THE POTENTIAL IMPACT OF FAST REACTORS AND FUEL RECYCLING SCHEMES ON THE UK'S NUCLEAR WASTE INVENTORY

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Abstract

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This work considers the impact of fast reactor fuel cycles on the UK's nuclear waste inventory, focusing on the disposition of the UK's plutonium stockpile and spent fuel from new build nuclear reactors.

Reprocessing spent fuel from nuclear reactors has led to a large stockpile of civil plutonium in the UK. At the end of reprocessing the stockpile was estimated to be 112 tonnes. This large stockpile of separated plutonium poses a proliferation concern and there is no strategy at present for UK plutonium disposition. The NDA's position paper in 2014 [1] stated the re-use of plutonium in a reactor as a preferred option. These options included Mixed OXide (MOX) fuelled Pressurised Water Reactors (PWR) and the use of plutonium in a Sodium-cooled Fast Reactor (SFR), PRISM, operated as a once-through plutonium burning fast reactor [1, 2]. As yet a preferred option has not been selected by the government.

Nuclear power is the UK's largest source of low-carbon electricity. Current plans aim to build 16 GWe of new reactors by 2050 [3] to replace the UK's current fleet.

This work considered PWR MOX and once-through SFRs for UK plutonium disposition, comparing their relative merits to the direct disposal of the plutonium stockpile in a geological repository. The waste performance of disposition options were compared using assessment criteria based on: Technology Readiness Level (TRL), final stockpile mass, repository size and radiotoxicity. To maximise the reduction of the UK's plutonium stockpile, closed SFR fuel cycles were also considered with scenarios aimed at improving waste performance. Once-through and closed SFR fuel cycles were also considered for the disposition of spent fuel from new build reactors.

Research presented in this thesis shows that UK waste disposition options are highly dependent on fuel cycle operating parameters. In once-through plutonium disposition options all scenarios increased repository size compared to direct disposal. Once-though SFRs increased repository size the least, where as PWR MOX reduced the stockpile mass most significantly. The most significant improvement in waste performance, using a closed fuel cycle up to 2150, required short reprocessing times and americium reprocessing. There were no additional improvements of significance with curium reprocessing and the choice of metallic or MOX fuelled SFRs had little impact on waste performance.

Preferred fuel cycle scenarios are dependent on the priority given to different assessment criteria. To compare fuel cycle scenarios on an even basis, decision analysis methods were presented using assessment criteria results from the fuel cycles modelled in this work. Decision analysis methods were designed so that the reader can apply their own priorities, through the use of weightings, to the assessment criteria to determine preferable fuel cycle scenarios.

Declaration

No portion of the work referred to in the thesis has been submitted in support of an application for another degree or qualification of this or any other university or other institute of learning.

Copyright Statement

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Definitions and Glossary

Definitions

Important terms used in this work.

1000 year radiotoxicity	Total radiotoxicity 1000 years after disposal in a repository.
2-tier fuel cycle	Recycling spent fuel through a thermal reactor then a fast reactor.
AM scenarios	Fuel cycle scenario recycling plutonium and americium.
Americium in-growth	The growth of ^{241}Am in a plutonium stockpile as a result fo the decay of ^{241}Pu.
Assessment criteria	A set of criteria used to assess the relative performance of a fuel cycle.
Breeding	A net production in fissile material during the operation of a reactor.
Burden on a repository	Having a large repository and long radiotoxicity lifetime, requiring long stewardship.
BURNER scenarios	Fuel cycle scenario using once-through burners and no reprocessing of spent fuel.
Burning	A net reduction in fissile material during the operation of a reactor.
Conversion Ratio (CR)	A measure of the ratio of fissile material produced to fissile material destroyed.
Cumulative decay heat	Estimation method for repository size.
Degradation	Reducing the fissile content or a material stream.

Direct Disposal	Conditioning material and sending material directly to a repository.
Disturbed repository	Where a repository would be compromised by a geological event, or human intrusion.
Enrichment	The weight percent of transuranics relative to total heavy metal.
Equilibrium	Recycling fuel through a reactor for enough cycles until the fuel feed and k_{eff} reach an equilibrium point.
Fast neutrons	Neutrons at energies close to the energy o neutrons produced from fission, ∼1–20 MeV.
FEED equilibrium	Recycling fuel through a reactor, replacing lost fissile material with fresh fuel from an external feed until an equilibrium is reached.
FEED scenarios	Fuel cycle scenario recycling fuel through a reactor, replacing lost fissile material with fresh fuel from an external feed.
Fertile	Can captures a neutrons to become fissile.
Fissile	Fissions readily at a range of neutron energies.
Fuel feed	Fissile feed of material used in a fuel cycle to fuel reactors.
FULL scenarios	Fuel cycle scenario recycling fuel through a reactor, reducing the number of reactors in each generation to maximise the reduction of stockpiled material.
FULL scenarios	Recycling fuel through a reactor using only reprocessed spent fuel as a fuel feed until an equilibrium is reached.
Hard spectrum	A neutron spectrum that is pushed towards fast energies.
Heavy Metal	There is no widely agreed definition of a heavy metal, but in this thesis it refers to uranium or heavier elements.
HI scenarios	Fuel cycle scenario using long reprocessing cooling times and high reprocessing losses.

Lifetime of radiotoxicity	The time it takes material to decay to radiotoxicities less than natural uranium.
LO scenarios	Fuel cycle scenario using short reprocessing cooling times and low reprocessing losses.
LWR offset	The ability of SFRs to generate electricity and offset the number of LWRs needed to reach nuclear power targets.
Metallic fuel (ZR)	U-TRU-10Zr fuel with a peak enrichment of 30 wt.%.
Moderator	A material used to effectively slow down neutrons to thermal energies.
MOX scenarios	Fuel cycle scenario using MOX fuelled reactors.
New build	New thermal reactors built in the UK.
One-through burner	Irradiating stockpiled material in a reactor once, without reprocessing.
Proliferation	The spread of weapons usable material or technology.
PU scenarios	Fuel cycle scenario recycling plutonium.
Radiotoxicity	Total dose as a result of ingestion, Sv.
Reactor park	A fleet of reactors, grouped together in a park based on their use or geographical considerations.
Repository	A geological excavation used to dispose of nuclear waste, isolating it for several hundred thousand years.
Reprocessing	Processing spent fuel in order to re-use the useful components in a reactor and dispose of the waste components.
Soft spectrum	A neutron spectrum that is pushed towards thermal energies.
Thermal neutrons	Neutrons at low energies, in thermal equilibrium with their environment, $\sim$0.025 eV.
Transmutation	The process of changing an isotope into a different isotope through fission or capture.
TRU scenarios	Fuel cycle scenario recycling all transuranics.
Waste performance	Measured by the performance of the following assessment criteria: repository size, 1000 year radiotoxicity, radiotoxicity lifetime and final transuranic inventory.

WASTE results	Reprocessing waste stream only results from FULL scenarios.
ZR scenarios	Fuel cycle scenario using metallic fuelled reactors.

Glossary

ABR	Advanced Burner Reactor
ADS	Accelerator Driven Sub-critical
AGR	Advanced Gas-cooled Reactor
ANL	Argonne National Laboratory
ASME	American Society of Mechanical Engineers
ASTRID	Advanced Sodium Technological Reactor for Industrial Demonstration
BISTRO	Optimized Two-Dimensional Sn Transport code
BOC	Beginning Of Cycle
BOEC	Beginning Of Equilibrium Cycle
BOL	Beginning Of Life
BU	Burnup
BWR	Boiling Water Reactor
CDH	Cumulative Decay Heat
CR	Conversion Ratio
DDR	Dimitrovgrad Dry Route
DECC	Department of Energy and Climate Change
DFR	Dounreay Fast Reactor
DIFF	Diffusion
EBR-I	Experimental Breeder Reactor I
EBR-II	Experimental Breeder Reactor II
ECCO	European Cell COde
EFPD	Effective Full Power Days
EFR	European Fast Reactor
ENDF	Evaluated Nuclear Data File
EOC	End Of Cycle

EPR	European Pressurized Reactor
ERANOS	European Reactor ANalysis Optimized calculation System
ESFR	European Sodium-cooled Fast Reactor
FBR	Fast Breeder Reactor
FBTR	Fast Breeder Test Reactor
FC	Fuel Cycle
FCCI	Fuel Cladding Chemical Interaction
FCMI	Fuel Cladding Mechanical Interaction
FFTF	Fast Flux Test Facility
FOI	Freedom Of Information
FP	Fission Product
GCR	Gas Cooled Reactor
GFR	Gas-cooled Fast Reactor
GHG	Green House Gas
GIF	Generation IV International Forum
GNEP	Global Nuclear Energy Partnership
GWd/t	Gigawatt days per tonne
GWe	Gigawatt electric
GWt	Gigawatt thermal
GWy(e)	Gigawatt years electric
HET	Heterogeneous
HEX	Hexagonal
HLW	High Level Waste
HM	Heavy Metal
HOM	Homogeneous
HTR	High Temperature Reactor
HWR	Heavy Water Reactor
IAEA	International Atomic Energy Agency
IFR	Integral Fast Reactor
ILW	Intermediate Level Waste
IMF	Inert Matrix Fuel
INL	Idaho National Laboratory

JAEA	Japan Atomic Energy Agency
LFR	Lead-cooled Fast Reactor
LWR	Light Water Reactor
MA	Minor Actinide
MOX	Mixed OXide
MSR	Molten Salt Reactor
MWe	Megawatt electric
MWt	Megawatt thermal
NDA	Nuclear Decommissioning Authority
NEA	Nuclear Energy Agency
NIREX	Nuclear Industry Radioactive waste EXecutive
NNL	National Nuclear Laboratory
ODS	Oxide Dispersion Strengthened
OECD	Organisation for Economic Co-operation and Development
ONR	Office for Nuclear Regulation
pcm	per cent mille (one-thousandth of a percent)
PFBR	Prototype Fast Breeder Reactor
PFR	Prototype Fast Reactor
PONI	Project On Nuclear Issues
PP	Physical Protection
PR	Proliferation Resistance
PRIS	Power Reactor Information System
PRISM	Power Reactor Innovative Small Modulus
PUREX	Plutonium Uranium Redox EXtraction
PWR	Pressurised Water Reactor
R&D	Research and Development
RWMD	Radioactive Waste Management Directorate
SFR	Sodium-cooled Fast Reactor
SMR	Small Modular Reactor
tHM	tonnes Heavy Metal
THORP	Thermal Oxide Reprocessing Plant
TR	Thermal Reactor

TRL	Technology Readiness Level
TRU	Transuranic
UK	United Kingdom
UOX	Uranium OXide
XS	Cross-section

Chapter 1

Introduction

The United Kingdom (UK) is planning an ambitious nuclear new build programme, currently aiming to build 16 GWe of new nuclear reactors by 2050 [3]. Simultaneously, the UK is deciding how to dispose of its large civil plutonium stockpile. There are many long-term issues that the nuclear industry needs to address, particularly waste. The preferred option for disposing of nuclear waste is in a geological repository which must isolate nuclear waste for several hundred thousand years. The UK has used public consultation to locate a site for a geological repository but has yet to find a site.

Several options have been suggested for the disposition of the UK's plutonium stockpile. Preferred options are re-use strategies, three of which are being considered: re-use in a thermal Light Water Reactor (LWR); re-use in a Heavy Water Reactor (HWR); and re-use in a Sodium-cooled Fast Reactor (SFR) [1]. Alternatively there is the direct disposal approach, that is, disposing of the UK's plutonium stockpile in a geological repository. Currently a decision has not been made on UK plutonium disposition. At present, the UK does not have a site for a geological repository and the main barrier to siting a repository is public perception. Eurobarometer polls show that the three key areas of public concern with regard to the nuclear industry are terrorism, nuclear waste, and proliferation [4].

This PhD looks at how fast reactor fuel cycles could address these areas of concern. Fast reactor fuel cycles were modelled using UK stockpiled material. LWR and SFR options for UK plutonium disposition were assessed to determine the relative merits of plutonium disposition options. Plutonium disposition aims to reduce the proliferation

concerns surrounding large stockpiles of separated plutonium. SFR fuel cycle models were extended to include closed fuel cycles, with an aim to reduce the burden the UK's plutonium stockpile would have on a repository. Reducing the burden on a repository would involve reducing repository size and the lifetime of waste with an aim to improving public perception of nuclear waste. SFR fuel cycle models then incorporated new build reactor Spent Nuclear Fuel (SNF), with an aim to reduce the burden that new build SNF would have on a repository. SFRs and their fuel cycle were assessed in terms of their Technology Readiness Level (TRL) and examples of decision analysis were used to show how the results of this study could be used to determine fuel cycles for further study, based on the goals and importance given to the criteria used to assess each fuel cycle scenario.

1.1 Key concepts

This section will introduce key concepts that will be used throughout this thesis. Definitions of common terms used can be found in the Definitions and Glossary at the beginning of this thesis.

Fuel cycle scenario A model of a potential nuclear fuel cycle accounting for reactor operation, facilities to support reactor operation, natural resources required, and the ultimate disposal of waste. Scenarios are developed to meet specific aims such as security of energy supply or reducing the generation of nuclear waste.

Fuel feed Feed of material used in a fuel cycle to fuel reactors.

Technology Readiness Level (TRL) Provides a degree of standardisation for comparing the development stage of different technologies. The TRL scale used has 10 levels. As the technology matures from level 1 to level 10, it moves from a scientific idea to a commercially deployed concept. TRL definitions used in this thesis are described in Table 2.1.

Cross-sections Nuclear cross-sections are used to describe the probability of a neutron under-going a reaction with a particular nuclei. Cross-sections are energy dependent and typically higher at thermal energies and lower at fast energies. Cross-sections represent absorption reactions (most commonly fission, capture and (n,2n) reactions) and scattering reactions (elastic and inelastic scattering).

Conversion Ratio (CR) Also called the Breeding Ratio (BR), CR is the ratio of fissile material produced to fissile material destroyed in a reactor,

$$\mathrm{CR} = \frac{\text{Fissile material produced}}{\text{Fissile material destroyed}} = \frac{\text{Rate of capture}}{\text{Rate of destruction}}. \tag{1.1}$$

Breeding The process of generating more fissile material from fertile material. Fissile material readily undergoes fission, such as ^{239}Pu. In a uranium-plutonium fuel cycle, breeding is the process of fertile ^{238}U capturing an neutron to produce fissile ^{239}Pu,

$$^{238}\mathrm{U} +^{1} \mathrm{n} \xrightarrow{\text{capture}} ^{239}\mathrm{U} \xrightarrow{\beta\text{-decay}} ^{239}\mathrm{Np} \xrightarrow{\beta\text{-decay}} ^{239}\mathrm{Pu}. \tag{1.2}$$

Breeder reactors have a CR > 1, where they breed more fissile material than they fission.

Burning The process of irradiating fuel in a reactor to reduce the fissile inventory of the fuel. Burner reactors have a CR < 1.

Transmutation The process of changing an isotope into a different isotope through fission or capture. Transmutation scenarios aim to reduce mass of Minor Actinides (MAs) in the fuel cycle, which are the main long-lived component of nuclear waste, by recycling them through fast reactors. MAs are neptunium (Np), americium (Am) and curium (Cm).

Waste performance Waste performance in this thesis is defined by the following fuel cycle assessment criteria: Repository size, radiotoxicity lifetime, radiotoxicity 1000 years after repository emplacement, and final inventory of transuranics sent to a repository

Table 1.1: World nuclear power reactors operating and under construction as of 2014 [5]. Pressurised Water Reactors (PWR), Boiling Water Reactors (BWR), Gas Cooled Reactors (GCR), Light Water moderated Gas cooled Reactors (LWGR), Heavy Water Reactors (HWR) and Fast Reactors (FR).

	Operating	**Construction**
PWR	279	56
BWR	78	4
GCR	15	1
LWGR	15	0
HWR	49	4
FR	2	2
Total	438	67

Radiotoxicity Described in this thesis as the total effective dose as a result of ingestion. The total effective dose is dependent on a radiation weighting factor and tissue weighting factor. More highly ionising radiation results in higher effective doses and the irradiation of more sensitive tissues results in a higher effective doses.

Repository Geological excavation used to dispose of nuclear waste, isolating it for several hundred thousand years.

1.2 Nuclear power's global outlook

At the end of 2014 there were 438 nuclear reactors operating which made up 10.2% of the world's electricity supply [5]. Two of these reactors were fast spectrum reactors and the rest were thermal-spectrum reactors, the majority of which were Pressurised Water Reactors (PWRs), Table 1.1.

Advantages of nuclear power Nuclear power is advantageous in terms of providing base-load power, low Green House Gas (GHG) emissions, and security of supply whilst having generation costs which are insensitive to fuel price [6]. With recent carbon emission targets and fluctuations in fossil fuel prices, there has been a resurgence of interest in nuclear power with 67 reactors currently under construction, and many new countries entering the nuclear market [5].

The cost of nuclear power is relatively insensitive to fluctuations in uranium price

[7]. The bulk of the cost associated with nuclear power is in the construction of a plant. The energy density of natural uranium is more than 30,000 times that of coal. As such, there is a low dependence on foreign imports which is advantageous in terms of security of supply.

At present there is an abundance of uranium, current estimates show that there is enough uranium for more than 150 years of nuclear power, based on current nuclear growth estimates [8]. Nuclear power has life cycle GHG emissions in the same range as renewable energy sources [9]. Currently, nuclear power is the only source of low carbon electricity with the potential to meet base-load electricity demand.[1]

Disadvantages of nuclear power Despite an overall increase in nuclear power worldwide, there has been a down turn in nuclear power in Europe and North America. The down turn is a result of the high upfront cost of nuclear power plants and the long construction time before before the generation of revenue, and public perception with regards to safety, terrorism,[2] proliferation and the disposal of waste [6]. In addition, uranium is a finite resource, although there is enough uranium for an estimated 150 years of nuclear power, it is not sustainable in the long run.

Despite the cost per kWh of nuclear power being competitive with other electricity sources, the up front cost of building a nuclear power plant is high. This combined with long construction timescales and the potential for delays, means that there is a lot of uncertainty and risk in building nuclear power plants [6, 7]. This can make nuclear power unfavourable for companies operating in a privatised energy sector.

The large scale of accidents such as Fukushima and Chernobyl lead to sharp drops in public perception which recover slowly. There are three additional factors which prevent nuclear power from having wide spread public acceptance: First is the concern of terrorist attacks, second the lifetime and disposal concerns surrounding nuclear waste, and finally, the potential for nuclear materials to be used for weapons [4].

At present there is no long-term solution for nuclear waste in the UK. Current plans are to construct geological repositories. Repositories aim to isolate nuclear waste in the

[1] In the near-term fossil fuels with carbon capture storage may become available, and in the long-term nuclear fusion may become available.

[2] It is worth noting that fuel cycles which minimise the transport of nuclear materials reduce the opportunity for terrorists to attack transported material. However, facilities still exist containing nuclear material, meaning there are still targets for terrorism.

ground for several hundred thousand years to allow radioactive material to decay to safe levels. Under nominal operating conditions radioactive releases from a repository would be small, but given such a long timescale there is a degree of uncertainty. Uncertainty and concerns surround the potential for a disturbed repository as a result of geological factors or human intrusion. A disturbed repository could result in the repository becoming compromised and the release of large quantities of radioactive material into the environment.

Thermal reactor technology Nearly all reactors currently in operation are thermal reactors, of these most are LWRs [10]. In uranium fuelled thermal reactors most fission reactions occur in the fissile component of uranium, ^{235}U, which makes up 0.7% of natural uranium. Most countries treat the spent fuel from thermal reactors as a waste product to be disposed of in a repository. Thermal reactor technology will be discussed in more detail in Section 1.4.1.1.

Potential for fast reactors Initial interest in fast reactors was a result of their ability to breed fissile material from the fertile component of natural uranium, ^{238}U, which makes up 99.3% of natural uranium. At the time of fast reactor development a global uranium shortage was expected and fast reactors were developed with the aim of extending the lifetime of natural uranium resources. However, uranium was more abundant than first thought, and the growth of nuclear power was much slower [11]. As a result, costly development of new fast reactor technology slowed in favour of thermal reactors which were already commercially available and cost competitive.

Resurgence in fast reactor research has been primarily a result of new nuclear countries that do not have natural uranium resources. Fast reactors are seen as a way to ensure security of supply, removing the dependence on uranium imports. In addition, interest in fast reactor research has come from the ability of fast reactors to transmute long-lived components of nuclear waste. Transmutation of long-lived isotopes would reduce the burden on a repository in terms of repository size and the lifetime of radiotoxicity. Reducing the burden on a repository would help address public concerns surrounding nuclear waste.

Despite the advantages of fast reactors they are not widely used, Table 1.1, as they are at a low stage of commercial development. Industrial deployment would be costly

and require fundamental changes to the nuclear fuel cycle. This makes fast reactors less economically competitive than thermal reactors and other electricity generation methods in the near-term.

1.3 Nuclear power's outlook in the UK

At the end of 2014 there were 16 nuclear reactors operating in the UK which supplied 17.2% of the UK's electricity supply, Table 1.2. Most of these reactors were Advanced Gas-cooler Reactors (AGRs) [5].

Table 1.2: Nuclear power reactors in the UK that are operating and permanently shutdown as of 2014 [5].

	Operating	**Shut-down**
PWR	1	-
Magnox	1	25
AGR	17	1
HWR	-	1
FR	-	2
Total	16	29

The UK has previously operated 29 nuclear power reactors which are now permanently shut-down. The majority of these are gas-cooled and graphite moderated Magnox reactors, which were the first generation of electricity generating reactors in the UK. At the time of Magnox reactor construction there was a surge in nuclear power interest and estimates of natural uranium resources were low. As a result, the UK began developing fast reactors with an aim to transition from thermal reactors to fast reactors [11, 12]. Mangox spent fuel was reprocessed, extracting the reusable plutonium and uranium from spent fuel. The separated plutonium was stockpiled in preparation for use in fast reactors.

As previously discussed, natural uranium turned out to be far more abundant than first thought and the uptake of nuclear power slowed down. This led to the end of the fast reactor programme in the UK. However, reprocessing of spent fuel continued, now extended to include spent fuel from the second generation of gas-cooled, graphite moderated reactors, AGRs, and contracts to reprocess foreign reactor fuel [12].

Present situation Reprocessing in the UK has led to a large stockpile of civil plutonium, in 2013 there were 99.6 tonnes of UK owned plutonium and 23.4 tonnes of foreign owned plutonium [13]. Reprocessing is expected to end between 2018 and 2020 [14, 15], leaving the UK with an estimated plutonium stockpile of 140 tonnes [1]. This large stockpile of separated plutonium poses a proliferation concern and there is no strategy for use or disposal of the stockpile. In the past there have been three long-term considerations for managing the UK's plutonium stockpile: direct disposal in a repository, use in a PWR as MOX before disposal, and long-term storage [16]. In 2011 the UK government opened a consultation on the UK's plutonium stockpile [17]. In response to this consultation, two extra disposition strategies were suggested. The use of plutonium in an HWR (CANDU), and the use of plutonium in an SFR (PRISM) operated as a once-through, metallic fuelled, plutonium burning fast reactor [1, 2]. In an NDA position paper in 2014 [1] the re-use of plutonium in a reactor was suggested as being preferable as it reduces the attractiveness of the stockpile, putting it out of reach in a highly active matrix before disposal in a repository. As yet a preferred option has not been selected by the government.

Nuclear power in the UK is the largest source of low-carbon electricity with a total capacity of 9.37 GWe in 2014. However, most nuclear power stations are due to shut by 2030 [18, 19, 20]. Current plans are to build new PWRs and BWRs (jointly refereed to as LWRs) to replace the current fleet, with a goal of 16 GWe of new build reactors by 2050 [3].

The UK has also released a Nuclear Energy R&D Roadmap, see Figure 1.3 [21]. Whilst the Nuclear Energy R&D Roadmap does not outline the future of the UK nuclear fuel cycle, it does outline potential research pathways based on different nuclear growth scenarios, including the operation of 16 to 75 GWe of new nuclear reactors by 2050. These fuel cycles will be discussed in more detail in Section 3.6.

1.4 Background

1.4.1 Nuclear reactors

Nuclear reactors generate heat which is used to produce steam to drive a turbine and produce electricity, Figure 1.1. The process of fission produces a lot of heat through the

splitting of a nucleus, with most energy coming from recoiling fission fragment which heat up the fuel. Fuel is wrapped in a cladding material and bundled into assemblies for structural integrity and refuelling purposes. Surrounding the fuel assemblies is a coolant which is used to transfer heat to a steam generator [10].

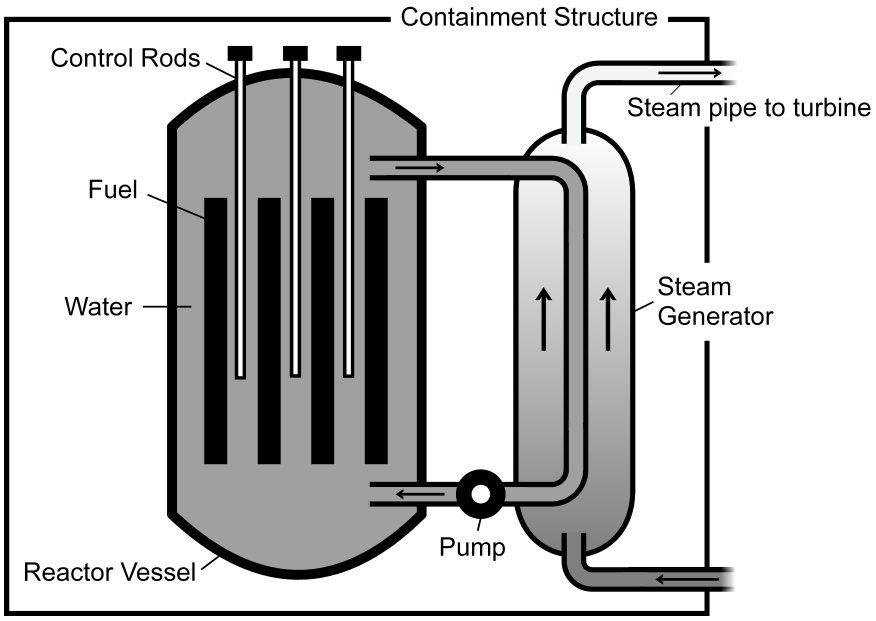


Figure 1.1: Diagram of a PWR, drawn by present author for Ref. [10].

Fission is the process of a heavy nuclide absorbing a neutron causing the heavy nuclide to split, producing fission fragments and neutrons. If one of the neutrons goes on to cause another fission, then a chain reaction is sustained. The probability of fission occurring is heavily dependent on the energy of the neutron. Nuclides where fission readily occurs at a variety of neutron energies are described as fissile [10].

Natural uranium's major constituents are fissile ^{235}U and fertile ^{238}U. Fertile means that the nuclide can capture a neutron to produce a fissile nuclide, for example ^{238}U captures a neutron and decays to ^{239}Pu which is fissile, equation 1.1.

The effective neutron multiplication factor, k_{eff} is used to describe the criticality condition of a reactor. A reactor is critical if $k_{eff} = 1$, where the neutron population in a reactor is maintained, a reactor is sub-critical if $k_{eff} < 1$, where not enough neutrons are produced to maintain the fission chain reaction, and a reactor is super-critical if $k_{eff} > 1$, where the neutron population rapidly increases. Operating reactors need a $k_{eff} = 1$, with intrinsic feedback loops in the design of the reactor to counteract small fluctuations in k_{eff}. k_{eff} is controlled by the addition or removal of neutron poisons,

which have a high neutron capture cross section, in the reactor. Control rods, which are made of neutron poisons, are inserted to shut-down a reactor making it sub-critical.

Neutrons produced from fission have high energies, at high neutron energies fission cross-sections are low, whereas at thermal neutron energies fission cross-sections are greater. Thermal reactors use a moderating material to slow neutrons to thermal energies so that fission reactions are more probable. Ideal moderating materials have a low atomic mass, high scattering cross-section and low absorption cross-sections to ensure that neutrons are slowed quickly and with minimal parasitic absorption. Fast reactors on the other hand do not use moderators keeping neutrons at high energies. Fast reactors therefore need a higher proportion of fissile material in the fuel and all other materials need to be minimised to reduce the amount of parasitic absorption and scattering from structural and coolant materials.

1.4.1.1 Thermal reactor technology

A diagram of a PWR is shown in Figure 1.1. In a PWR water is used as a moderator and coolant. Thermal reactors, such as PWRs, optimise the spacing of fuel pins to allow for the optimum fuel to moderator ratio. This is the point where the probability of fission is maximised whilst minimising the amount of absorption in structural, coolant and moderator materials. Having the optimum fuel to moderator ratio means that there is a reactivity feedback loop. If k_{eff} becomes greater than one there is an increase in temperature which causes the moderator to expand, moving away from the optimum fuel to moderator ratio, reducing the amount of moderation and reducing k_{eff}.

Most thermal reactors use UO_2 ceramic fuel which is chemically inert and has a high melting temperature which is advantageous in accident conditions. To sustain a fission chain reaction in most thermal reactors, the fissile component, ^{235}U, must be enriched above natural levels of $\sim$0.7% to >3%, to account for the loss of neutrons due to parasitic absorption. Fuel is also enriched to increase the irradiation time between refuelling which reduces the time spent refuelling and increases the capacity factor.

When the fissile component of fuel is depleted to the point where a chain reaction can no longer be sustained the fuel is replaced. The fuel burnup is measured as either the atomic percentage of heavy metal isotopes fissioned, at. %, or as the thermal

power generated per unit mass, GWd/t.

Over the in-core lifetime of fuel there is a build-up of fission products. Heavier isotopes than uranium, transuranics (TRU), also build-up due to successive captures in uranium and transuranics. The ratio of fission to capture cross-section of TRUs at thermal energies is such that heavier transuranic isotopes are generated with no net reduction if are recycled through thermal reactors.

1.4.1.2 Fast reactor technology

Fast reactors are different to thermal reactors in a number of ways. Fast reactors aim to keep the neutron energy spectrum close to the fission neutron energy spectrum. It is preferable to have a small core with low structure and coolant volume to minimise the presence of scattering isotopes which can soften[3] the neutron spectrum. The fuel to coolant ratio is limited by the ability of the coolant to remove heat from the fuel. At fast energies, cross-sections are around 100 times lower than thermal cross-sections. As such, higher fissile content fuel is needed in fast reactors to maintain the fission chain reaction and the power density is much higher than in a thermal reactor [22]. Liquid metals have been used as coolants as the nuclei have a minimal scattering effect and the high thermal capacity of a metallic coolant is effective at removing heat given a high power density and low coolant volume.

As a result of the low cross-sections at fast energies compared to thermal energies, the neutron diffusion length in longer due to the lower probability of interactions. Longer diffusion lengths results in more neutrons being leaked from the core in fast reactors, $> 20\%$ compared with $\sim 3\%$ in LWRs. With high leakage, reflector materials are used to surround the core and improve the neutron economy. Neutron diffusion length is 10-20 cm in fast reactors, the order of a fuel assembly, compared to 2 cm in an LWR, the order of a fuel pin. As such, LWRs have a significant flux depression across fuel pins, which is not as significant in a fast reactor.

In fast reactors more neutrons per absorption are produced than in thermal reactors. One neutron is needed to cause another fission, the rest are either parasitically absorbed (coolant, structural or fuel material) or leaked from the core. More neutrons per fission combined with lower cross-sections for parasitic absorption results in fast

[3]Push the neutron spectrum towards thermal energies.

reactors having extra neutrons that can be absorbed in fertile isotopes to breed fissile material, equation 1.1. As leakage is a large contributor to the loss of neutrons, putting a dense fertile material around the core can utilise the leaked neutrons to breed fissile material.

For TRUs the fission to capture ratio is higher at fast energies than thermal energies. At fast energies, the number of neutrons produced per absorption is greater than one. As such, all TRUs positively contribution to the neutron balance of a fast reactor. In a thermal reactor only fissile TRUs contribute positively to neutron balance. As a result, recycling TRUs in fast reactors can have a net reduction in higher TRUs as a result of successive captures and fissions, this process is called transmutation. Recycling TRUs in a thermal spectrum will degrade the isotopic vector and build-up higher actinides with each recycle.

1.4.2 The nuclear fuel cycle

There are three types of fuel cycle: open, partially-closed and closed. These are shown in Figure 1.2. Fuel cycles start with the mining, milling and chemical leaching of natural uranium to form Uranium Ore Concentrate. Natural uranium contains ∼0.7% ^{235}U, however most thermal reactors require a greater proportion of ^{235}U. To enrich the concentration of ^{235}U it is converted into UF_6 and passed through centrifuges which preferentially separate the heavier and lighter uranium compounds. The enriched uranium is converted into a chemically stable powder, UO_2, which is pressed into pellets and sintered. These are stacked in fuel pins and bundled to make fuel assemblies which are loaded into a reactor. Heat is generated from the fission of ^{235}U, leaving behind fission products and activation products which are highly radioactive. Spent fuel is sent to cooling ponds to allow fission products to decay, which results in the decay of the total activity and decay heat generated by the fuel. The step after cooling depends on the fuel cycle:

- Open fuel cycle – Fuel is cooled for longer and conditioned for disposal in a repository.
- Partially-closed fuel cycle – Fuel is reprocessed, where chemical separations are used to remove the waste component for disposal and separate useful material

that can be recycled as fuel. The waste stream is immobilised for disposal. The reprocessed fuel material is fabricated into fresh fuel for re-use in a reactor. Once the new fuel has been irradiated it is stored, conditioned and disposed of in a repository.

- Closed fuel cycle – In a fully closed fuel cycle spent fuel is continually reprocessed and recycled in reactors. This is not possible using thermal reactors as the reprocessed fuel vector degrades with continued recycling to the point where it impedes thermal reactor operation. In fast reactors this is not the case and fully closed fuel cycles can be sustained.

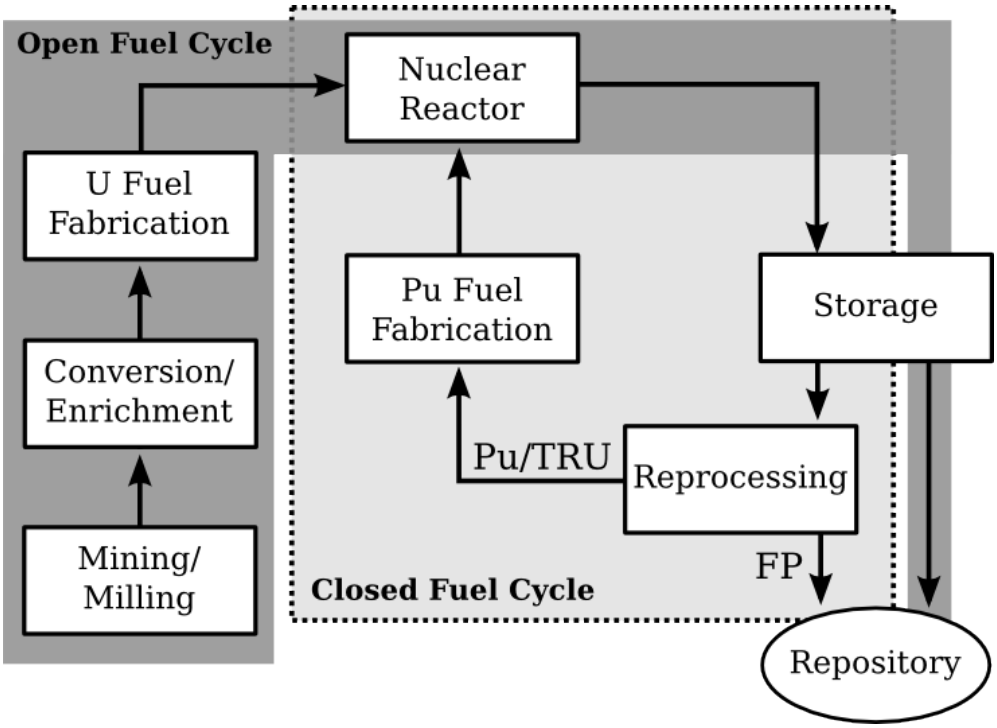


Figure 1.2: Diagram of the nuclear fuel cycle.

There are three main types of fast reactor closed fuel cycle:

- Breeding fuel cycles where fast reactors are operated with a CR > 1 to breed excess fissile material to start up new reactors.
- Steady state fuel cycles with CR $= 1$ where reactors are operating in equilibrium, producing the same amount of fissile material as they burn, preventing the build-up of fissile material whilst removing the dependence on natural uranium.
- Burner or transmutation fuel cycles with CR < 1 to reduce the quantity of TRUs in the fuel cycle, minimising the size of a repository and radiotoxicity lifetime.

A repository is designed to isolate nuclear waste until it has decayed to safe levels, typically several hundred thousand years. The size of a repository is determined by the decay heat generated by waste, with High Level Waste (HLW) canisters spaced out to ensure that peak temperature limits are not exceeded. Other factors also contribute to repository size such as criticality concerns. High plutonium content waste will have to be spaced out so that criticality cannot occur. Intermediate level waste (ILW) which does not generate heat is also disposed of in a repository, but takes up a much smaller area as there are no thermal limitations.

The total radiotoxicity of material in a repository is the total ingested dose. Long-lived contributors to radiotoxicity are not mobile in repository condition, as such, total releases to the biosphere from a repository will be low under nominal operating conditions. However, over the hundred thousand year timescale a repository could be disturbed by geological events or human intrusion. A disturbed repository could result in large radioactive releases to the biosphere.

1.5 Aims of this study

The UK has the largest civil plutonium stockpile in the world with no current disposition route in place. In addition, the UK is planning to build 16 GWe of new nuclear reactors. There have been several studies as part of the UK's nuclear R&D roadmap which investigate large scale build-up of nuclear power in the UK and transitioning to fast reactors to reduce the dependence on natural uranium.

The NDA's preferred plutonium disposition routes are once-through use of plutonium in a reactor. A PWR, HWR and SFR have been outlined as credible options but a decision has not been made on a final disposition route, and the relative merits of these options have not been discussed in the open literature. The key concerns which influence public perception of nuclear power in Eurobarometer polls are terrorism, waste and proliferation [4]. Terrorism concerns cannot easily be addressed by fuel cycle design. Proliferation concerns can be reduced by burning the UK's plutonium stockpile or immobilising the plutonium and putting it out of reach in a repository. Waste concerns could be reduced with transmutation fuel cycles.

The aim of this work is to investigate fast reactor fuel cycles in a UK context,

considering the UK's plutonium stockpile as well as new build LWRs and the potential impact of fast reactors on final waste inventories. This will be considered over several timescales and fuel cycle options will be assessed against a range of assessment criteria. Specifically this work aims to:

- Assess the TRL of fast reactor and fuel cycle concepts;
- Down select fast reactor fuel cycle options suitable for the UK;
- Model the build-up of the UK's plutonium stockpile, and SNF from new build LWRs, for use in reactor and fuel cycle modelling;
- Neutronics design of fast reactors for use in UK fuel cycles, fuelled with UK stockpiled materials;
- Model once-through UK plutonium disposition fuel cycle options to assess their relative merits;
- Model closed fast reactor fuel cycle scenarios using the UK's plutonium stockpile to assess their impact on waste performance;
- Model fast reactor fuel cycle options using the UK's plutonium stockpile and new build LWRs to assess their impact on waste performance;
- Assess the performance of fuel cycle scenarios against direct disposal scenarios and once-through LWR scenarios, showing the relative improvement factors and TRL of each fuel cycle;
- Present all results as numerical values that can be normalised and weighted for use in decision analysis to determine the best fuel cycles for further study.

A review of fast reactor and fuel cycle options is required to select options for a fast reactor fuel cycle in the UK. The TRL of these fuel cycles and fast reactor concepts will be assessed and used to compare fuel cycle scenarios in terms of their development needs.

There is little publicly available information on the UK's civil plutonium stockpile. As such, plutonium stockpile build-up must be modelled from the operating histories of reactors and reprocessing plants. The aim is to make an appropriate model of the UK's plutonium stockpile that can be used as the basis for fuel cycle and neutronics models.

To accurately model the impact fast reactors have on UK nuclear materials, reactor neutronics models are required. The aim is to design fast reactors that are optimised for the purpose of the fuel cycle scenario. Results from the neutronics models can be used in the fuel cycle models to ensure that the reactor component is accurately represented. Reactor models must be developed for each fuel cycle scenario, accounting for different fuel cycle parameters.

Fuel cycle models require a fuel feed (the UK's plutonium stockpile and, where appropriate, spent fuel from new build LWRs) and a reactor based on neutronics models. Each fuel cycle scenario has a range of parameters that can be varied to determine how this influences the performance of a fuel cycle.

Once-through SFR fuel cycles for plutonium disposition and their relative performance to PWR MOX is not presented in the open literature. This will be discussed as it pertains to UK plutonium disposition. Closed SFR fuel cycles for stockpile reduction have been under represented in the literature and use very low TRL fuel cycles. Low and high TRL fast reactor fuel cycles will be compared in a UK context to see how stockpile reduction is influenced.

Assessment criteria for a fuel cycle will be: TRL, electricity generated, fuel cycle lifetime, final TRU inventory, repository size, radiotoxicity lifetime, radiotoxicity in a disturbed repository scenario, and the bare sphere critical mass of the final waste stream. The term waste performance is defined in this thesis as the measure of the certain assessment criteria: repository size, 1000 year radiotoxicity, radiotoxicity lifetime and final transuranic inventory. Results will be given as the relative improvement when compared to reference scenario such as PWR MOX, direct disposal, or a once-through LWR scenario, as appropriate.

Final results for each assessment criterion will be represented as improvement factors over a reference case. Improvement factors can be normalised and weighted based on the relative importance of each assessment criteria. The aim is to show how the results from this thesis can be used in decision analysis to determine the best scenarios for further study and which type of scenarios are preferable based on different drivers. The author understands that the importance given to each of the assessment criteria can vary with personal opinion, but examples will be given.

1.5.1 Limitations

There are several key limitations of this study. It is important to outline the main limitations of this study, they will be summarised here and discussed in more detail in later chapters.

TRL is a broad way of determining the development needs of a technology. Some technologies with the same TRL will have very different development timescales based on experience with related technology.

There were many assumptions and limitations in estimating the UK's plutonium stockpile. Most significantly, average Magnox and AGR burnups were used, where as burnup changes over the operating lifetime of reactors. Estimating new build also had limitations, the average burnup, build rate, and final capacity of reactors operated are all subject to change so an approximation was used.

JEF-2.2 was the only cross-section library available in ERANOS. Cross-sections are the fundamental limitation of neutronics modelling. More up to date cross-sections would be preferable but were not available.

Assessment criteria used to determine the performance of fuel cycle scenarios do not cover all aspect of fuel cycle performance. The assessment criteria do not consider cost, proliferation resistance or the impact on fuel cycle facilities such as shielding requirements.

The cumulative decay heat method for estimating repository size was not accurate but adequate for indicating relative repository size. Design of a repository was not known, making it difficult to model or estimate repository size. Repository size in the fuel cycle models assumed all waste was sent to a repository at the end of the fuel cycle. Waste was not sent to a repository during a fuel cycle scenario.

1.5.2 Outline of chapters

Chapter 2 is a TRL assessment of SFRs and SFR fuel cycles. Chapter 3 reviews literature on previous fast reactor fuel cycle studies to determine gaps in the field. Chapter 4 outlines the fuel cycle scenarios studied in this thesis and the methods used to study them. Chapter 5 estimates the UK's plutonium stockpile and spent fuel from the UK's new build programme. These were used as fuel feeds for reactor

design and fuel cycle modelling. Chapter 6 presents results for SFRs designed for the fuel cycle scenarios. Chapter 7 presents results for once-through SFR burner fuel cycles and PWR MOX as plutonium disposition options. Chapter 8 presents results for closed SFR fuel cycles for UK plutonium disposition, aiming to reduce the burden on a repository. Chapter 9 presents results for once-through and closed SFR fuel cycles for the disposition of the UK's plutonium and spent fuel from new build reactors. Chapter 10 outlines a methodology for using the results in this thesis thesis in decision analysis. Finally Chapter 11 summarises this work, drawing conclusions and outlining the limitations as well potential areas for further work.

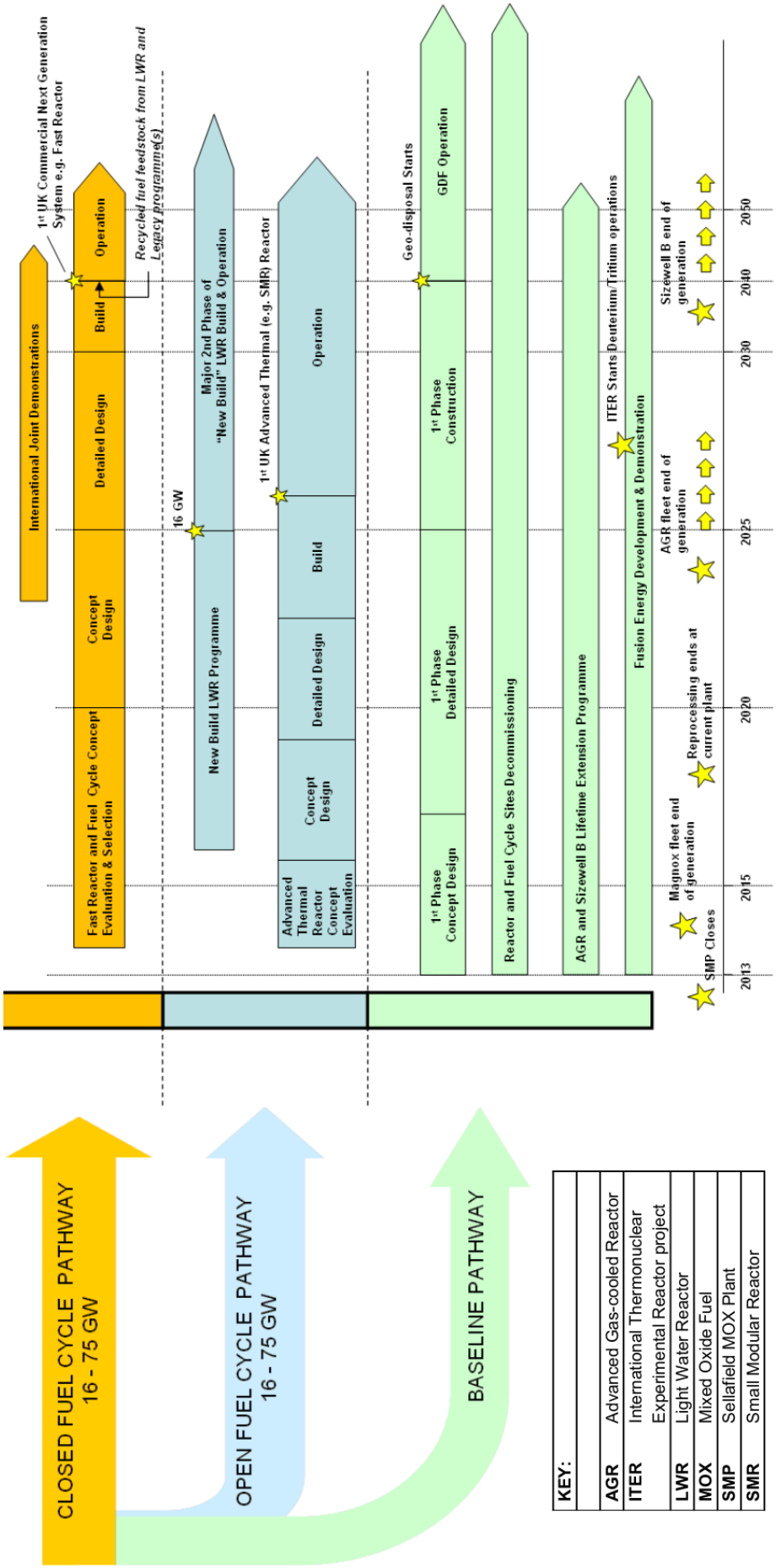


Figure 1.3: Nuclear Energy Technology Pathway Options. Taken from Ref. [21].

Chapter 2

TRL assessment of SFR fuel cycles

2.1 Introduction

There are many fast reactor concepts and fuel cycles being studied internationally. More advanced concepts can have advantages over current technology, however the development required could make advanced concepts undesirable.

It is important to consider the TRL of fast reactors concepts, fuel cycle concepts and the fuel cycle as a whole to understand which types of fuel cycle are deployable in the near-term, and which fuel cycles will need considerable investment and development. Definitions of the TRL scale used in this work can be found in Table 2.1.

TRL assessment of fast reactors and fuel cycle facilities was based on the present author's judgement of information available in the literature. The TRL assessment forms a few parts:

- Fast reactor TRL assessment;
- Selection of fuel and fast reactor parameters;
- Fuel cycle facilities TRL assessment, focussing fuel fabrication and reprocessing.
- Overall TRL assessment of closed SFR fuel cycles.

It is worth noting that other factors are relevant to development as well as TRL, including cost, technical expertise, and time-scales for development. These additional factors were considered outside the scope of this work and would require large assumptions, rendering any assessment inaccurate.

Only fast reactors concepts with solid fuel were considered. Sections on fast reactor TRL assessment have been taken from work done by the present author during a secondment, writing technical reports for DECC as part of a joint contract between The University of Manchester and the National Nuclear Laboratory [23,24,25]. The present author was responsible for chapters on Sodium-cooled Fast Reactors (SFRs), Lead-cooled Fast Reactors (LFRs) and Gas-cooled Fast Reactors (GFRs). TRL assessment for LFRs and GFRs have been removed, with a summary of results in Section 2.2.

For the purpose of brevity, aspects not directly applicable to the neutronics modelling of SFRs or fuel cycle modelling have been moved to the Appendix, these include: out-of-core reactor components (Appendix A), review of fabrication and reprocessing methodologies as well technical issues associated with fabrication and reprocessing (Appendix B).

A summary of the discussions in this chapter have been tabulated for ease of reference, without the need to read the accompanying justification. The SFR TRL summary can be found in Table 2.5, the reprocessing TRL summary can be found in Table 2.6 and the fuel fabrication TRL summary can be found in Table 2.7.

2.2 Down selection of a fast reactor coolant

Table 2.2 shows the TRL for reference, near-term demonstration fast reactors. These were assessed as part of previous work by the present author [23]. SFRs have a much higher TRL than LFRs or GFRs. Even though GFRs and LFRs have advantages, such as coolant compatibility with water and air [22], or a neutron energy spectrum the favours breeding or transmutation [26]. Any advantages of an LFR or GFR are insignificant compared to the development requirements to build a basic commercial demonstrator. Equally, in terms of the neutronic performance of the reactor, the selection of metallic or oxide fuel will have more of a neutronics impact in terms of breeding and transmutation than the choice of coolant [26]. The impact of fuels is discussed further in Section 2.4. As SFRs have the highest TRL of all fast reactor concepts, they were the only reactor concept considered in this study. Detailed discussion of SFRs and their TRL can be found in Section 2.3.

Table 2.1: Definitions for the TRLs used in this report. Adapted from Ref. [23]

TRL	Definition & Description
1	**Basic principles** Research identifies the principles that underlie the technology *eg. A particular material or chemical has been identified that shows promising applications*
2	**Basic concept** Practical applications of basic principles are formulated into a basic concept *eg. Core design or fuel type with preliminary modelling or experiment design with an initial screening of materials and methods*
3	**Experimental demonstration of basic principles** Basic principles or a component is successfully demonstrated *eg. Fuels have been fabricated, subjected to out-of-core testing and examined or a chemical extraction process has been demonstrated*
4	**Lab scale demonstration of concept** Principle components are integrated into a basic system *eg. Representative assembly sections have been manufactured and test reactor irradiation trials of individual materials / components have been conducted with limited success*
5	**Lab scale demonstration in representative environment** Basic system is successfully demonstrated *eg. Materials / components have been irradiated and performed successfully in representative conditions such as a reactor test loop or xhemical extraction process on real HLW feed liquor.*
6	**Prototype plant construction** A pilot or prototype is constructed (much more representative than the basic system) *eg. Small scale reactor that consists of many components that will be in the final design has been constructed. Small throughput, batch reprocessing. Small throughput fuel assembly glovebox line*
7	**Prototype plant demonstration** A pilot or prototype is successfully demonstrated *eg. Small scale reactor has been operated successfully with representative conditions*
8	**Demonstration plant and qualification of concepts** A small scale system has been constructed and commissioned that is designed to meet all of the reference requirements *eg. Smaller than commercial scale plant has been constructed and commissioned.*
9	**Successful operation of actual system** Industrial scale system is successful under operational conditions *eg. Reference reactor has performed successfully under operational requirements. Industrial reprocessing of thermal reactor fuel.*
(10)	**(Widespread, reliable and long term operation of many actual systems)** *eg. Commercialised LWR technology.*

Table 2.2: Summary of solid fuel fast reactors, their TRL and relative advantages and disadvantages [23].

Reactor	TRL	Advantages	Disadvantages
SFR	9	1) Most deployable fast reactor in the near-term. 2) Potential to use fissile materials sustainably. 3) Potential to minimise the lifetime of nuclear waste and the burden on a repository. 4) UK has existing experience at Dounreay.	1) Issues with sodium leaks, which can cause fires and component damage, shutting down reactors. 2) The UK is behind in terms of research, however the UK is still well placed to contribute to international collaborations.
LFR	5	1) Lead is chemically compatible with air and water (unlike the SFR). 2) Lead freezing has benefits in terms of physical protection and shielding.	1) Corrosion issues, most significant for pumps and fuel reloading. 2) Limited civil experience. 3) Coolant activation.
GFR	3	1) Chemically inert and transparent coolant. 2) Higher outlet temperatures relative to other FRs therefore useful for process heat applications. 3) UK is well placed to re-engage based on current experience with gas cooled reactors.	1) Immature materials development (fabrication and performance). 2) Components need developing (blowers, DHR), and significant testing on safety features (backup pressure, heavy gas injection system).

2.3 Sodium-cooled fast reactors

At the end of this section there is a brief summary of TRLs and their justification in Table 2.5.

2.3.1 Technical overview

The Sodium-cooled Fast Reactor (SFR) is the most developed fast reactor concept. SFRs have been in operation since 1951 with EBR-I, which was the first electricity generating nuclear reactor. The sodium coolant does not significantly moderate neutrons making the spectrum much harder than thermal reactors, and has a higher thermal conductivity than thermal reactors. Therefore, the core can cooled be much more compact and still have adequate cooling. Outlet temperatures are typically up to 550 °C, at which there are relatively few applications for process heat. As such, SFRs are

aimed at electricity production with a closed fuel cycle to better utilise uranium resources or in transmutation scenarios, to burn long-lived waste from thermal reactor parks.

SFRs can have either a pool- or loop-type configuration, as shown in Figure 2.1. Pool-type configurations have become more widely adopted in later SFR designs, as can be seen in Table 2.4. This is due to inherent passive safety features available with a pool configuration. The sodium pool has a high thermal capacity which allows passive cooling systems to adequately cool the core in the event of an accident. In addition, loss of coolant accidents (LOCAs), which are usually caused by leaks in the primary coolant circuit, are prevented by all primary pipework being retained inside the reactor vessel [27].

There are some advantages to a loop configuration, which is currently being pursued by Japan. Maintenance is simpler as isolated cells can be used for heat exchangers and less shielding is required to limit activation of the secondary sodium loop. Coupling of the heat exchange system is tighter so it responds faster to temperature changes in the reactor, which enhances control factors and temperature dependent feedback systems. It is also possible to change the positioning of the intermediate heat exchanger to enhance natural circulation of the loop. Intermediate heat exchange position can be used to improve the transfer of heat from the core to an ultimate heat sink in an accident condition where there is a loss of pumping [27].

Modern SFR designs have sufficient active and passive safety features. Aspects such as positive void coefficients that were inherent to previous SFR designs have now been engineered out with modern core configurations (eg. the French SFR design, Advanced Sodium Technical Reactor for Industrial Demonstration (ASTRID)), or have been designed around with active features. Other safety and reliability issues that are still concerns include pump failures or leaks in pipes and steam generators. The latter leads to sodium-water or sodium-air interactions, which can cause fires or explosions, damaging components with the potential to put a reactor out of service for prolonged periods.

One of the best ways to obtain a high breeding gain is through the use of radial breeder blankets. However, this produces plutonium with a very high proportion of ^{239}Pu which is suitable for weapons. As such, it is preferable to omit radial breeder

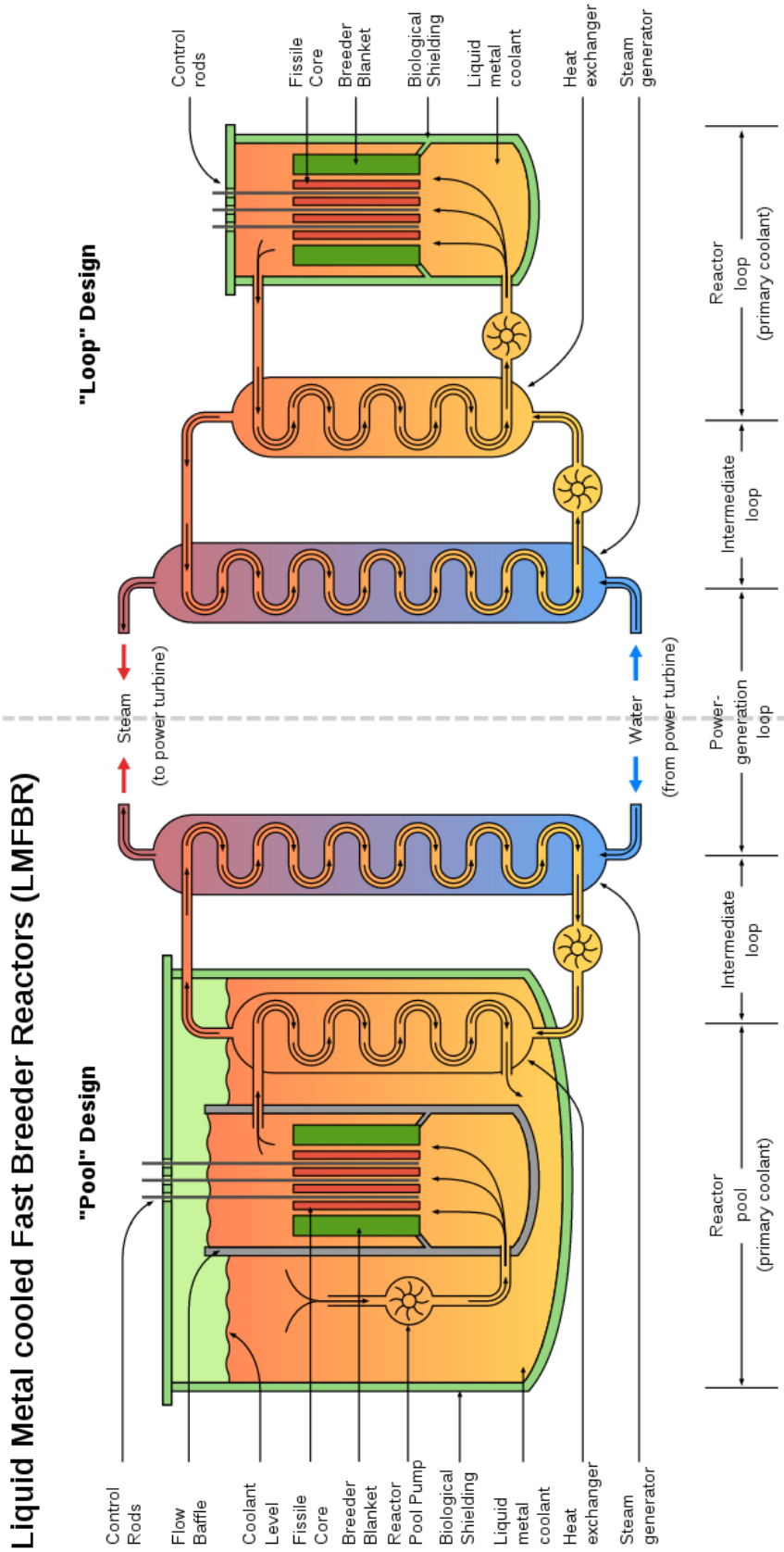


Figure 2.1: Comparison of pool-type and loop-type SFR designs, from Ref. [28].

blankets, which limits the maximum breeding gain [29].

Assuming the long-term use of nuclear power, conversion ratios (CR) of just above one is required. This would keep reactors fuelled without producing excess fissile material. SFRs also have the potential to contribute to partitioning and transmutation (P&T) scenarios, using an SFR as a burner with a CR < 1. However, despite there being test fuel assemblies fabricated and irradiated, there is no experience operating reactors for transmutation. SFRs set up as burners can work in the fuel cycle in two ways: Minimising the waste of a fast reactor fleet by recycling Minor Actinides (MAs), including them in the fuel or blanket region; or using a few fast reactors to burn the TRU inventory from an LWR fuel cycle.

There has been considerable experience with SFRs worldwide, including in the UK which operated the Prototype Fast Reactor (PFR) and Dounreay Fast Reactor (DFR) reactor. PFR and DFR tested several fuel types and cladding materials ranging from common austenetic alloys to more unusual refractory metals and low-swelling nickel alloys, see Table 2.4. At present, SFR designs typically use austenetic and ferritic martensitic (FM) alloys for cladding and wrapper materials. These in-core materials have limitations in terms of radiation damage and temperature. Therefore, R&D programmes for more advanced reactors look at developing materials that retain their characteristics up to high burnups over a broad range of temperatures.

Rankine steam-cycles for power production are the basis for nearly all designs, using an intermediate loop to separate active sodium from the steam-cycle. The development of gas Brayton cycles for power conversion are considered as part of many national programmes. This is an attractive concept as it removes the issue of sodium-water interactions and increases the efficiency of a system.

SFRs are being developed as part of many national programmes and therefore vary significantly in their design and use. Their size ranges from SMRs to large > 1 GWe commercial power generators. The typical goals for SFR development are:

1. Efficient use of natural resources, aiming for a breeding ratio up to 1.3 and doubling time of 30 years for the whole system,

2. Waste minimising capability by the inclusion of all transuranics (TRUs) from spent fuel,

3. Proliferation resistance by avoiding the production or separation of pure plutonium,

4. Active and passive safety,

5. Economic competitiveness.

2.3.1.1 Typical SFR design

Describing an typical SFR design is difficult due to the different aims of national programmes. Current reactor designs lie in three broad areas: modular (typically $<$ 300 MWe), medium sized (typically $<$ 600 MWe) and large ($>$ 600 MWe) [30]. As such, here we define the SFR reference design as something to meet the minimum requirement of a medium to large ($>$ 300 MWe), near-term commercial demonstrator.

Based on current reactor experience, the fuel would be based on MOX aiming at an average burnup of 10 to 15 at%. Cladding and wrapper materials would be based on early austenitic and FM stainless steels which have been extensively tested but limited to 100 to 150 dpa (variants on 316, 15-15Ti and HT9 alloys). Performance of the cladding under irradiation is the main limiting factor, restricting the fuel burnup to less than 15 at%. The reactor would be pool-type with a Rankine steam cycle using mechanical pumps, and mature steam generator designs (straight or helical tube).

Using current SFR experience as a guide would result in an SFR design that could be built and operated with todays experience, using fuels, materials and components which have extensive operating experience, giving it a high TRL. Some of the aims of SFR R&D programmes are not met by this reference design. The TRLs for more advanced concepts that can meet these requirements are discussed in Section 2.3.5, which include MA burners, advanced in-core materials, components and power conversion systems. Such large diversity originates from the different experience and aims of national research programmes. As such, the base reference design meets the requirement of a commercial prototype similar to those being designed and built in China, France, India and Russia.

2.3.1.2 Current Design Types

Reactors which are under construction are a good indicator of what a near-term reactor would look like. Similarly, SFRs which are in the design stage show what new aspects are being considered for future reactor concepts. The specifics of materials and components are discussed in Sections 2.3.2 and 2.3.3.

- PFBR (India - Under construction, operation 2014) – 500 MWe demonstrator using MOX fuel and D9 austenetic steel cladding, aiming at a burnup of 10 at%. Design includes an advanced FM steel being used for the vertical shell and tube steam generator [31, 32].
- BN-800 (Russia - Under construction, operation 2014) – 800 MWe design using basic austenetic steel and MOX fuel. Design is almost identical to BN-600 (Table 2.4) but scaled up and using plutonium containing fuel [30].
- BN-1200 (Russia - Design stage) – 1200 MWe commercial reactor based on BN-800. Design and materials are similar, the most significant difference is the consideration of mixed nitride fuel [33].
- AFR-100 (USA - Design stage) – 100 MWe SMR with long life U-Zr fuelled core aiming at a 30 year refuelling interval. It is designed to use several novel components: a supercritical CO_2 (S-CO_2) Brayton cycle, electromagnetic (EM) pumps, advanced austenitic cladding and a novel twisted tube intermediate heat exchanger [34].
- ASTRID (France - Design stage) – 600 MWe demonstrator using MOX fuel with advanced austenitic steel cladding (AIM1) and FM steel wrapper (EM10). The power conversion cycle is based on advanced concepts: either an inverted steam generator for a steam cycle, or Printed Circuit Heat Exchanger (PCHE) for an inert, nitrogen Brayton cycle. Other advanced concepts include the use of EM pumps [35, 36, 37].
- ESFR (Europe - Design stage) – Large commercial design, aiming at a power output of 1500 MWe. MOX or mixed carbide are the reference fuel options, with a second phase core demonstrating MA bearing oxide fuel. ODS steel has been suggested for the cladding with advanced FM steel (EM10) for the wrapper material [38, 39, 40].

- JSFR (Japan - Design stage) – The only loop-type design listed. Intended to have a large power output of 1500 MWe. The design features high burnup oxide fuel (up to 25 at%) with ODS steel cladding, aiming to reach 250 dpa. Piping and steam generators will use advanced steels, 9-12Cr FM steel [41, 42].
- PRISM (GE-Hitachi - Design stage) – An SMR designed consisting of 311 MWe modules using U-Pu-10Zr fuel and qualified FM steel cladding, HT9. It is designed to use EM pumps for the coolant and use a conventional steam cycle [30].

2.3.2 Key materials and chemistry

2.3.2.1 Fuel

In the near-term, reactors are expected to use oxide based fuels due to significantly greater experience available. Oxide fuels would not include MAs due to the lack of significant experience.

In the medium-term, fuels are likely to still be oxide based, but in the long-term, the development of more advanced fuels is likely. Metallic fuels are one of the more commonly investigated fuels, especially in the USA, with nitride and carbide considered in some national programmes. Nitride fuel is planned for BN-1200 in Russia where there is experience operating BR-10 with UN fuel [30, 33, 43]. India currently leads the way with carbide fuels which are currently used in FBTR and are under long-term consideration for use in the European SFR project ESFR [31, 38, 44]. Metallic fuels have been operated in the EBR-II and DFR reactors, and are part of many national R&D programmes. China, India, South Korea and the USA consider metallic fuels as a long-term design goal [45, 46, 47, 48, 49, 50].

Many long-term commercial SFR designs consider the potential to incorporate MA into their fuel [38,43,51]. The fuel can be designed in two ways [39,52,53,54,55,56,57]:

- Homogeneously – Minor Actinide Bearing Driver Fuels (MADF) have small quantities of MAs homogeneously mixed with the fuel. The expected concentration of minor actinides within the fuel is < 5 wt.%. This is limited by the negative effect MA bearing fuels have on safety aspects and fuel cycle facilities.[1]

[1]Safety issues include sodium void worth, Doppler effects, reactivity swing and delayed neutron fraction.

This has advantages in terms of proliferation with highly active fuel and the potential to develop reprocessing techniques where there is no isolated plutonium stream.

- Heterogeneously – Minor Actinide Bearing Blankets (MABB) used in the core periphery, with MAs in more significant quantities, expected to be between 10 to 40 wt.%. MAs will be in an inert matrix or uranium based sub-assembly positioned in or around the core. Studies have shown that this has less of a negative impact on safety aspects of the core. However, it would require a more technically complex fabrication facility dedicated to high MA content sub-assemblies.[2]

MADF, with an MA content < 5 wt.% have been irradiated in tests such as SUPERACT, NIMPHE, AM1, SHPERE, GACID, FUTRIX, CONFIRM, METAPHIX, AFC-1 and AFC-2 with americium and neptunium in oxide, nitride and metallic fuel [44, 57, 58, 59, 60, 61, 62, 63]. A more detailed overview of MA bearing fuels that have been fabricated can be found in Section B.2. Most of these studies have focused on the impact on fuel properties, reactivity and control effects [47, 64]. At present there has been no real experience with carbide fuels or the addition of curium into fuel.

There is less experience with MABBs. To the authors' knowledge SUPERFACT has been the only experiment. However, there are currently irradiation tests underway (the MARIOS and DIAMINO experiments) [58].

2.3.2.2 Cladding and wrapper

Materials for use in an SFR need to have good mechanical and corrosion properties up to high levels of irradiation at low temperature where brittleness can occur, and at high temperature in order to be safe in accident conditions. In the near-term, in-core materials should be capable of 100 to 150 dpa, but in the long-term the development of materials should allow for greater irradiation up to 200 dpa if not higher.

In the near-term, cladding and wrapper materials would be based on materials that have been qualified or may be qualified in a short timescale. The main examples are

[2]This would be completely separate from the more simple U-Pu fuel fabrication facility.

austenitic stainless steels which are variants on 316 and 15-15Ti [65], the low-swelling nickel-based super alloy PE16 and early FM steels like HT9 [30, 41, 66].

In the medium- to long-term, cladding and wrapper materials that show promise are: advanced austenitic stainless steels, such as AIM1/2 [67] and NF709 [48]; advanced FM steels, such as EM10/12 [67], NF616 and HCM12A; high-Cr ODS steels, typically a 9Cr-ODS [38, 42, 68]; and materials with surface treatments and refractory alloy coatings [67, 69]. These types of materials need testing and qualification in relevant conditions, although some already have relevant experience or tests underway as part of national programmes. There is irradiation experience with AIM1 and EM10 which are being considered for cladding and wrapper materials in the French ASTRID design. Similarly, but to a lesser extent, ODS steels have been used for sample fuel pins and irradiated in BOR-60 and MONJU [67, 70, 71, 72, 73, 74, 75]. Refractory liners have been looked at internationally with the use of niobium with a vanadium liner within DFR. They have also been the subject of research in civil and space based reactors in the USS and France [30, 67, 76, 77].

2.3.3 Other reactor specific issues

For the purpose of brevity out-of-core material and components have been moved to Appendix A. Whilst important for the TRL assessment of SFRs they do not feature in the SFR design and fuel cycle modelling aspects of this thesis.

2.3.4 Operating parameters

This section onwards is original work and not taken from from the authors previous work in Ref. [23, 24, 25].

To get an idea of the limiting factors in SFR core design, key design and operating parameters have been listed in Table 2.3. This list covers demonstration reactors > 840 GWth, commercial designs and relevant reactor designs based on PRISM which are of particular interest to this study.

Full thermal hydraulic and safety assessment of fast reactor designs is outside of the scope of this study. However, it is important to have feasible reactor concepts. Therefore certain design aspects will be kept within the range of previous reactor

designs. Fuel and coolant volume ratios need to be in an acceptable range to make sure heat is effectively removed from the fuel. Height to diameter ratios should be selected to ensure that leakage is enhanced during accident conditions to promote negative feedback effects. Linear power should be low enough to ensure that there is a large enough margin to fuel melting in accident conditions. Design and operating parameters were based on a PRISM-type reactor design, shown in Table 2.3. With the reactor design processed discussed in Chapter 4.

The fabrication, performance and operating parameters of metallic and oxide fuels will be discussed further in Section 2.5.2, including MA bearing fuels.

2.3.5 TRLs and justification

Table 2.5 summarises the TRL conclusions for SFRs. The TRL for each reactor concept is related to the experience of SFRs using the specified materials and components, not the TRL of the materials and components themselves.

There are many variations in SFR design. Advanced concepts outlined in this section broadly summarise some of the more significant and actively researched variations from national programmes. These aim at improving characteristics such as safety, reliability and efficiency. The typical design outlined here is based on materials and components that have been developed extensively in past SFR programmes. Therefore it represents the type of SFR that could be constructed today with minimal supporting R&D. That said, there are near-term demonstration reactors that are being developed with more advanced components and materials. As such, this shows that any near-term systems can be used to test more advanced materials and components as a stepping stone to developing more advanced long-term designs.

The typical design described in Section 2.3.1.1 would use oxide fuel, established materials and conventional steam cycle. This has been assigned a TRL of 9 due to significant international research programmes and successful performance of materials and components in prototype and demonstrator reactors. These types of reactors can be seen in Table 2.4. The main advantage of this design is that it is deployable in the near-term.

Advanced fuels are considered over MOX due to their better thermal conductivity and increased fuel density, which is advantageous in SFRs. The increased fuel density

Table 2.3: SFR design parameters, adapted from Ref. [30] and used to determine typical SFR design parameters.

Reactor	GWth	Diam (m)	Height (m)	H/D	CR	Enrichment zones I(%)	M(%)	O(%)	M/I	Fuel Vol %	BU Gwd/t	Peak Lin. kW/m	Peak/ Av Lin
COMMERCIAL													
Super-Phenix 1	2.99	3.70	1.00	0.27	1.18	16.0	19.7		1.23	37.0	60-90	48.0	1.6
Super-Phenix 2	3.60	3.97	1.20	0.30						37.0	85-136	48.0	1.6
DFBR	1.60	2.99	1.15	0.38	1.20	11.0	16.0		1.45	39.0	90-110	41.0	1.64
CDFR (UK)	3.80	3.00	0.78	0.26	1.15	15.0	20.5		1.37	25.0	115-170	43.0	1.54
BN-1600	4.20	4.45	0.88	0.20	1.10	18.2	21.1		1.16	41.5	115-170	48.7	1.62
BN-800	2.10	2.56	1.00	0.39	0.98	19.5	22.1	24.7	1.13	34.0	66-98	48.0	1.55
EFR	3.60	4.05	1.07	0.26	0.98	18.3	26.9		1.47	36.1	134-190	52.0	2
BN1800	4.00	5.17	0.80	0.15		14.8				44.6	66-118	41.0	1.71
JSFR	3.53	5.38	1.00	0.19	0.85	11.5	13.0		1.13	36.0	150-220	40.0	1.60
DEMO													
Phenix	0.56	1.39	0.85	0.61	1.16	18.0	23.0		1.28	37.0	100-150	45.0	1.67
SNR-300	0.76	1.78	0.95	0.53	1.10	25.0	36.0		1.44	29.5	57-86	36.0	1.57
PFBR	1.25	1.97	1.00	0.51	1.05	20.7	27.7		1.34	29.7	77-113	45.0	1.57
MONJU	0.71	1.80	0.93	0.52	1.20	16.0	21.0		1.31	33.5	80-94	36.0	1.71
PFR	0.65	1.47	0.91	0.62	0.95	22.0	28.5		1.3	35.0	150-200	48.0	1.78
CRBRP	0.98	2.02	0.91	0.45	1.24	32.8				32.5	50-74	40.3	1.51
BN-350	0.75	1.58	1.00	0.63	0.00	17.0	21.0	26.0	1.24	38.0	58-97	40.0	1.67
BN-600	1.47	2.05	1.03	0.50	0.85	17.0	21.0	26.0	1.24	37.5	60-97	47.0	1.68
KALIMER-150	0.38	1.56	1.00	0.64	1.05	21.1				37.6	88-121	28.7	1.43
ALMR	0.84	2.43	0.66	0.27	1.23	21.0	25.2		1.2	37.8	90-125	31.0	1.63
PRISM-type [78, 79, 80, 81] (Relevant designs to this study)													
ABR Zr (prelim)	1.00	2.18	1.01	0.46	0.75	16.1	20.2	24.2	1.25	29.3	100	39.3	1.525
ABR MOX (prelim)	1.00	2.18	1.37	0.63	0.75	21.3	26.6	32.0	1.25	41.7	100	30.1	1.483
ABR Zr (final)	1.00	2.40	0.86	0.36	0.84	18.6	24.6		1.32	39.0	93		
ABR MOX (final)	1.00	2.40	1.15	0.48	0.82	20.5	23.7	30.4	1.28	41.1	132		
PRISM (UNF)	0.84	2.45	0.66	0.27	0.72	18.9	22.7		1.2		87.5		1.4

Table 2.4: Worldwide operating experience of SFRs, adapted from Ref. [27, 30, 82].

Reactor	First Critical	Country	Power MWth	MWe	Type	Fuel	Cladding
Experimental							
EBR-I	1951	USA	1.2	0.2	Loop	U[1]	-
BR-5/10	1958	Russia	8	0	Loop	UN (PuO_2)[2]	Cr16 Ni15 Mo3 Nb
DFR	1959	UK	60	15	Loop	U-7%Mo[3]	Nb
LAMPRE	1961	USA	1	0	Loop	Molten Pu-Fe	-
Fermi	1963	USA	200	61	Loop	U-10Mo	Zr
EBR-II	1963	USA	62.5	20	Pool	U-Zr[4,5,6]	SS316
Rapsodie	1967	France	40	0	Loop	MOX	SS316
BOR-60	1968	Russia	55	12	Loop	MOX[5,6]	Cr16 Ni15
SEFOR	1969	USA	20	0	Loop	MOX	Na
KNK-II	1972	Germany	48	20	Loop	UOX & MOX[5]	1.4970
FFTF	1980	USA	400	0	Loop	MOX[5]	SS316, HT-9
FBTR	1985	India	40	13	Loop	PuC-UC	Na
JOYO	2003	Japan	50-140	0	Loop	MOX	SS316
CEFR	2010	China	65	23.4	Pool	UO_2	Na
Demonstration							
BN-350	1972	Russia	750	130	Loop	UOX[5]	Cr16 Ni15 Mo2+MnTiSi (CW)
Phenix	1973	France	563	255	Pool	MOX[5,6]	Cr 17 Ni 13 Mo 2.5 Mn 1.5 Ti Si
PFR	1974	UK	650	250	Pool	MOX[5]	SS316, PE16
BN-600	1980	Russia	1,470	600	Pool	MOX[5]	Cr16 Ni15 Mo2+MnTiSi (CW)
MONJU	1994	Japan	714	280	Loop	MOX	Mod-316
Super-Phenix 1	1985	France	2,990	1,242	Pool	MOX	Cr 17 Ni 13 Mo 2.5 Mn 1.5 Ti Si

[1] Mk I U, Mk II & III U-Zr, Mk IV Pu-Al.
[2] PuO_2 (BR-5 MK I), UC (BR-5 Mk II), BR-10 UN, (U,Pu)N.
[3] Mk II U-Cr, Mk II & III U-Mo.
[4] U-Fs Mk I-II, Mk III-IV U-Zn, Mk V U-Pu-Zr.
[5] These tested multiple carbide and/or nitride fuel pins and assemblies.
[6] Irradiated some test assemblies containing MAs.

improves the neutronics of the system and allows for higher breeding gains. However, there are issues with advanced fuels such as activation of advanced fuel matrices and under-developed fuel cycles. Metallic fuels are a promising case as they do not significantly interact with the coolant. Advanced fuels are assigned a TRL of 6. Prototype reactors have used metallic, carbide and nitride fuels as well as performing significant R&D with test pin irradiations. In some cases such as U-10Zr (qualified and used in EBR-II), UN (used in BR-1) and (U,Pu)C (used in FBTR) prototypes have been demonstrated successfully with full fuelled cores and are therefore assigned a higher TRL of 7.

MA bearing fuels allow for the management of long-lived waste, minimising the life time of radiotoxicity and required thermal capacity of a repository. There are however issues due to the fuel cycle being under developed and the negative impact MAs have on fuel performance and reactor control aspects. Minor Actinide bearing Driver Fuels (MADF) are assigned a TRL of 5. There have been multiple irradiation tests in representative conditions with americium and neptunium included in fuels. The introduction of curium into the fuel would lower this to a TRL of 2, as minimal work has been done in this area. Minor Actinide Bearing Blankets (MABB) with high MA content are TRL 3, with minimal experimental experience.

Cladding materials are the main limitation on burnup. Therefore advanced materials aim at increasing fuel burnup to improve efficiency and enhancing safety aspects by performing well up to higher temperatures in accident conditions. However, it is difficult to identify materials that retain their properties up to high temperatures and irradiation doses. Materials which do perform well in these conditions tend to have significant issues in terms of fabrication and joining. Advanced steel cladding materials are assessed as having a TRL of 5. Advanced austenitic, FM and ODS steels are all assigned a TRL of 5 as fuel pins have been fabricated using these materials and irradiated (some cases are waiting for Post Irradiation Examination (PIE)). However, reactors have not operated using these materials for the main driver fuel. The use of refractory alloys as cladding and liners are assigned a TRL of 4 due to limited operating experience stemming from in-core performance issues. Some experience exists due to the use of niobium cladding in the UK's DFR as well as research into more advance refractory alloys in France and the USA. Whilst some advanced FM and austenitic

steels are qualified, or close to being qualified, they are limited in their use and require more development and further qualification.

TRL justification of out-of-core materials and components has been moved to Appendix A.

Table 2.5: SFR TRL summary table.

SFR	TRL	Justification
Reference design w/ UOX or MOX	9	Based on vast experience gained in operating large reactors. However, there has been no commercial deployment and most reactors are prototypes or demonstrators with low capacity factors.
Metallic, carbide or nitride fuels	5 – 7	There have been pilot reactors operating with these fuels and even more test irradiations. However, no large demonstration reactor has operated using any of these as the driver fuel.
MA bearing fuels	2 – 5	There have been successful experiments irradiating test fuel pins containing MAs. However, no research has gone beyond using them in a basic system. Carbide MA fuels and fuels containing Cm are assigned a TRL of 2 due to no experience. Oxide, nitride and metallic fuels containing Np and Am up to 3 wt% (MADF) are assigned a TRL of 5 due to multiple experiments, some up to high burnup. Similar fuels with 10-40 wt% Am content (MABB) are assigned a TRL 3 due to limited experimental work.
Adv. cladding material	4 – 5	Advanced austenetic, FM and ODS steels have been used as fuel pin cladding, irradiated in experimental reactors and performed well. However, they are not qualified and have not been used in a whole system so have been assigned a TRL of 5. There is limited experience with refractory alloys and liners, so they are assigned a TRL of 4.

2.4 Down selection of SFR fuels

Most significant factor that will influence reactor performance is the fuel. Fuel choice will dictate the type of fabrication and reprocessing facility and have the greatest impact on the spent fuel inventory.

Oxide fuels are the most widely used and highest TRL. Alternative ceramic fuels (carbide and nitride) and metallic fuels have lower TRLs but have several advantages over oxides. The heavy metal density of non-oxide fuels is much greater. As a result, lower enrichments are possible and cores can be smaller for the same heavy metal

inventory. This is advantageous in terms of breeding, increasing the heavy metal loading of fertile material in a core. Alternatively, smaller cores are more efficient, reducing the amount of coolant and structural material available for parasitic neutron absorption. The better thermal conductivity of alternative fuels means that average fuel temperature is lower giving greater margins to melting in accident conditions [83].

Oxides have approximately two oxygen atoms for every heavy metal atom, neutron scattering from light oxygen causes some moderation and a softening of the neutron spectrum. Carbide, nitride and metallic fuels have less moderating atoms per heavy metal atoms so the neutron spectrum is harder. This has several affects:

- More neutrons per fission;
- Greater fission to capture ratio in heavy metal nuclides resulting in more fertile fission, and less of a build-up of higher actinides through capture;
- Lower neutron losses as a result of parasitic absorption but more neutron losses as a result of leakage.

Overall, carbide, nitride and metallic fuels result in more neutrons available for breeding or transmutation than oxide fuels. In an Argonne National Laboratory (ANL) study, there were 15% more neutrons available in carbide fuelled fast reactors compared to oxide, and 22% more neutrons in metallic fuelled fast reactors [22].

Metallic fuels have the highest density and hardest neutron spectrum compared to carbide and nitride fuels. Therefore, metallic fuel will have the most significant impact on fast reactor neutronics. As such, metallic fuels were selected for this study and compared to oxide fuels, which have the softest spectrum and lowest heavy metal density. This will allow the relative merits of metallic fuels to be assessed against MOX fuel to determine if the development of low TRL metallic fuels is worth while over higher TRL MOX fuels. Previous studies have shown that, in transmutation scenarios, oxide fuels require higher enrichments than metallic fuels to get the same transuranic consumption rates [84].

2.5 Fuel cycle facilities

In fuel cycle models the type of facilities represented must be realistic. Reactors, fuel fabrication and reprocessing techniques must be compatible, with fuel cycles developed as an entire system rather than individual components.

Fast reactor fuel cycles have had limited use worldwide. A lot of fast reactor fuel cycle technology has been tested and piloted to different extends. Advanced fuel cycles for transmutation, using minor actinide (MA) bearing fuels have no industrial experience. A lot of research has looked at transmutation fuel cycles, modelling the fuel cycles and developing fundamental methods. The main limitation of fast reactor fuel cycle studies is the use of fuel cycle technologies that have not been demonstrated on a commercial scale.

To address the limitations of advanced fuel cycle studies, fuel cycle facilities were reviewed and assigned TRLs. The TRL assessment aimed to give an overview of the types of fuel cycle facilities considered for fast reactors, their advantages, disadvantages, and stage of development. In assigning facilities a TRL it is possible to test a range of fuel cycle options in fuel cycle modelling to determine the relative merits of low TRL fuel cycles compared to high TRL fuel cycles.

At the end of Section 2.5.1 and 2.5.2 there is a brief summary of TRLs and their justification. For reprocessing technology Table 2.6 and for fabrication technology Table 2.7. This section gives a brief overview of the experience of fuel cycle technology and their limitations for TRL assessment. A full description of technology and as well as experimental to demonstration scale experience can be found in Appendix B.

2.5.1 Reprocessing

Reprocessing was originally developed as part of nuclear weapons programmes to extract plutonium from low burnup spent fuel. Reprocessing was then developed to extract plutonium from commercial thermal reactors to close the nuclear fuel cycle. As a result, all commercial scale reprocessing uses the PUREX process to extract uranium and plutonium from thermal reactor fuel irradiated to less than 50 GWd/t. There have been demonstrations of thermal MOX and fast reactor fuel being reprocessed but not on the same scale.

The development of reprocessing technologies for fast reactors are based on two drivers, waste reduction and sustainable fuel cycles. Waste reduction focuses on reprocessing as part of P&T scenarios to reduce waste volumes. Reprocessing for P&T scenarios involves the separation of long-lived MAs as well as plutonium. Reprocessing as part of sustainable fuel cycles aims to extend the lifetime of natural uranium resources, which was the original driving force for commercial reprocessing. The recent renewal of interest in fast reactor reprocessing for sustainability is a result of emerging nuclear countries with limited fissile resources (eg. India, China) looking to ensure security of supply.

The aims of reprocessing R&D are: Improved economics, enhanced proliferation resistance of sites and products, reduced solid waste and liquid effluents, improved safety, and public acceptance for their sighting and operation. For fast reactor closed fuel cycles there are additional technical goals: Handle higher burnup fuel; short turn around of spent fuel, therefore short cooling time and therefore ability to handle high activity material; potential to extract MAs; flexibility on fuel material and fissile content.

Broadly speaking there are two main areas if reprocessing technologies being developed, aqueous routes (based on commercial PUREX technique) and dry routes (pyro-electrochemical and fluoride volatility techniques developed for fast reactors). The TRL and justification has been summarised in the following section. For examples of technology development stage, technical advantages and disadvantages, please see the supplementary work in Appendix B.1.

2.5.1.1 TRL Justification

TRLs have been assigned based on the experience of each technology with different fuels. These are described in detail below, with a summary in Table 2.6.

Aqueous PUREX reprocessing of thermal reactor uranium fuels was assigned a TRL of 10 as it is currently done on an industrial scale in several countries, Table B.1 [85, 86, 87, 88, 89].

PUREX reprocessing of thermal and fast reactor MOX has been demonstrated in thermal reprocessing plants and in pilot plants, Table B.1 and B.2 [87, 89, 90, 91].

Whilst there are some difficulties with reprocessing these fuels, the issues have been resolved for pilot reprocessing plants, with fuel diluted with UOX in commercial UOX reprocessing plants. Due to several successful pilot scale reprocessing plants, but the relatively small amount of irradiated MOX being reprocessed, thermal and fast MOX PUREX have been assigned a TRL of 7.

There are many advanced aqueous reprocessing techniques being studied to extract plutonium and MAs, Table B.3. Some are post-PUREX extractions, to extract MAs after plutonium, and others are co-extraction alternatives to PUREX, to extract MAs and plutonium [85, 86, 87, 90, 92, 93, 94, 95]. These have only been demonstrated on a lab scale. These experiments have used representative conditions and HLW feed materials but have not been demonstrated in a pilot plant, and several issues have yet to been resolved such as waste routes and solvent cleaning. As such, advanced aqueous reprocessing methods have been assigned a TRL of 4 – 5, depending on the type of technique.

PUREX reprocessing of metallic zirconium fuels have issues with explosions due to nitric acid U-Zr reactions. These issues have been resolved, but to the present author's knowledge, lab scale zirconium fuel reprocessing has not been demonstrated [96,97,98]. As such aqueous reprocessing of zirconium based metallic fuels has been assigned a TRL of 3.

Dry International experience with pyro-reprocessing techniques are summarised in Table B.4 and B.5.

Pyro-reprocessing methods for uranium extraction from metallic fuel has been piloted in the USA to extract uranium from EBR-II spent fuel, extracting the uranium and disposing of the TRU and FPs [87, 94, 95, 99, 100]. This has been successfully demonstrated and a larger system has been designed to process all of EBR-II's fuel. As such, uranium extraction from metallic fuels has been assigned a TRL of 7, and extraction of plutonium a TRL of 6 due to the relatively high losses and further development requirements.

Methods for extracting uranium and plutonium from MOX fuel have been piloted in Russia with the DDP route [95, 99, 101, 102, 103]. This has been very successful with a range of UOX and MOX fuels, Table B.4. The DDP route has has also been

used much more extensively for unirradiated materials to produce fuel feeds for vibrationally compacted fuels. The DDP methods has been demonstrated successfully with representative conditions and fuels, as such it has been assigned a TRL of 6.

Dry methods to extract MAs as well as plutonium from metallic and oxide fuels have been demonstrated in Russia, Europe and other countries on a lab scale [101, 102, 104, 105]. Results have been successful but there are outstanding issues such as waste processing and salt cleaning. As a lot of experiments have used representative fuels and conditions, but only on a lab scale, pyro-reprocessing methods to extract plutonium and MAs from oxide or metallic fuels have been assigned a TRL of 4.

2.5.1.2 Overview

If economics was the main priority then aqueous PUREX reprocessing methods are preferable due to lower development needs. If transmutation scenarios are the main goal for the fuel cycle then MAs require extraction with either pyro-reprocessing or advanced aqueous methods. If proliferation resistance is a concern then pyro-reprocessing methods are preferable, as they favour on-site facilities which reduces material transportation and pyro-reprocessing techniques do not produce pure plutonium streams. If large centralised facilities are a priority for economies of scale than aqueous methods are preferable. Generally speaking aqueous methods have lower losses than pyro-reprocessing methods, however pyro-reprocessing may become competitive with further development. If short cooling times are a priority, then pyro-reprocessing methods are preferable as they are not sensitive to radiation, unlike aqueous reprocessing techniques which have issues with solvent degradation.

2.5.2 Fabrication and fuel experience

There has been extensive fabrication of uranium oxide (UOX) and mixed oxide (MOX) fuels for thermal reactors. There has been significantly less fabrication of of ceramic and metallic fuels for fast reactors. Plutonium based fuels and fabrication techniques are related to reprocessing programmes which provide the plutonium feed for fuel fabrication.

The focus of this section will be on the previously selected fuels, metallic U-TRU-Zr and ceramic MOX fuels. Despite their use in fast reactors, they have not been produced

Table 2.6: Summary of TRLs assigned to reprocessing methods

Method	Fuel	Rep	TRL	Justification	Details	Comments
PUREX	TR UOX	U/Pu	9	Commercial facilities Thorp, UP2/3	Upto 99.9% U and Pu separation, <5 years cooling for <50 GWd/t	Longer cooling for higher burnup
PUREX	TR MOX	U/Pu	7	Demonstrated in commercial UOX plants	UP2 required mixing of fuel with UOX	More Pu, heat and activity than UOX. Longer cooling time, dissolver time, more solvent degradation.
PUREX	FR MOX	U/Pu	7	Pilot plants & used in commercial UOX plants	Short cooling times demonstrated in pilot plants <6 months (83 GWd/t)	Same as TR MOX but more significant due to more Pu & higher burnup.
Adv-Aqu	-	U/TRU	4-5	Several methods proven on lab scale with HLW feed	Range of recovery efficiencies	Issues: Clean solvents, waste/effluent solutions, scale-ability.
Aqueous	FR Metal	-	3	Experiments on U-Zr nitric acid explosions	To the authors knowledge there have been no lab scale demos other than dissolution tests.	Fluorine needed to prevent explosions. Dry processes may be more favourable.
Pyro	FR Metal	U/Pu	6-8	Pilot for EBR-II U-Zr fuel	99.7% U recovery, 1 t/y batch demo designed for all EBR-II fuel	TRU and FP are waste.
Pyro	FR MOX	U/Pu	6	DDP Pilot plant	Tons of fresh fuel, 40 kg irradiated fuel, upto 240 Gwd/t, >95% recovery.	Higher losses for irradiated fuel, expected to be be 99% at industrial deployment.
Pyro	FR MOX	U/TRU	4	Lab scale: Russia DOVITA, EURATOM, Korea	20 kg batch oxide reductions, gram scale co-deposition.	Issues: Materials corrosion, salt cleaning, waste forms.
Pyro	FR Metal	U/TRU	4	Lab scale: GNEP, ITU experiments with representative materials.	Gram scale experiments, 50 kg batches in USA, ITU reprocessed MA METAFIX fuel	Issues: Materials corrosion, salt cleaning, waste forms.

on a commercial scale. A lot of development is needed before MA bearing fuels can be fabricated on a commercial scale for advanced fuel cycles. Fuel fabrication methods aim at to produce fuel which performs well in-core and enable fuels to be reprocessed easily.

This section discusses the experience with MOX and metallic fuels focusing on fabrication and irradiation performance. For MA bearing fuel, minor Actinide Bearing Driver Fuel (MADF) is the primary focus, but higher enrichment Minor Actinide Bearing Blanket (MABB) fuel is also discussed, which is high MA content fuel situated heterogeneously in the periphery around the core.

This section discusses the experience with MOX and zirconium metallic fuels focusing on fabrication and irradiation performance. The TRL and justification has been summarised in the following section. For examples of technology development stage, technical advantages and disadvantages, please see the supplementary work in Appendix B.2.

2.5.2.1 TRL and Justification

TRLs have been assigned based on the experience of each technology with different fuels. These are described in detail below, with a summary in Table 2.7.

Oxide fuels The fabrication and use of thermal reactor MOX fuel was assigned a TRL of 10. Many LWRs utilise MOX fuel on an industrial scale, Table B.6, with fabrication facilities consistently producing well performing fuel [89, 106].

Several fast reactors have been operated with MOX fuelled cores. Dedicated facilities have been used to fabricate pellet MOX for the lifetime of reactors [89]. Whilst fast reactors have not been as prolific as thermal reactors, the production and use of MOX fuel is well understood and has been produced on a commercial scale, Table B.6. As such, fast reactor MOX has been assigned a TRL of 9.

VIPAC MOX fuel has been tested in several countries. In Russia it has been successfully demonstrated for whole BOR-60 cores, and tested in BN-350 and BN-600 [103]. However, VIPAC MOX has only been produced on a small scale for a small reactor, therefore it has been assigned a TRL of 8.

MOX fuels, pellet or VIPAC, containing MAs have been fabricated and irradiated

as part of many experimental programmes. A summary of the key programmes, using fuel with $< 40\%$ TRU have are shown in Table B.7. These experiments have been successful and taken place in representative fast reactor conditions and irradiated to representative burnup levels. As such, MA containing MOX fuel has been assigned a TRL of 5, as fabrication has never been scaled up to the pilot plant level.

Metallic Fuels U-10Zr fuels have been qualified for use in fast reactors in the USA and demonstrated on the whole core level in EBR-II [49]. U-10Zr fuels have shown good performance in representative conditions and in accident conditions. As such, U-10Zr fuels have been assigned a TRL of 8.

U-Pu-10Zr fuels were being tested for qualification in EBR-II and the FFTF before the USA SFR research programme was ended [107, 108]. Many pins of U-19Pu-10Zr have been irradiated in representative conditions and up to high burnup, showing good performance. As such U-Pu-10Zr fuel has been assigned a TRL of $6 - 7$ (dependent on enrichment).

U-TRU-Zr fuels have had a similar level of in-core testing in representative conditions as TRU MOX fuel. A summary of the main experiments can be found in Table B.8. Whole U-TRU-Zr fuel pins have been fabricated for EBR-II using the same fabrication methodsas U-10Zr and U-Pu-10Zr, performing well in-core, but with americium volatility issues during fabrication resulting in americium losses. Smaller pins and samples have been made with alternative methods that do not have americium losses. As such, U-TRU-Zr fuels have been assigned a TRL of 5. It is worth noting that whilst U-TRU-Zr fuels have the same TRL as MA bearing MOX, however, the development time would be longer due to less experience with metallic fuels. A drawback of TRL assessment is that the broad levels do not capture past experience of related technology.

2.5.2.2 Overview

If economics was the main priority then MOX fuels are preferable due to lower development needs. If in-core performance such as transmutation or breeding were a priority than metallic fuels may be preferable. If transmutation is a priority then MA bearing fuels are required, with MA bearing MOX fuel expected to have a shorter

Table 2.7: Summary of the TRLs assigned to fuels based on fabrication and irradiation experience.

Fuel	**Process**	**TRL**	**Justification**	**Details**	**Comments**
TR MOX	Pellet	10	Many industrial scale plants	Several successful powder methods used.	UK was unsuccessful operating on a commercial scale with SMP.
FR MOX	Pellet	9	Industrial scale fabrication of FR cores	CFCa - 110 tonnes FR MOX.	
FR MOX	Vipac	8	Demonstrated at RIAR	Used in BOR-60 (whole core)	UOX and MOX fuel from pyro feed
FR TRU-MOX	-	5	FR irradiation tests	Range of Np, Am, Pu fuels manufactured and successfully irradiated up to representative burnups (19.5 at%)	Vibration and Pellet processes tested, no Cm used, Am volatility issue.
FR U-Zr	Inj	7-8	U-10Zr fuel qualification	Whole core EBR-II irradiated, 1000's assemblies made ANL-W	Safety tests and several cladding materials tested.
FR U-Pu-Zr	Inj	6 – 7	U-19Pu-10Zr demonstrated but not qualified	244 fuel pins made, irradiated to 19.3at%, 31% enrich tested	Programme was closed, issues with range of Zr concentrations and unknown phases.
FR U-TRU-Zr	-	5	FR Irradiation tests	Whole pins with MA tested, also smaller samples with range of TRU, RE and Zr content irradiated successfully up to representative burnups (11-30 at%)	Am volatility during fabrication is an issue, swelling issues, no Cm

development time than MA bearing metallic fuels.

2.6 SFR fuel cycles

It is important to assess the TRL of fabrication, reprocessing and reactor technology combined in a duel cycle, as well as individually. This section will discuss real demonstrations of closed, fast reactor fuel cycles and assess the TRL of different closed fuel cycle options, which is summarised in Table 2.8.

2.6.1 Demonstrations of closed SFR fuel cycles

Partially closed fuel cycles, reprocessing LWR fuel and recycling plutonium into MOX fuelled LWRs gave been demonstrated around the world. There have been large scale demonstrations of fast reactors without a closed fuel cycle, using uranium fuel or plutonium from thermal reactors. Fully closed fast reactor fuel cycles have been demonstrated on a relatively small scale. In this thesis a closed fast reactor fuel cycle is described as spent fast reactor fuel being reprocessing, fabricated and irradiated again in a fast reactor. There are four clear demonstrations of this, which are described in this section.

In the USA, EBR-II used melt refining to reprocess 2.3 tonnes of metallic fuel, refabricated for EBR-II with a very short turn around of two months [105]. Although this represents a closed fast reactor fuel cycle the reprocessing method is not representative. Melt refining, which has not been discussed in this chapter, uses the reaction of melted fuel elements with a crucible as an extraction method. Melt refining leaves unwanted FPs and actinides in the crucible which is not very efficient, retaining 90 to 95% of uranium, plutonium and noble metals [88].

In France, spent fuel from Rhapsodie was reprocessed and used in Phenix. Phenix spent fuel was reprocessed and fabricated into fresh MOX fuel, some of this was recycled up to 3 times through Phenix [87]. This clearly demonstrates a closed fuel cycle with aqueous reprocessing and MOX fuel.

In Russia, kilogram quantities of BOR-60, BN-350 and BN-600 UOX and MOX fuel has been reprocessed at RIAR using pyro-reprocessing methods. The PuO_2 product from reprocessing was mechanically mixed with fresh UO_2 and used to make VIPAC

MOX fuel for the BOR-60 reactor. Although plutonium recovery was only 95.6%, this demonstrates a closed fast reactor fuel cycle using pyro-reprocessing methods and VIPAC MOX fuel [99].

In the UK, PFR MOX has been reprocessed totalling 3.7 tonnes of plutonium from fuel and blanket assemblies. Whilst most of this has been stored as PuO_2 some was fabricated into MOX fuel and recycled in PFR [89]. It is unclear from reports how much fuel was recycled through PFR.

2.6.2 Summary of fuel cycle options

The TRL of fuel fabrication, SFRs using them and reprocessing techniques, as assessed by the present author, are summarised in Table 2.8. There are a range of ways that fast reactor fuel cycle facilities could be set up, with some not being compatible.

The overall development level of a fuel cycle cannot be determined from the assessment of individual components. Although components of a fuel cycle may have similar TRLs, the overall development time of a fuel cycle might be long. If each component of a fuel cycle is at the lower end of a TRL, the combination of all of the development needs into one complete system may result in a lower overall TRL.

The aim of Table 2.8 was to assess the TRL of fast reactor fuel cycles as a whole. Whole fuel cycle TRL assessment tries to account for the compatibility of facilities and the longer development timescales of some fuel cycles. In addition, all curium recycling fuel cycles were assigned a TRL of 2. To the present author's knowledge, the fabrication of curium bearing fuels has not been tested and the operation of reactors with high curium content fuels has also been untested.

In later chapters high and low TRL fuel cycles are used as general terms to separate the near-term concepts from more advanced concepts. A TRL of 5 or above is deemed high for a fast reactor closed fuel cycles. A TRL of 5 is not high in of itself, but it was selected so that at least one fuel cycle can be used for transmutation scenarios can be considered as a high TRL transmutation scenario. A TRU (plutonium, neptunium, americium) fuelled MOX reactor with aqueous reprocessing was assigned a TRL of 5, and is considered as the highest TRL concept that can be used for transmutation. All other transmutation scenarios have low TRLs, less than 5. High TRL fuel cycles also include plutonium only recycling fuel cycles.

Table 2.8: TRL of SFR fuel cycle options.

	Fab.	SFR	Rep.	TRL	Comments
					Once through
O1	MOX (9)	(9)	-	9	Has been demonstrated internationally.
O2	U-Pu-10Zr (7)	(6)	-	6	Fuel requires qualification.
					Pu rep Only: Burner or breeder reactor
C1	MOX (9)	(9)	Aq (7)	8	Has been demonstrated in France and the UK. Shortest development time.
C2	MOX (9)	(9)	Py (6)	7	Has been demonstrated in Russia on small scale. Longer development time than C1. Potential for shorter cooling times, fewer process steps, higher losses than Aq, impure Pu stream (PR) and all on-site to reduce transport (PP).
C3	U-Pu-10Zr (7)	(6)	Py (6)	6	Partially demonstrated in USA with uranium fuels. Better breeding potential than C1 & C2, same advantages as C2.
C4	U-Pu-10Zr (7)	(6)	Aq (3)	3	Would not be selected over C3.
					TRU Fuels: Transmutation or PR
C5	TRU-MOX (5)	(4)	Aq (4)	5	More PR than C1. Although same TRL as C6 & C7, shorter development time as more MOX and Aq experience.
C6	TRU-MOX (5)	(4)	Py (4)	4	Shorter cooling so better for transmutation than C5, however longer development times for pyro. Same advantages as C2.
C7	U-TRU-Zr (5)	(4)	Py (4)	4	Harder spectrum means better transmutation than C5 or C6. However, less experience with fuels, reactor and reprocessing so longer development time
C8	U-TRU-Zr (5)	(4)	Aq (3)	3	Would not be selected over C7.
C9	Cm fuel (2)	(3)	(3)	2	A lot of development required.

O – Open fuel cycle.
C – Closed fuel cycle.

Chapter 3

Literature review of fuel cycle studies

3.1 Introduction

Chapter 2 reviewed fast reactor fuel cycle technologies to assess their TRL. This chapter reviews previous fuel cycle studies and the UK fuel cycle to determine gaps in knowledge.

There have been many fuel cycle studies in the open literature, however, very few specific to the UK. The UK is in a unique situation with a large stockpile of plutonium to deal with and an ambitious new build reactor programme.

The aim of this chapter is to get a broad idea of what fuel cycle studies typically assess, the types of parameters used and how they influence results. A broad range of studies were reviewed, focusing on key work from IAEA or NEA technical documents as well as UK specific studies. Typical parameters for fuel cycle scenario studies were reviewed and gaps in previous research assessed, both in terms of general fuel cycle studies and UK specific fuel cycle studies. An overview of the considerations for further study can be found in Section 3.7.

3.2 Aims of fast reactor fuel cycles

Aims of national and international projects for fast reactor fuel cycle development are based on similar drivers for the sustainable growth of nuclear power. The following list

of aims is adapted from the OECD NEA goals: preserving resources and the environment; minimising waste volume, radiotoxicity of waste and size of a repository; safety; reliability; proliferation resistance (PR) and physical protection (PP); public acceptance; and economic competitiveness with alternative energy sources [109, 110, 111].[1] Preserving resources and the environment includes uranium, land for facilities, GHG emissions, employment and long-term security of supply independent of international politics.

These aims are broad, most of which could apply to the ethical development of any new technology. The importance of each of the above aims will vary depending on national drivers. However, the primary goals for nuclear countries considering fast reactors is either sustainability and security of supply, or minimising the burden of waste. China and France are two example cases where fast reactors are part of long-term energy policy.

China With electricity consumption set to rise, China plans to generate 15% of all electricity from nuclear power by 2150. This involves the development of breeders, to maximise uranium resources ensuring security of supply. These will be deployed commercially from 2020 and become the main source of nuclear power by 2100 [50].

France In 1991 France passed a law on long-lived nuclear waste. In 2006 this was updated to include goals on a waste solution, requiring the study of partition and transmutation scenarios [109]. Current work looks at deployment of low conversion ratio SFRs, starting with a prototype in 2020 [50].

3.3 Overview of fast reactor fuel cycle studies

Fuel cycle studies consider international scenarios, national scenarios, or specific reactor park scenarios, with objectives of a fuel cycle varying. In general, there are there are two main drivers for fast reactors fuel cycles. The first is based on sustainability and security of supply, reducing the demand for natural uranium and the dependence

[1] A summary of the broad goals of the following collaborative programmes: Nuclear Energy Agency (NEA), Generation IV International Forum (GIF), IAEA International project on Innovative Nuclear reactors and fuel cycles (INPRO), International Framework of Nuclear Energy Cooperation (IFNEC), European Sustainable Nuclear Energy Technology Platform (SNETP).

on raw material imports. The second driver is focused on waste performance, reducing the volume of waste that needs to be stored, the size requirements of a repository and the radiotoxicity lifetime of waste. To combat these drivers, there are three ways that fast reactors can be operated in a fuel cycle:

- CR > 1 – Breed excess fissile material, either to start new fast reactors or in some cases to support thermal reactors.
- CR $= 1$ – Iso-breeder fast reactors, used to produce enough fissile material to sustain their operation. Any growth of fast reactors would come from external sources of fissile material, such as reprocessed spent fuel from thermal reactors.
- CR < 1 – Burners used to destroy more transuranics than they produce. These would be used to reduce the size of transuranic stockpiles in terms of mass, repository size and radiotoxicity (waste performance factors). Fuel cycle studies typically use low CR fast reactors to support the back end of thermal fuel cycles, burning thermal reactor spent fuel to improve waste performance. Alternatively, burners could be used to reduce stockpiled material, typically in nuclear phase out scenarios.

Some studies consider 2-tier fuel cycles, where spent fuel is recycled once-through a thermal reactors before being used in a fast reactor.

Fuel cycle models simulate uranium enrichment, fuel fabrication, thermal reactors, fast reactors, cooling, storage, reprocessing and a repository. Parameters are chosen for facilities in the fuel cycle, such as reprocessing losses, elements to be recycled, cooling time, reactor generating capacity, reactor design, and how long a fuel cycle scenario is modelled for.

The results and assessment criteria used for a fast reactor fuel cycle study depend on the objectives of the scenario, typically sustainability or waste reduction. Typical assessment criteria are:

- Natural uranium requirements – Different fuel cycles will require different quantities of natural uranium. This is dependent on the number of thermal reactors being operated. Reprocessing and recycling fuel in reactors will offset the number of reactors that need to be fuelled with uranium.

- Stored material buffers – The mass or volume of material being stored, as well as the lifetime of storage facilities.
- Radiotoxicity of waste – The ingested dose of material is used to compare the relative risk of materials over time. The evolution of radiotoxicity after disposal can be compared to that of natural uranium to see how long it takes material in a repository to decay to the same level as the natural uranium, termed radiotoxicity lifetime. Radiotoxicity lifetime indicates the intergenerational liability of nuclear waste, which can influences public opinion on nuclear waste [4].
- Decay heat and repository size – The relative size of a repository impacts on the cost of a fuel cycle and public perception [4]. A single small repository is preferable in terms of minimising the size of the nuclear legacy, planning, sighting and public acceptance. Repository size is dependent on the heat generated by waste.
- Facility requirements – The annual throughput of a fabrication or reprocessing plants determine the number of plants and size needed as well as the transport requirements. The properties of the materials being used will influence technology choices, their cost and development needs. High activity materials require shielding and remote handling. Very hot fuel require changes in transport and handling methods. High fissile content materials will require criticality safe geometry and neutron absorbers as well as international inspection and safeguards in term of proliferation.
- Deployment – The rate at which new fast reactors can be built, based on the material being produced from thermal or fast breeder reactors.
- Proliferation – The fuel cycle can be assessed qualitatively based on the materials and facilities, relating to how easy it is to handle material or adapt a facility for clandestine purposes. Proliferation resistance of fast reactors is dicussed in more detail in Appendix D. Materials in the fuel cycle can also be quantitatively assessed in terms of how much is available and how much would be needed for a weapon.
- Cost – Many studies have estimated the cost and financial risk associated with fuel cycle development.

Assessment criteria are typically presented as the relative improvement over a reference fuel cycle. Typically the reference fuel cycle is the same generating capacity of once-through LWRs, which corresponds to the use of current technology. Once-through LWRs represent the highest TRL and most economically competitive fuel cycle to see if the improvement factors of advanced fuel cycles are large enough to warrant their development. Alternatively, for nuclear phase out scenarios, the reference case is the initial stockpile of material in the fuel cycle to determine the improvement of a fuel cycle scenario over the direct disposal of stockpiled materials.

Studies that focus on reprocessing and improvements on waste performance typically present results for the reprocessing waste stream only. These studies assume that only the waste from reprocessing is sent to a repository and, at the time of the assessment, material buffered in the fuel cycle is ignored, assuming it will be used in the future. The contribution of reprocessing to waste over the fuel cycle lifetime does not account for any final inventories sent to a repository, giving an infeasible best case scenario. Some fuel cycle studies consider waste streams and all material in the fuel cycle at the time of assessment, to show the relative merits of a fast reactor fuel cycle by the assessment date. Assessing all material in the fuel cycle at the time of assessment shows the progress of the fuel cycle if the fuel cycle was stopped at the assessment date and all material sent to a repository.

Fuel cycles are typically assessed at 2100–2200, looking at the relative improvement factors of a fuel cycle over a reference case by the end of the century.

3.4 Results from previous studies

Fuel cycle studies vary based on the initial starting condition of the fuel cycle and fuel cycle parameters. A broad range of fuel cycle studies will be discussed based on their results. Results for the assessment criteria in each study have been given as relative improvement factors over their reference cases for ease of comparing results. The aim is to illustrate the scale of improvements that are possible when operating fast reactor fuel cycles, and how these depend on fuel cycle parameters. It is not the intention to directly compare studies, which servers no purpose due to the different starting conditions and drivers of studies. Results for studies generally presented in two ways:

1. The impact a fuel cycle has on the total nuclear material at the assessment date. Either relative to stockpiled material or relative to a once-through LWR fuel cycle.

2. The impact a fuel cycle has on the reprocessing waste stream, assuming that reprocessing waste is the only material sent to a repository. Not accounting for any material left in the fuel cycle. This is usually given relative to the total SNF from a once-through LWR fuel cycles.

3.4.1 Natural uranium requirements

Fuel cycle scenarios tend to quote the natural uranium saved when the fuel cycle is compared to the same generating capacity of once-through LWRs. Many studies have targets for peak uranium demand [111, 112]. The reduced dependency on uranium is a function of the ratio of fast reactors to LWRs, the CR of fast reactors and the timescales considered.

Fast reactor burner scenarios reduce uranium use, but it depends on reactor design.In a USA scenario using a very low CR (0.5) SFR using reprocessed fuel from LWRs, uranium demand was reduced by a factor of 1.16–1.23 by 2100, compared to a once-through LWR fuel cycle [113, 114, 115].

Breeder reactors can reduce uranium demand by a factor of greater than 50 depending on their rate of deployment and ratio of LWRs to fast reactors [116]. In a USA scenario, using iso-breeders, the uranium demand reduced by a factor of 1.32–1.72 by 2100 [113, 114, 115]. In an NEA study on fuel cycle sustainability [117], iso-breeders reduced uranium demand by a factor of 1.27 by 2100, a CR of 1.2 reduced uranium demand by a factor of 1.73–1.9 by 2100. Given longer timescales uranium demand drops even further with the maximum improvement of 5.64 by 2200 [117].

3.4.2 Radiotoxicity

Radiotoxicity is defined in Section 1.1. The release of radioactive nuclides into the biosphere from a repository is not very sensitive to the use of partitioning and transmutation schemes [88]. The radioisotopes most likely to escape into the biosphere and be ingested are ^{99}Tc, ^{129}I, ^{93}Zr, ^{135}Cs, and ^{14}C which have half-lives of 100,000 to

1,000,000 years [118]. Under normal repository conditions the doses to people as a result of release of these isotopes to the environment are well below regulatory limits. The dose released from a repository is proportional to the number of fissions and not dependent on the use of thermal or fast reactors.

The total radiotoxicity in a repository is only a concern in the case of a disturbed repository, where a repository is compromised then there is the potential for large releases to the biosphere. This could either be a natural occurrence (earthquake, ice age) or human intrusion (intentional or accidental such as mining) [119]. The most significant contributors to radiotoxicity decay away in 300 to 500 years. To reduce this further requires the transmutation of long-lived radioisotopes, the most significant of which are (given as approximate orders of magnitude): ^{241}Am for 10,000 years, ^{239}Pu and ^{240}Pu for 100,000 years [120]. Note, to get the maximum reduction in radiotoxicity curium needs to reprocessed as ^{240}Pu is produced from the decay of ^{244}Cm which would otherwise be sent to the reprocessing waste stream [120]. Measuring improvements in long-term radiotoxicity is either done by the time it takes for waste to decay to natural uranium levels termed 'radiotoxicity lifetime', or the total radiotoxicity at a specified date, assuming a disturbed repository.

In a USA scenario, using low-CR SFRs and recycling all TRUs, the improvement on the total radiotoxicity by 2100, assumed disturbed repository in 1000 years, was 1.6. If a 2-tier scenario was used then the radiotoxcity improvement was 1.1. With partitioning of Cs and Sr, radiotoxicity improvement was 2.1 (1.3 for 2-tier) [113, 114, 115, 121]. Japanese scenarios looking at phasing out nuclear power with 240 years of dedicated SFR burners, managed to reduce the lifetime of radiotoxicity by a factor of 10 [122]

Considering reprocessing waste streams only, the reduction in radiotoxicity is much greater. With relatively low losses, a 2-tier scenarios reduces the radiotoxicity lifetime by a factor of 6, using just fast reactors with plutonium only reprocessing has an improvement factor of 15, and using fast reactors, recycling all TRUs, has an improvement factor of 750, corresponding to radiotoxicity lifetime of 400 years. However, these improvement factors vary greatly based on reprocessing losses [84, 110, 123, 124]. These results are similar for most studies, with variations depending on the losses from reprocessing, the best case scenario with no losses is a radiotoxicity lifetime of 270 years,

where only FPs are present in the waste stream [111, 125]. Reduction factors greater than 10 are only possible with MA reprocessing [111].

3.4.3 Decay heat and repository size

The main limitation on repository size is the heat generated by waste. There are several temperature limitations such a the peak temperature of the waste canister wall and the peak mid-drift temperature between waste canisters. These limitations are dependant on repository design and geology. Typically, the peak drift temperature ($\sim$100°C) is the limiting factor of a repository [111]. Several studies have modelled the build-up of heat in a repository, using codes which specifically model heat transport in geological conditions (eg. CODE_BRIGHT or PORFLOW) [119]. Others studies have used finite element codes such as AQAQUS for thermal analysis [126]. In many cases approximations are used such as cumulative decay heat (CDH). CDH is the integration of decay heat from time of emplacement in a repository to the time when peak mid-drift temperature is expected, this is typically 500-2000 years depending on the study [114, 121, 124, 127].

Decay heat is determined by different isotopes which decay over different time scales. For the first 50 years ^{137}Cs ($t_{1/2}$ = 30 y) and ^{90}Sr ($t_{1/2}$ = 28.8 y) dominate decay heat, along with ^{244}Cm ($t_{1/2}$ = 18 y) if curium or americium are recycled [125]. After 50 years ^{241}Am ($t_{1/2}$ = 432.2 y) and ^{238}Pu ($t_{1/2}$ = 87.7 y) are the most significant contributors to decay heat. The most significant contributor to decay heat up to 1000 years is ^{241}Am after which the contributions of ^{240}Pu ($t_{1/2} = 6.56^3$ y) and ^{239}Pu ($t_{1/2} = 2.41^4$ y) become more relevant [128].

Compared to a once-through LWR scenario, a USA scenario with 0.5 CR SFRs reduced the repository size by a factor of 1.6 by 2100 with all TRUs recycled, with Cs and Sr removal the improvement was 2.1 [113, 114, 115, 121]. In a French study with plutonium and americium recycled in SFRs and 70 years of cooling, repository size was reduced by a factor of 2 by 2150. With curium reprocessed, the repository size decreased by a factor 2.5, and with curium reprocessing and 120 years of cooling the repository size decreased by a factor of 4.6 [129].

A Japanese scenario looking at nuclear phase out and 240 years of SFR burners to reduce stockpiled material, reduced repository size by a factor of 4.7 [122]. Similarly,

a German scenario looking at nuclear phase out and ADS[2] reactors reduced repository size by a factor of 4 by 2100 [124].

When considering reprocessing waste streams only, the potential reduction in repository size is much greater. With low losses, LWR MOX can reduce repository size by a factor of 3.2, using a 2-tier scenario by a factor of 6.7, and using SFRs with all TRUs recycled by a factor of 8.3 [50, 110]. However USA studies have shown that this is very sensitive to losses and the TRUs recycled. A reduction of 4.3–5.4 for plutonium and americium reprocessing would increase to 4.4–5.7 with the inclusion of curium [113, 114, 115, 121]. When partitioning of Cs and Sr is included this increased to 10–43 and 10.5–225 respectively.

Main factors which influence repository size are the heat from FPs and the quantity of TRUs sent to a repository. Heat from FPs can be reduced by cooling prior to repository emplacement or partitioning. A key factor, shown in a Japanese study, is reprocessing cooling time which can lead to more decay of ^{241}Pu to ^{241}Am. A cooling time of 5 years before reprocessing rather than 20 years had an improvement of $\sim$1.7 on repository size for plutonium recycling scenarios [130].[3]

The RED-impact study considered the influence of different geologies on repository size. In a scenario reprocessing all TRUs, a repository size could be reduced by a factor of 2.5 in granite and 3.2 in clay [111, 119, 131]. A similar type of study, with different reprocessing losses, achieved a reduced repository size by a factor of 4.2 in granite, 3.5 in clay and 15.9 in Tuff (Yucca Mountain) [111, 112]. Comparing the granite and clay loading of these two studies, granite has a smaller repository size in one study, and clay has a smaller repository size in the other.

There are factors other than decay heat which influence repository size. Criticality concerns can lead to a larger repository footprint when high plutonium content fuels are used, requiring a larger area for waste packages.

[2]Accelerator driven sub-critical reactors, with very high loading of TRUs that would not be safe in a normal operating fast reactor. The higher TRU loading means lower CR and faster reduction in stockpile size.

[3]Read from a figure.

3.4.4 Curium issues in the fuel cycle

Curium decay chains contribute to long-term radiotoxicity and decay heat, so it is preferable to include curium in fuel, as shown in the previous sections, to reduce radiotoxicity and repository size. However, curium reprocessing has significant handling issues due to ^{244}Cm which is a strong neutron emitter [123]. Curium gradually builds up at the beginning of a fast reactor closed fuel cycle, where all TRUs are recycled, until an equilibrium is reached. The main reason to reprocess curium is to prevent the daughter isotopes in its decay chain being present in the waste stream. Whilst most curium decay heat is gone in a short time scale (^{244}Cm $t_{1/2}$ =18 y) ^{244}Cm decays to ^{240}Pu which contributes to decay heat and radiotoxicity over a 100,000 years [128]. By not recycling curium a lot of curium and ^{240}Pu will be present in the reprocessing waste stream sent to a repository.

The main issue with curium is shielding fabrication facilities from neutrons, and the lack of experience with curium bearing fuels and reprocessing. To the present author's knowledge there have been no fuels manufactured containing curium. ^{244}Cm levels greater than 5 wt.% will require significant neutron shielding of a fabrication plant to prevent high worker doses [118]. Neutron emissions for SFR SNF decay by factor of 4 over a cooling period of 20–30 years [132]. Decay heat and neutron emissions from fuel during fabrications is the same order of magnitude for fuel streams containing neptunium–americium, with the inclusion of curium neutron emissions would be more than two orders of magnitude greater [133]. This would cause issues in terms of facility operation and shielding, but could have benefits in terms of proliferation resistance, making fuel material harder to process and handle increasing intrinsic proliferation resistance, Appendix D.

3.4.5 Once-through burner SFRs

Only one example of a once-through burner SFR fuel cycle was found in the literature, based on USA's disposition of weapons grade plutonium. This scenario irradiated plutonium in an SFR for a short time to reduce the ^{239}Pu proportion and increase the handling dose [134]. Compared to LWR MOX, a once-through SFR was twice as expensive. The main issues were the designing, building, licensing and qualifying of

fuel as well as uncertainty in the design. The waste performance of a once-though SFR was not reported. How the waste performance of PWR MOX and once-though SFRs compare is a significant gap in the literature.

3.5 Parameters

This section outlines fuel cycles parameters used in previous studies and those selected for the study presented in this thesis. Most fast reactor fuel cycle models consider facilities that are not feasible with current technology, therefore operating parameters are estimated.

Deployment times Commercial scale deployment of fast reactors varies across studies. In new nuclear countries which are developing their nuclear programme quickly, such as China and India, SFR deployment is typically 2020–2025 [50]. In Russia and France, which have a slower nuclear build rate but more experience with SFRs, deployment timescales are 2030–2035 [50, 124, 135]. In the UK, USA and some NEA scenarios, deployment timescales are 2040–2050 [21, 115, 117, 121]. In this work an SFR deployment date of 2040 was chosed, based on previous studies related to the UK's nuclear outlook.

Reprocessing cooling times There have been a range of cooling times in real reprocessing scenarios, Table B.2. LWR fuel cooling is usually longer than SFR fuel cooling in fuel cycle scenarios due to the use of older facilities, this can be as long as 10 years [114]. Studies using pyro-reprocessing usually have a 1 year cooling time, with 1 year for reprocessing and fabrication [50, 113]. Many studies look at the influence of cooling from low 2–5 year periods to high 10–20 year periods [117, 130]. Short cooling times have better waste performance due to less americium in-growth and less material being buffered in cooling ponds, increasing fuel availability for reactor operation. In this work a short cooling time was considered to be 1 year and a long cooling time considered to be 10 years, giving a broad range of results. Fabrication and reprocessing times were assumed to be 1–2 years.

Reprocessing losses Many studies look at the impact of losses on reprocessing waste streams from 0.1–10% losses [84,111,121,125]. Plutonium losses typically range from 0.1–0.5% based on current reprocessing technology [88,130]. MA losses in fuel cycle scenarios are either treated the same as plutonium or as having greater losses, typically ranging from 1–5% [111,125,130]. Modern aqueous reprocessing technology can have plutonium and uranium losses as low as 0.1–0.2%. Future aqueous methods aim at 0.1% for neptunium–americium and 0.3% for curium, with $< 5\%$ LAs in the MA stream. The same is expected for pyro-reprocessing but curium losses being greater, roughly 5%, with $< 10\%$ LAs in the MA stream [88]. In this work low losses were considered to be 0.1% and high losses considered to be 0.5%Pu, 1%Am, 5%Cm, giving a broad range of results.

Waste streams Studies tend to have Pu, Pu+Am or all TRU as reprocessing streams. Inclusion of neptunium has a minor influence on waste performance [50, 110,111,125]. Waste streams include removal of Cs and Sr or prolonged cooling of SNF for 100 to 380 years to allow ^{244}Cm, Cs and Sr to decay [121,130]. In this work Pu, Pu+AM and all TRUs were considered. Prolonged cooling prior to disposal was also considered in the range of 50 to 350 years.

3.6 The UK

At present the UK's goal is to build 16 GWe of new LWRs by 2050 [3]. The first of which, Hinkley Point C, is due to come online by 2023 [136]. Reprocessing of fuel at THORP is due to stop in 2018 [14,15] and all Magnox reactor fuel should be reprocessed by 2017–2020 [15]. This will leave the UK with an estimated plutonium stockpile of 140 tonnes including foreign owned plutonium [1]. As part of the consultation on the UK's plutonium stockpile, several credible options for plutonium disposition outlined, with no decision made as yet [17]. Beyond 2050 there is no policy for the UK nuclear industry, but there has been development of a Nuclear Energy R&D Roadmap [21] outlining potential pathways for the UK research, based on three key pathway scenarios shown in Figure 1.3:

- Baseline – Continue operating current reactors, with no new reactors and the

design and operation of a geological repository from 2040.

- Open fuel cycle (16 to 75 GWe) – Baseline plus 16 GWe of LWRs by 2025 and up to 75 GWe of new build LWRs by 2050.
- Closed fuel cycle (16 to 75 GWe) – Baseline plus 16 GWe of LWRs by 2025 and up to 75 GWe of new build LWRs and fast reactors by 2050. With the first fast reactor deployed in 2040 and the beginning of LWR reprocessing in 2040.

3.6.1 UK Plutonium

In the past there have been three long-term considerations for managing the UK's plutonium stockpile. Direct disposal a repository, use in a PWR as MOX before disposal, and long-term storage [16]. In 2011 the UK government opened a consultation on the UK's plutonium stockpile [17]. In response to this consultation, two extra plutonium management strategies were suggested. The use of plutonium in a PHWR (CANDU), and the use of plutonium in an SFR (PRISM) operated as a once-through, metallic fuelled, plutonium burning fast reactor [1, 2]. The NDA's position paper outlined plutonium re-use strategies as the preferred options as they reduce the attractiveness of the stockpile, putting it out of reach in a highly active matrix [1]. As yet a preferred option has not been selected by the UK government.

3.6.2 UK SFR closed fuel cycle studies

NNL, as part of studies for DECC, have considered large scale nuclear growth scenarios to improve energy security, reduce carbon emissions and improve long-term sustainability [127]. Results from this study were published as the closed fuel cycle scenario in the UK R&D roadmap technology pathways report [21]. This focused on a target of 75 GWe by 2050, transitioning from LWRs using UOX fuel to MOX fuelled fast reactors.

Transitioning to 75 GWe of SFRs would lead to an estimated 80% cut in CO_2 emission by 2050. The maximum fast reactor fleet was directly dependent on the number of thermal reactors operated. With a 5 year cooling time, fast reactor capacity would be 10-20% smaller than LWRs by 2050 and with a 2 year cooling time the SFR and LWR capacity would be equal.

The decay heat of spent fuel being reprocessed was assessed, 5 year cooled SFR MOX (88 GWd/t) was similar to 18 year cooled PWR MOX (55 GWd/t) and 2.6 times higher than 6 year cooled LWR UOX (55 GWd/t). Geological disposal in bentonite clay was considered, assessing the final repository size. The cumulative decay heat method was used, summing over 450 years. With one generation of SFRs, recycling only plutonium, the reduction in repository size was a factor of 1.8 compared to a once-through LWR fuel cycle. With the reprocessing of neptunium and americium, repository size was reduced by a factor of 2.5. Considering the reprocessing waste streams only, the repository size was reduced by a factor of 2.5 for plutonium recycling and 14 with neptunium and americium recycling.

3.7 Considerations for further study

This chapter has given an overview of previous UK and international fuel cycle studies. This section will highlight areas where further investigation may be relevant. The down selection of fuel cycle scenarios to study is discussed in Chapter 4.

The UK is currently looking at plutonium disposition routes. There is only one current fuel cycle study looking at fast reactor fuel cycles in the UK. This UK study focuses on a large growth of nuclear power with high CR SFRs. There are no publicly available studies looking at UK plutonium disposition with SFRs. Equally there are no studies looking at low CR SFRs in the UK for transmutation scenarios, or to reduce the UK's stockpile in a phase out scenario.

Internationally, few studies consider the reduction stockpiles. Studies which do consider stockpile reduction consider it in the context of nuclear phase out and the total reduction of a stockpile. These studies tend to use low TRL ADS reactors, or low TRL SFR fuel cycles with short cooling times and all TRUs recycled. Higher TRL SFR scenarios for stockpile reduction would be of interest, considering longer cooling times and no curium reprocessing. No studies have considered once-through SFRs as burners to reduce stockpiled material and how once-through SFRs compare to PWR MOX as a disposition method. Studies looking at several generations of fast reactors for maximum stockpile reduction do not consider the continued operation of LWRs, where SFRs could offset the number new build LWRs.

Results from the literature show that waste performance is dependent on reprocessing cooling time and the TRUs recycled. When the waste stream from reprocessing is considered as the only material sent to a repository, then reprocessing losses also have a significant impact on waste performance.

Reprocessing parameters that were used in this work were estimated from previous studies. Advanced reprocessing techniques have not been developed to a commercial scale for relevant operating experience to be used. Reprocessing parameters are summarised in Table 4.3.

Chapter 4

Methods

This chapter outlines the methods used in this thesis to compare the performance of potential UK waste disposition options, aimed at reducing the UK's nuclear waste inventory. Various fuel cycle options were tested, each representing a unique fuel cycle scenario to be modelled. Assessment of the relative performance of disposition methods was a multi-step process, which required:

1. Definition of UK waste disposition options and potential fuel cycle scenarios to study;
2. Estimate the mass and isotopic content of UK waste streams;
3. Design a reactor for waste disposition to meet the requirements outlined in (1);
4. Model the fuel cycle scenario outlined in (1);
5. Assess the fuel cycle's performance using fuel cycle assessment criteria;
6. Compare the performance of the various fuel cycle scenarios;
7. Use decision analysis methods to determine which fuel cycle scenarios merit further study.

The approach used for each step is presented in Table 4.1. This chapter outlines the software used to model reactors and fuel cycles, the fuel cycle modelling variables used and the procedures used for each of the following results chapters.

This research was designed to test the impact of fast reactor fuel cycles on the UK's nuclear waste inventory. The UK's nuclear waste inventory includes the UK's plutonium stockpile and SNF from new build reactors. The impact of fast reactor fuel cycles

Table 4.1: Key steps and procedures used in this thesis.

Step	**Approach**	**Software/ Technique used**	**Sec.**
Definition of UK waste disposition options and potential fuel cycle scenarios to model	Review literature on UK nuclear programme and previous UK fuel cycle studies	N/A	4.2
Estimate the mass and isotopic content of UK plutonium stockpile and waste streams from new build reactors	Model the build-up of stockpiled material, using UK reactor operating histories and reprocessing histories	ORION	4.3.1
Design a reactor for waste disposition to meet the requirements outlined in (1)	Optimised a reference reactor to meet the aims of a fuel cycle scenario	ERANOS Python	4.3.2
Model the fuel cycle scenarios outlined in (1)	Use fuel feed determined in (2) and the reactor designed in (3)	ORION	4.3.3
Assess each fuel cycle's performance using fuel cycle assessment criteria	Define assessment criteria and post-processed results from ORION to get required information	Python	4.3.4
Compare the performance of the various fuel cycle scenarios	Compare the relative performance of each fuel cycle scenario to each other and direct disposal option using the assessment criteria	Improvement factors	4.3.5
Use decision analysis methods to determine which fuel cycle scenarios merit further study	Normalised and weighted improvement factors for a number of test cases	Decision Analysis	Ch. 10

was determined based on assessment criteria, these are: power generated; lifetime of the fuel cycle; final inventory; radiotoxicity lifetime , radiotoxicity at 1000 years, assuming a disturbed repository scenario; repository size; and bare sphere critical mass of final material.

The procedures used to model and assess UK fast reactor fuel cycle scenarios are outlined below and discussed in more detail in Section 4.3. Figure 4.1 shows how parameters are used and how the procedures are linked to get results:

1. Fuel feed assessment – Determine the mass and isotopic vector of fissile material that would be used to fuel a reactor in UK fuel cycles. Fuel feeds were the UK's

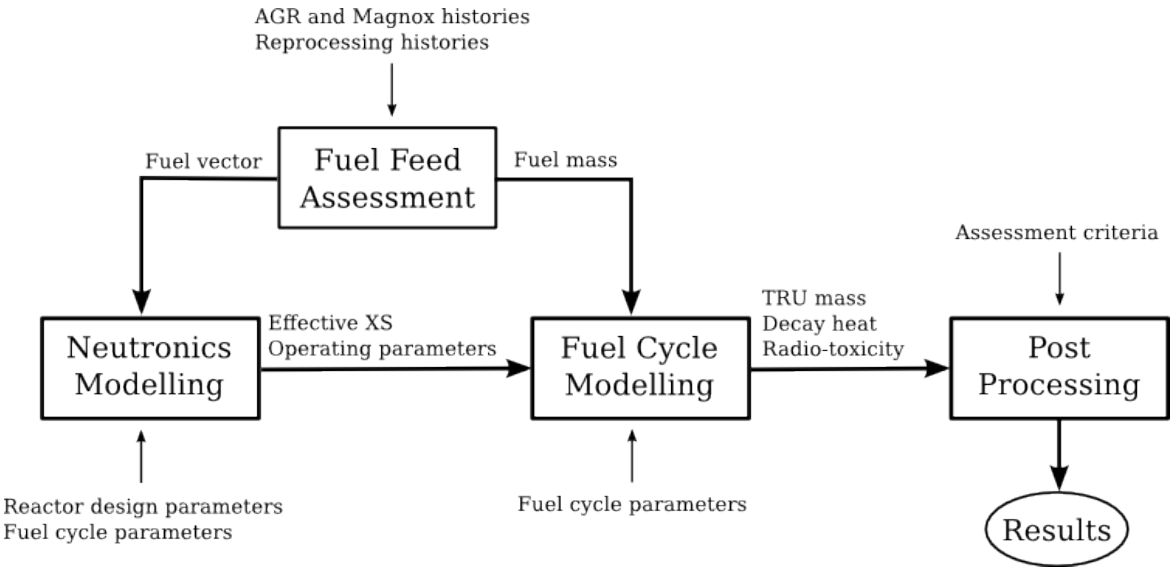


Figure 4.1: Methodology used for fuel cycle analysis.

separated plutonium stockpile and transuranics reprocessed from UK new build reactor SNF. Fuel feeds were modelled in ORION.

2. Fast reactor neutronics modelling – Use fuel feeds to design a reactor to match fuel cycle scenario aims, outlined in Section 4.2. ERANOS was used to perform k_{eff} and burnup calculations to optimise reactor design parameters to meet the aims of the fuel cycle, whilst ensuring that a reactor remains within feasible operating conditions, discussed in Section 4.3.2. The neutronics model was used to produce effective cross-sections which were used in fuel cycle models to simulate in-core depletion.

3. Fuel cycle modelling – Represent fuel cycle scenarios outlined in Section 4.2 with a set of fuel cycle parameters to calculate material streams and final inventories. ORION used the fuel feed, fuel cycle parameters and reactor design from previous steps. Each fuel cycle scenario had its own ORION model using different fuel cycle and reactor parameters.

4. Analysis of fuel cycle results – Post-processing of ORION streams and inventory results to get assessment criteria, discussed in Section 4.3.4. Assessment criteria were used to compare the performance of each fuel cycle scenario.

Decision analysis is discussed in Chapter 10. The purpose of decision analysis is for the user to apply weightings to the assessment criteria to determine the best fuel cycle scenarios for further study. Assessment criteria for all fuel cycle scenarios have

been represented as single value improvement factors, discussed in Section 4.3.4 and 4.3.5, which were normalised and used for decision analysis.

4.1 Modelling tools

The software used for neutronics and fuel cycle modelling are presented in this section to give an overview of the techniques used before discussing how the software was used to model UK waste disposition scenarios. Neutronics modelling was used to design fast reactors and export effective cross-sections to simulate in-core depletion in fuel cycle models. Fuel cycle models were used to simulate the fuel cycle and calculate the final waste inventory sent to a repository.

4.1.1 Fast-reactor neutronics code: ERANOS

For neutronics modelling the deterministic code European Reactor ANalysis Optimized code System (ERANOS) was used. One-group effective cross-sections were produced from neutronics modelling and used in fuel cycle models to represent a reactor over its lifetime, discussed in Section 4.1.2.1.

ERANOS2.0 has been developed as a validated deterministic code for reliable fast reactor neutronics calculations [137]. ERANOS comes with the JEF-2.2 multi-group cross-section library. All cross-sections come in broad 33-group and 172-group schemes, with the main resonance and scattering nuclei also represented in a fine 1968-group scheme. ERANOS uses separate calculation modules that are linked together using the LU scripting language. Modules of particular importance are: the cell lattice code (ECCO); flux solvers using diffusion and transport methods in 2D RZ and 3D hexagonal geometry (HEX-Z); the burnup module; and process modules which are used to output inventories, fluxes and conversion ratios.

The measurements of interest from ERANOS are k_{eff} and burnup fuel inventory. k_{eff} was used to ensure that reactor designs were feasible, based on shut down margins and burnup reactivity loss. Inventory results were used to simulate reprocessing, replacing the initial fuel vector with the new, reprocessed spent fuel vector. There are two main steps in modelling a reactor in ERANOS:

1. ECCO cell calculation – This requires cross-section data, material compositions,

fuel assembly (cell) geometry. ECCO has several calculation routes that account for resonance shielding and neutron slowing down to generate effective cross-sections for a cell.

2. Whole core flux calculation – The cross-sections from ECCO, as well as user defined core geometry and meshing, are entered into a flux solver which solves eigenvalue problem for the whole core to get k_{eff}.

4.1.1.1 Cell calculation

Effective cross-sections produced by ECCO are used for whole core calculations in ERANOS and to simulate in-core depletion in ORION fuel cycle models. As both reactor design and fuel cycle modelling depend on cross-sections from ECCO, it the most vital part of the neutronics calculation.

Two main factors must be accounted for in a fast reactor cell calculation: resonance self-shielding effects of heavy nuclei and elastic slowing down effect of light and intermediate nuclei (fuel matrix, coolant, and structural material) [138]. The thermalisation of neutrons can be ignored as it is assumed that neurons are absorbed or leaked from the core and do not reach thermal energies [27]. In the case of a homogeneous medium, condensed effective cross-sections are given by,

$$\sigma_g = \frac{\int_{E_g}^{E_{g-1}} dE\sigma(E)\phi(E)}{\int_{E_g}^{E_{g-1}} dE\phi(E)}. \tag{4.1}$$

Where g is the condensed energy group, σ_g is the effective cross-section in energy group g, $\sigma(E)$ is the initial cross-section as a function of energy, and $\phi(E)$ is the neutron flux.

Fine energy groups are required to represent the slowing down of neutrons in a fast reactor [138]. Elastic scattering from intermediate and light nuclei characterises the energy dependence of neutron flux in the cell. Scattering, based on two body Newtonian kinematics, is used to get a neutron slowing down flux [139]. Without absorption the flux spectrum is proportional to $1/E$, with the width of the flux spectrum being dependent on the amount of scattering [27].

Resonance self-shielding uses the sub-group method. The energy group structure used in ECCO is not fine enough to account for the shape of the resonances so probability tables are used to represent the sub-group resonance structure [138, 140]. The

impact of the resonances on neutron flux is dependent on the background scattering cross-section,

$$\sigma_0 = \frac{N_s \sigma_{s,s}}{N_r}, \tag{4.2}$$

where $\sigma_{s,s}$ is the total cross-section of the scattering nuclei and N_s and N_r are the number densities of scattering an resonant nuclei. If the background scattering cross-section is large then the neutron flux will look like the slowing down spectrum, as scattering dominates and resonance absorption is not significant enough to influence neutron flux. As the impact of resonances on g flux spectrum is dependent on background cross-sections, sub-group probability tables are given for a range of typical background cross-sections.

Equation 4.1, used for effective cross-sections can also be used to describe the sub-group contributions which accounts for resonance self-shielding. However, the equation needs to be re-written as a function of cross-section and a probability table $p(\sigma)$ [141],

$$\sigma_g = \frac{\int_{E_g}^{E_{g-1}} dE\sigma(E)\phi(E)}{\int_{E_g}^{E_{g-1}} dE\phi(E)} = \frac{\int_{\sigma_{min,g}}^{\sigma_{max,g}} d\sigma p(\sigma)\sigma\phi(\sigma)}{\int_{\sigma_{min,g}}^{\sigma_{max,g}} d\sigma p(\sigma)\phi(\sigma)}. \tag{4.3}$$

Where $p(\sigma)$ is the probability distribution of cross-section in the energy group E_g to E_{g-1}.

4.1.1.2 Whole core calculation

Condensed effective cross-sections from ECCO are used for whole core flux calculations. Whole core-calculations were used in this thesis to ensure that a reactor design would operate within a range of feasible k_{eff} values, determined in Section 4.3.2.2. The flux profile from the whole core calculation was also used to normalise ECCO fluxes, to get accurate condensed cross-sections for ORION. The level of detail required for k_{eff} calculations and flux normalisation is low, allowing for the use of diffusion flux solvers in ERANOS. For most fast reactor calculations diffusion theory is adequate to solve the multi-group eigenvalue problem [27],

$$\begin{aligned} &\text{Leakage} + \text{Removal} = \text{In Scattering} + \text{Fission}, \\ &-D_g\nabla^2\phi_g + \Sigma_g\phi_g = \sum_{h=1}^{g-1}\Sigma_{s,h\to g}\phi_h + \frac{\chi_g}{k_{eff}}\sum_{h=1}^{G}\nu\Sigma_{f,h}\phi_h. \end{aligned} \tag{4.4}$$

Where $-D_g\nabla^2\phi_g$ is the rate of leakage from the reactor of neutrons in group g, $\Sigma_g\phi_g$ is the total reaction rate for neutrons removed from group g due to scattering or

absorption, $\Sigma_{s,h\to g}\phi_h$ is the rate of scattering from group h into group g, χ_g is the number of fission neurons produced in group g, k_{eff} is the neutron multiplication factor and $\nu\Sigma_{f,h}\phi_h$ is the rate of fission at energy h multiplied by the number of neutrons produced per fission.

Core geometry is meshed and the volume averaged eigenvalue problem is solved for all mesh points [142]. As the flux distribution is dependent on the solution of the eigenvalue problem, a first guess is made and the problem is solved iteratively until results converge on a value of k_{eff}.

4.1.2 Fuel cycle modelling code: ORION

Fuel cycle modelling codes represent the fuel cycle as material buffers and facilities with streams connecting them. Buffers can represent storage, a geological repository, or initial materials such as a plutonium stockpile. Facilities perform operations on material streams such as cooling, reprocessing, fuel fabrication or in-core depletion. Buffers and facilities are connected by streams, allowing material to flow between them. Fuel cycle codes model the movement of all isotopes and their decay in broad time steps, calculating the inventory of buffers and material streams at each time step.

ORION, which was developed at NNL [143], was used for fuel cycle modelling in this thesis. ORION tracks 2552 nuclides over time steps of one month to one year. Reactor build rates can be user defined, or dynamically set based on the build-up of fissile materiel in the fuel cycle. If fissile fuel feeds vary over the fuel cycle, ORION can change the enrichment of fuel based on reactivity equivalence coefficients. Final inventories can be tracked to any year after the fuel cycle model has ended, calculating the build-up and loss of all isotopes due to decay. Buffers and streams can be represented in terms of mass, activity, radiotoxicity, decay heat, and spontaneous neutron emissions. For radiotoxicity, effective dose coefficients are taken from taken from ICRP-68 [144].

An example of an ORION closed fuel cycle model and the graphical user interface is shown in Figure 4.2. Streams are the numbered green lines and the types of buffers and facilities are shown in the red box, described as:

- Feed – Specifies annual isotopic masses to be fed to a buffer, representing a

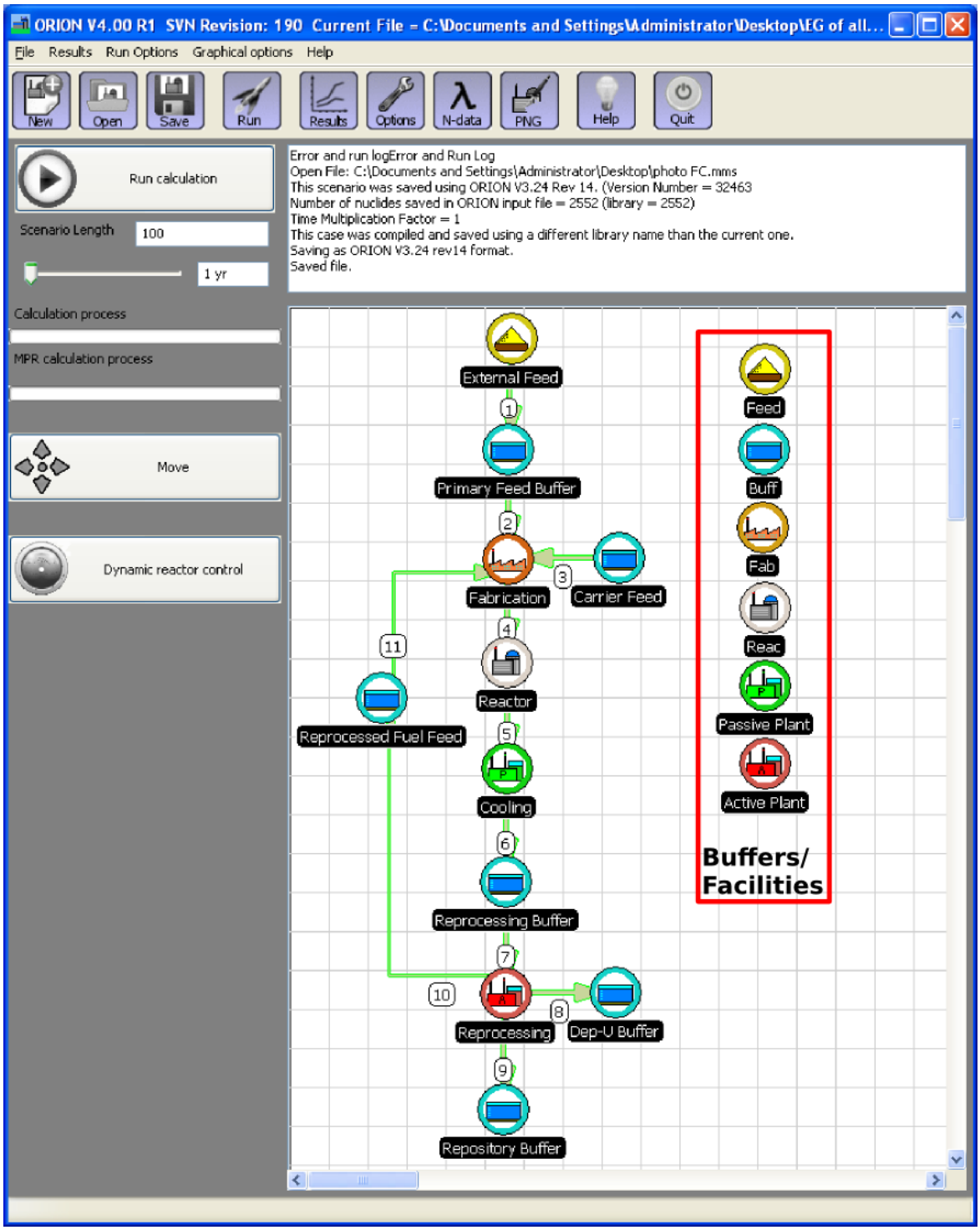


Figure 4.2: ORION graphical user interface with an example of a closed fuel cycle. Streams are the numbered green lines, and the types of buffers and facilities are shown in the red box [143].

known external feed such as plutonium from a reprocessing plant.

- Buffer – Used to store material. Buffers can be initialised with material, for example a natural uranium buffer could be used to represent a uranium mine. Buffers are also used to represent a repository (e.g. "Repository Buffer") and before a facility to represent interim storage before there is a demand for the buffered material (e.g. "Reprocessing Buffer" and "Reprocessed Fuel Feed").
- Fabrication – Fabricates uranium or plutonium fuel for a reactor. Uranium fuel fabrication takes a uranium stream and enriches it to the level required for the reactor, sending enrichment tails to a buffer. Plutonium fuel specifies a fissile feed and a fertile carrier feed, mixing the feeds to an enrichment specified by the reactor. The annual throughput of a fabrication facility is dictated by reactor demand, but the hold up time for the fabrication process is specified by the user.
- Reactor – Discussed in Section 4.1.2.1.
- Active plant – Represents reprocessing. Active plants take an input stream and separates isotopes across different output streams. The user specifies a peak annual throughput and hold up time for the process.
- Passive plant – Used to represent cooling ponds or to move material between buffers. Passive plants have a peak annual throughput and hold up time for the process. Unlike active plants materials are not separated in passive plants.

The example fuel cycle in Figure 4.2 uses an 'External Feed' mixed with a 'Carrier Feed' at the 'Fabrication' step, to generate fuel for the 'Reactor'. The 'Reactor' irradiates the fuel by performing depletion calculations and spent fuel is cooled at the 'Cooling' stage before being reprocessed at the 'Reprocessing' stage. The 'Reprocessing' stage separates fuel material from the spent fuel. Fuel material is sent to the 'Reprocessing Fuel Feed' buffer to be fabricated as new fuel, and all other materials are sent the 'Repository Buffer'.

4.1.2.1 Reactors

Reactors are defined in ORION by several static parameters: electrical power, thermal power, heavy metal inventory, enrichment, operational lifetime, capacity factor, cycle

length, fuel density, and power density. In addition to these static parameters, in-core depletion is modelled using tabulated spent fuel compositions; or using effective cross-sections produced by a reactor neutronics model.

This section will describe the use of effective cross-sections for in-core depletion calculations. A cross-section file for a fuel region contains one group microscopic cross-sections averaged over the fuel region. Cross-sections were generated using reactor neutronics models, with some post-processing required to condense and format the cross-sections. Post-processing uses utility codes written by NNL [143].

ERANOS was used to model SFRs and produce neutron fluxes and effective, self-shielded cross-sections for ORION. Post-processing utilities condense the cross-section data to one group using the flux data,

$$\sigma_{xm} = \frac{\int \sigma_{gxm}(E)\phi_g(E)dE}{\int \phi_g(E)dE}. \tag{4.5}$$

Where σ_{gxm} is the effective cross-section for reaction x of nuclei m in energy group g; ϕ_g is the average neutron flux in energy group g; and σ_{xm} is the condensed effective cross-section, also called the reaction rate.

ORION tracks more isotopes than ERANOS, missing isotope data is taken from the EAF-2007 library [145] of infinitely diluted cross-sections. It is assumed that isotopes that are not modelled in ERANOS are present in low enough quantities to not have a significant self-shielding effect. If there is no self-shielding then, the use of infinitely diluted cross-sections is valid. EAF-2007 cross-sections are condensed to one group using ERANOS flux data.

Reactor flux is calculated in ORION from the user defined power density and a list of MeV per fission for each of the key nuclides. Depletion calculations take place over the ORION time-step using the flux (ϕ), reaction rates (σ_{rm}), decay constants (λ_m), and atomic densities of each nuclei (N_m) in the fuel,

$$\begin{aligned}\frac{dN_k}{dt} = &\overbrace{\phi \sum_{i=l}^{m} N_i \sigma_{fi} y_{i\to k}}^{\text{Produced from fission}} + \overbrace{\phi \sum_{j=l}^{m} N_j \sigma_{cj} \gamma_{j\to k}}^{\text{Produced from capture}} + \overbrace{\phi \sum_{z=l}^{m} N_z \lambda_z \alpha_{z\to k}}^{\text{Produced from decay}} \\ &- \underbrace{\lambda_k N_k}_{\text{Loss to decay}} - \underbrace{\phi N_k \sigma_{ak}}_{\text{Loss to absorption}}.\end{aligned} \tag{4.6}$$

Where σ_{fi} is the fission cross-section of nuclei i; $y_{i\to k}$ is the number of nuclei k produced per fission of nuclei i; σ_{cj} is the capture cross-section of nuclei j; $\gamma_{j\to k}$ is the number

of nuclei k produce per capture in nuclei j; $\alpha_{z\to k}$ is the number of nuclei k produced per decay of nuclei z; and σ_{ak} is the cross-section for absorption in nuclei k.

4.2 Fuel cycle scenario definitions

This section outlines reactor and fuel cycle parameters tested in the fuel cycle studies. Parameters were selected to cover a broad range of fuel cycle options. These range from the most realistic, high TRL parameters, to those which have lower TRLs but are expected to perform better against the assessment criteria described in Section 4.3.4.

There are two key areas in the future of the UK's nuclear fuel cycle, that give two potential fuel streams for fast reactor fuel cycle scenarios: (1) The UK's plutonium stockpile; (2) The UK's plutonium stockpile and SNF from new build LWRs. Fuel cycle scenarios that use these two feeds will be considered separately as they represent different UK situations and require different deployment levels for fast reactors.

NNL has investigated a large build scenarios as part of the UK R&D pathways [21]. The UK R&D pathways are the only up to date UK fuel cycle studies in the literature. The closed fuel cycle pathway transitioned from LWRs to sustainable SFRs with a CR $\geqslant 1$. As high CR SFR fuel cycles have already been considered in a UK scenario, they were excluded as potential option for this study. The aim of low-CR fast reactor fuel cycles is to reduce stockpiled material inventories.There are three levels of low CR fast reactor deployment that are of interest:

1. Once-through BURNER – Operating once-through SFRs to irradiate all stockpiled material without any further reprocessing. This is the scenario that has been suggested for UK plutonium disposition with PRISM.
2. FEED equilibrium scenario – Operating a small number of SFRs with reprocessing for one to two generations, until all the stockpile has been irradiated once. The FEED scenario aims to assess the impact of fast reactors in a closed fuel cycle over a short timescale.
3. FULL equilibrium scenario – Operating as many SFRs as possible given the stockpiled material available, for several generations to get the maximum reduction in stockpile material. Each generation of SFRs will operate less SFRs as

Table 4.2: Potential fuel cycles scenarios to study. Outlining reactor design goals, fuel feed (Pu stockpile with and without new build LWRs), fuel cycle options, reactor build rates (years / reactor), and fuel cycle assessment date in the model (some are To Be Evaluated (TBE)).

CR	Feed	Fuel Cycle	Build Rate	Ass. Year	Info
Lo	Pu	BURNER	5	2100	PRISM-type scenario
Lo	Pu	FEED	5	2150	PRISM /w recycling to increase impact
Lo	Pu	FULL	1	TBE	Max inventory reduction
Hi	Pu	BURNER	5	2100	PRISM with high U efficiency
Hi	Pu	FEED	5	2150	(Similar to NNL) Increase U efficiency
Hi	Pu	FULL	1	TBE	(Similar to NNL) Transition to FR
Lo	Pu+LWR	BURNER	5	2150	PRISM-type scenario
Lo	Pu+LWR	FEED	5	2200	PRISM /w recycling to increase impact
Lo	Pu+LWR	FULL	1	TBE	Max inventory reduction
Hi	Pu+LWR	BURNER	5	2150	PRISM with high U efficiency
Hi	Pu+LWR	FEED	5	2200	(Similar to NNL) Increase U efficiency
Hi	Pu+LWR	FULL	1	TBE	(Similar to NNL) Transition to FR

the mass of fuel in the fuel cycle reduces.

Table 4.2 outlines all possible combinations of fuel cycle studies. Most of the high CR scenarios overlap with NNL scenario studies so they are not considered, leaving eight possible scenarios. Each of the fuel cycle scenarios in Table 4.2 will have several sub-scenarios based on reactor and fuel cycle variables discussed in the following section.

4.2.1 Reactor and fuel cycle parameters

An overview of fuel cycle parameters from the literature can be found in Section 3.5. Parameters were selected for fuel cycle studies based on those which would have the greatest impact on final inventories and those which had the highest TRL. Overall sixty SFR fuel cycles were modelled: eight BURNER fuel cycle scenarios using the UK's plutonium stockpile in Chapter 7; twelve FEED scenarios and twelve FULL scenarios using the UK's plutonium stockpile in Chapter 8; four BURNER scenarios, twelve FEED scenarios and twelve FULL scenarios using the UK's plutonium stockpile and SNF from new build LWRs in Chapter 9. The different parameters tested in each sub-scenario are outlined in this section.

Table 4.3: Closed fuel cycle reprocessing parameters used for sub-scenarios.

	LO-PU	**LO-AM**	**LO-TRU**	**HI-PU**	**HI-AM**	**HI-TRU**
Cooling (y)	1	1	1	10	10	10
Fab+Rep (y)	1	1	1	2	2	2
Losses (%)	0.1 Pu	0.1 Pu	0.1 Pu	0.2 Pu	0.2 Pu	0.2 Pu
		0.1 Am	0.1 Np		1.0 Am	1.0 Np
			0.1 Am			1.0 Am
			0.1 Cm			5.0 Cm

SFRs were deployed in 2040 and built at a rate of one module per year in FULL scenarios and one module every five years for BURNER and FEED scenarios, see Table 4.2. BURNER and FEED scenarios were assessed at 2100–2200, and FULL scenarios were assessed when the stockpile was reduced to a point where no further reactors could be operated.

Each of the initial fuel feeds were modelled in BURNER, FEED and FULL scenarios with a range of sub-scenarios testing different reactor and fuel cycle parameters. Testing a range of reactor and fuel cycle parameters allowed the merits of different technologies to be compared relative to their TRL. Preliminary sensitivity studies determined the combined effect of cooling time and reprocessing losses. Long-cooling times and high reprocessing losses pushed results in the same direction. As such, high reprocessing losses and long cooling time were combined (HIGH scenarios), and low reprocessing losses and short cooling times were combined (LOW scenarios). HIGH and LOW fuel cycle parameters are compared in Table 4.3.

A total of twelve sub-scenarios were outlined for FEED and FULL scenarios, using:

- Three reprocessed fuel feeds – Reprocessing of plutonium (PU scenarios), plutonium and americium (AM scenario), or all TRUs (TRU scenario).
- Two reprocessing parameters – Long cooling times and high losses (HI scenario), or low cooling times and low losses (LO scenario), Table 4.3.
- Two reactor fuels – Metallic (U-TRU-10Zr) and MOX fuelled reactors were designed and tested for each scenario. Two fuels were compared to see how the harder flux spectrum and higher fuel density of U-TRU-10Zr metallic fuels compare to higher TRL MOX fuels. Reactor design will be discussed further in Section 4.3.2.

A total of four[1] sub-scenarios were outlined for once-through BURNER scenarios using:

- Two fuel feeds – AM scenarios which had americium in-growth included in the fuel, and PU scenarios which had americium in-growth removed before fuel fabrication.
- Two reactor fuels – Metallic (U-TRU-10Zr) and MOX fuelled reactors were designed and tested for each scenario.

4.3 Procedures

The procedure for UK fast reactor fuel cycle analysis had four key components which were outlined at the beginning of this chapter, Figure 4.1, and described in detail in the following sections.

4.3.1 Fuel feed assessment

Fuel feed assessment is the process of determining the mass and isotopic content of stockpiled material that will be used in fuel cycle scenarios. Two feeds were required for the fuel cycle scenarios outlined for study in Section 4.2, the UK's plutonium stockpile and SNF from new build LWRs. There is no public information on the isotopic content of the UK's plutonium stockpile or new build SNF. Fuel feeds were assessed in Chapter 5 using ORION models so the final results could be used as a direct feed in further fuel cycle scenario modelling. Data used in the fuel feed models came from the open literature and Freedom of Information (FOI) requests. This section describes a brief overview of the methods used to assess the fuel feeds, with Chapter 5 containing more detail on the sources of data, assumptions made and limitation.

Plutonium stockpile The majority of the UK's plutonium stockpile is reprocessed Magnox and AGR fuel. The build-up of spent fuel from the Magnox and AGR fleet was required, along with the history of Magnox and AGR fuel reprocessing. Data on UK reactor operating histories, average fuel burnup, reactor inventory, and spent fuel compositions were used to determine the annual Magnox and AGR SNF generated,

[1] In Chapter 7 there were eight BURNER scenarios as high CR BURNER fuel cycle scenarios were modelled as well as low CR BURNER scenarios in the initial scoping of this study.

which was used as a feed in ORION. The reprocessing histories of B205 and THORP reprocessing plants were used to model the buffering of SNF prior to reprocessing and the build-up of separated plutonium in the UK. It was possible to compare plutonium stockpile results to ONR data published annually on the mass of the UK's plutonium stockpile, as well as estimates for the final stockpile size at the end of reprocessing. However, it was not possible to check the isotopic content of the stockpile as this information is not publicly available.

New build LWR New build SNF was estimated based on the UK's current goal of 16 GWe by 2050, with the first PWR coming online in 2023. New build plans are likely to change, as such there is no guarantee of build rates, lifetime of reactors, capacity factor or final generating capacities. With so much uncertainty a relatively simple approximation for new build was used, operating 1.65 GWe PWRs with an average burnup of 48 GWd/t and a constant build rate from 2023 to 2050.

4.3.2 Fast reactor neutronics modelling

ERANOS was used to design and model reactors for each fuel cycle scenario. A feasible range of design and operating parameters is outlined in Section 4.3.2.2. The design process for reactors had several stages:

1. Benchmarking and validation of ERANOS neutronics modelling techniques;
2. Defining a reference reactor design based on UK fuel cycle aims;
3. Develop a script to optimised reference reactor design by adjusting fuel volume and enrichment parameters;
4. Develop a script to run a reactors with reprocessing to an equilibrium point;
5. Find an optimum design of a reactor for each fuel cycle scenario.
6. Extract cross-sections and design data for use in ORION.

In addition to these stages, sensitivity studies were performed, Appendix C, to determine appropriate mesh size, burnup steps and burnable zone sizes to get accurate results and fast running simulations.

4.3.2.1 Benchmarking and validation

ERANOS2.0 was tested against an IAEA benchmark for deterministic fast reactor codes [146], comparing results to CEA, using ERANOS2.2, and other deterministic codes. A reference, PRISM-type 1 GWth SFR design was tested in ERANOS, with a range of different solution methods, and compared against the SERPENT Monte-Carlo code to show that results were comparable across codes.

4.3.2.2 Reference reactor

It was important to consider a fast reactor design that was applicable to the UK situation and flexible in terms of design to cope with a range of fuel cycle scenarios. A small PRISM-type reactor was selected as PRISM has been suggested for UK plutonium disposition. There are many adaptations of the PRISM concept, which use oxide or metallic fuels as well as a range of conversion ratios. This is ideally suited to this study and allows for metallic (ZR) and MOX fuelled reactors to be compared, to determine whether there are any advantages in developing metallic fuelled reactors over higher TRL MOX fuelled reactors.

The exact design specification of a PRISM reactors was not available so a variant on the PRISM reactor was developed based on two reactors, the 840 MWth low-CR PRISM-UNF design [147] and the 1 GWth Advanced Burner Reactor (ABR) design, which is based on PRISM [81, 148]. Reactors were designed for UK fuel feeds and fuel cycle goals which met key design constraints described below. The design process used a reference 1 GWth design and varied fuel pin radius, enrichment and the size of the inner and outer fuel regions.

Neutronics design parameters Preliminary ABR designs had three enrichment zones with a range of conversion ratios [81]. Without breeder blankets the maximum CR was ~ 1. With limitations on enrichment taken as 40 wt.% for oxide fuel and 30 wt.% for U-Pu-10Zr metallic fuel, the lowest CR for the ABR study was ~ 0.75 [81]. In benchmark studies of the ABR, a CR of 0.7 was selected with two enrichment zones for the metallic fuelled core, similar to the PRISM UNF design [148].

As the fuel feed and reactor aims of a UK scenarios are different to previous studies, it was necessary to optimise reactor designs for UK scenarios. Reasonable limitations

Table 4.4: Parameters used for neutronics reactor design [79, 81, 117, 148].

Parameter	**Oxide**	**Metal**
Average fuel temp (°C)	1027	534
Structure/Coolant temp (°C)	432.5	432.5
Core Height (cm)	114.9	85.5
Core Diam (cm)	218	218
Fuel Vol (%)	< 50	< 50
Fuel Dens (g/cc)	10.97-11.46	15.7
Smear density (%)	85	75
Fuel Matrix	1.98 O	10 Zr
Peak Enrich (%)	< 40	< 30
I/O enrich ratio	1.25	1.25
k_{BOC}	<1.08	<1.08
Δk_{eff} (pcm)	3500	3500
Peak Linear (kW/m)	< 33	< 40
Power Peaking	< 1.7	< 1.7
Burnup (GWd/t)	100	100

on fuel design were taken as the boundary conditions for reactor design. Fuel design limits were based on previous reactor designs, summarised in Section 2.3.4.

The end requirement for reactor design was cross-sections for ORION, so some simplifications were made to make oxide and metallic cores more comparable. The average burnup was set to 100 GWd/t, and both reactors were limited to two enrichment zones.

Reactor design and operating parameters selected are presented in Table 4.4. Most parameters were taken from previous studies of a PRISM-type reactor, summarised in Table 2.3. Specifically, a maximum k_{eff} of 1.08 was taken from previous studies [79] and validated against neutronic assessment of the control rod worth. A maximum enrichment of 30% was chosen based the highest enrichment U-Pu-10Zr fuel successfully tested in the U-Pu-10Zr development programme [149, 150]. A maximum enrichment of 40 wt.% was chosen for MOX fuel based on reprocessing limitations. Aqueous reprocessing limits MOX to 45 wt.% plutonium [89, 106] and the CAPRA-CADRA programme envisaged enrichments between 40-45 wt.% [151].

4.3.2.3 Script for optimum reactor design

Python was used to run a range of quick, low precision ERANOS models for initial design scoping. Fuel volume and enrichment were optimised to achieve the lowest or

highest CR whilst keeping the design parameters within the allowable range outlined in Section 4.3.2.2. A python script, using the routine shown in Figure 4.3, tested reactor designs and tabulated design options which operated within the allowable parameters in Table 4.4.

Methods were investigated to reduce the run time of reactor optimisation and equilibrium calculation times, Appendix C. Batch-wise reactor refuelling was not used, instead the whole-core was irradiated from 0 GWd/t to an average burnup of 100 GWd/t to reduce the number of ECCO calculation step. Without batch-wise refuelling the BOC and EOC points were assumed to be where the average whole-core burnup reached the same point as the average batch-wise burnup at the BOC and EOC. This assumed that the reactor flux profile does not change significantly from 0 GWd/t to 100 GWd/t, which it does not.

4.3.2.4 Script for equilibrium reactor design

In a closed fuel cycle scenario a recycling equilibrium point can be reached. At equilibrium every subsequent recycling step has the same spent fuel composition. During fuel recycling the fuel feed vector degrades, requiring an increase in enrichment until an equilibrium point is reached. For fuel cycle modelling, an equilibrium reactor needs to be designed to ensure the peak allowable fuel enrichment is not exceeded.

A sub-routine was added to the script for optimum reactor design, described in Section 4.3.2.3, where the equilibrium sub-routine was run instead of an ERANOS burnup calculation. The equilibrium sub-routine ran 30 recycling steps to ensure a reactor was close to its equilibrium point. If the reactor exceeded any of the design limitations, the equilibrium sub-routine would exit and a new reactor design would be tested. Two methods of fuel recycling were implemented to reach two different equilibrium points,

- FEED equilibrium – Depleted fuel was cooled, reprocessed elements were extracted and used as the fresh fuel. Any reduction in fuel mass was replaced with stockpile/feed material.
- FULL equilibrium – Depleted fuel vector was cooled, reprocessed elements were extracted and used as fresh fuel. The depleted fuel vector was scaled up with no addition of feed material.

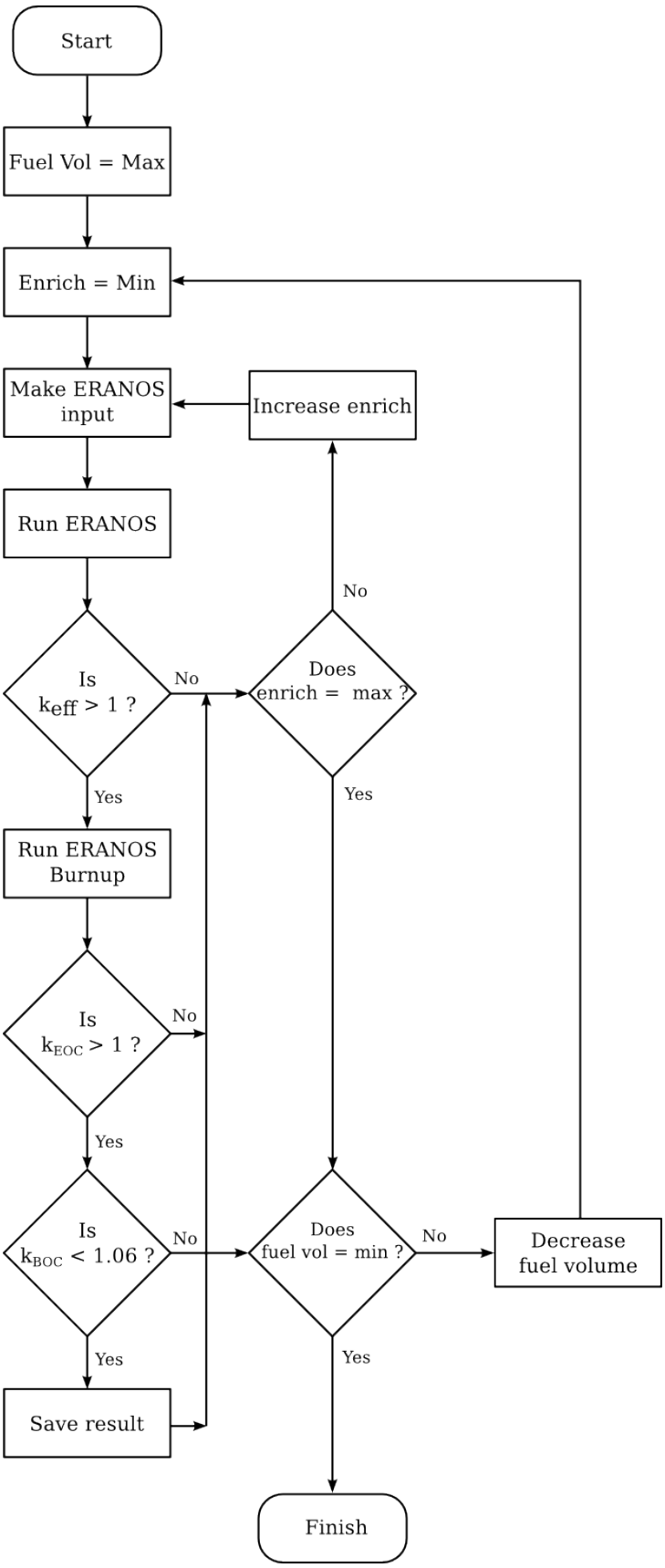


Figure 4.3: Python routine used for reactor design scoping in ERANOS.

Two methods were considered to account for the required change in fuel enrichment as a result of fuel feed degradation: first by adjusting enrichment based on equivalence coefficient calculations; and second by choosing the final enrichment and keeping enrichment fixed for recycling steps, assuming the same equilibrium point is reached. The fixed and variable enrichment methods were tested to show that the same equilibrium point was reached, Appendix C. The fixed method was chosen as the optimisation method was quicker. The present author was able to select a narrow range of enrichments to test based on previous reactor results, reducing the number of parameters tested in the optimisation routine.

4.3.3 Fuel cycle modelling

ORION was used to model the fuel cycle scenarios in this work. Initial fuel feeds, described in Section 4.3.1, were used with reactor parameters determined from the neutronics design, described in Section 4.3.2. There were three types of fuel cycle scenario, once-through BURNERS, FEED equilibrium scenarios and FULL equilibrium scenarios. Several sub-scenarios were tested for each of the BURNER, FEED and FULL scenarios, based on different reactor and fuel cycle parameters described in Section 4.2.

4.3.3.1 ORION models

Figure 4.4 shows an example of a closed SFR fuel cycle scenario used in this study, modelled in ORION. ORION once-through BURNER models are discussed in more detail in Chapter 7, FEED and FULL equilibrium models are discussed in more detail in Chapter 8.

In Figure 4.4 the UK plutonium stockpile and new build LWR SNF fuel feeds (described in Section 4.3.1) were used to generate representative fuel feeds in the 'Pu Stock' and 'LWR Feed,' buffers. Fuel was fabricated using uranium enrichment tails, 'U Tails' as the carrier feed. Fuel was fabricated for the inner and outer reactor regions and irradiated in 'SFR IF' and 'SFR OF' using reactor parameters and cross-section from ERANOS (Section 4.3.2). For BURNER scenarios spent fuel was sent to a repository buffer. For FEED and FULL equilibrium scenarios, spent fuel was reprocessed and recycled fuel material is sent to the 'Rep Feed' buffer to fabricate new

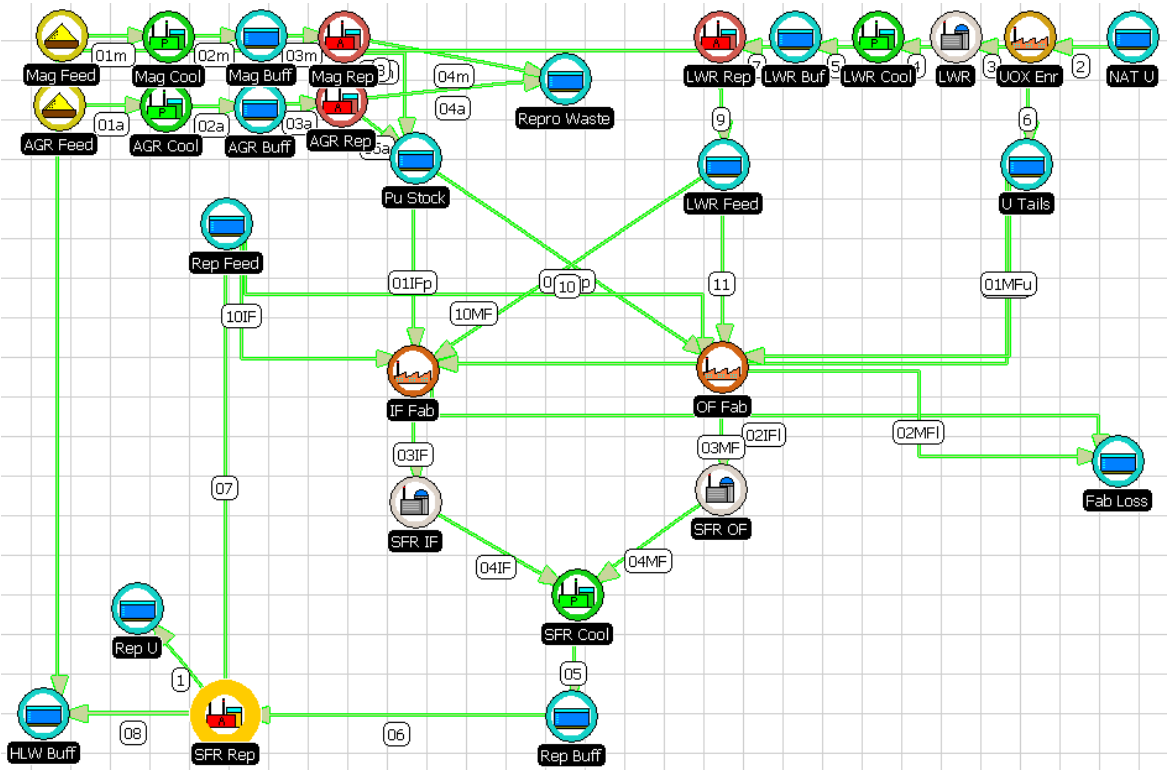


Figure 4.4: Closed SFR fuel cycle in ORION, using the UK's plutonium stockpile and SNF from new build LWRs as fuel feeds.

fuel.

For BURNER and FEED scenarios ORION was used to model a suitable number of reactors to irradiate all stockpiled material by the assessment date of the fuel cycle. The assessment date of FULL scenarios was dependent on how long it took to reduce the stockpile to the point where it is too small to operate any further reactors. A comparison of the FEED and FULL build scenario and the impact they have on fuel cycle inventory can be seen in Figure 4.5.

Direct disposal and once-through LWRs Stockpiled material was also modelled to represent direct disposal and used as a reference case to compare fuel cycle scenarios. UOX fuelled LWRs were also modelled, to compare SFR scenarios to once-through LWR scenarios with the same generating capacity.

PWR MOX Cross-section and fluxes for a sample PWR MOX assembly were used in ORION to estimate the total electricity that could be generated in a PWR with stockpiled material. Whilst it is understood that a 100% MOX fuelled PWR is unlikely, the ORION model estimated the total electricity generated by MOX fuel assemblies

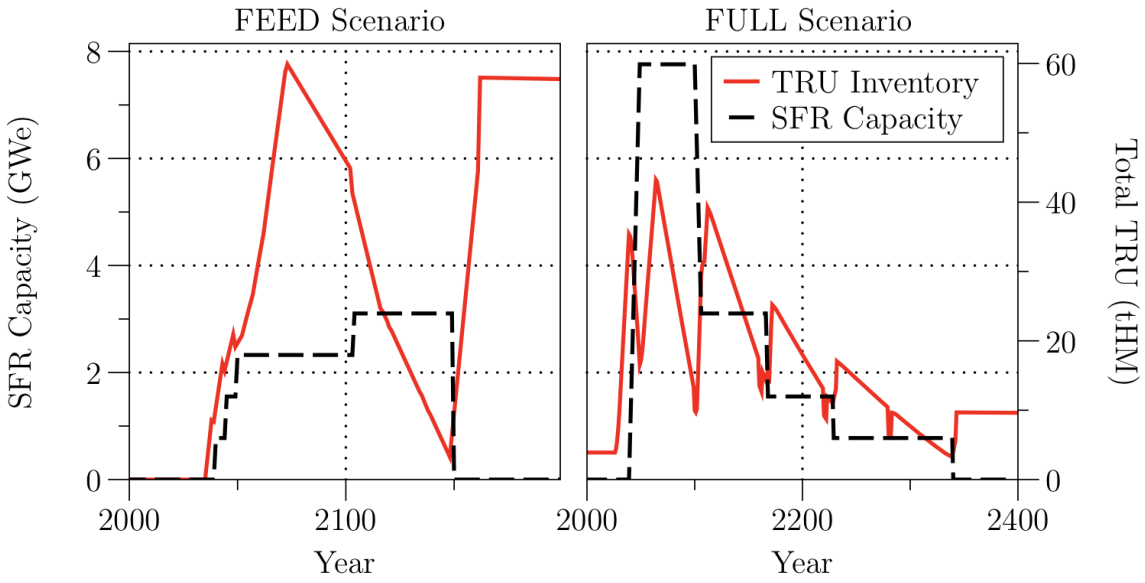


Figure 4.5: Build-up of SFRs in an example FEED and FULL fuel cycle. Showing the build-up of SFRs and the impact on transuranics buffered in the fuel cycle. (Note that transuranics build up from 2040, due to a separation step removing americium from the plutonium)

in PWRs. In plutonium stockpile BURNER scenarios, a PWR MOX scenario was compared to SFR scenarios so that three of the UK's options for plutonium disposition could be compared: direct disposal, PWR MOX, and irradiation in a PRISM-type SFR.

Americium in-growth With the in-growth of ^{241}Am in stockpiled material, due to the decay of ^{241}Pu, plutonium only fuel cycles (PU) required an additional americium removal step prior to fuel fabrication. An active plant was used prior to fuel fabrication to separate americium from plutonium, sending americium to a repository.

WASTE stream only results Many fuel cycle studies, discussed in Chapter 3, only consider the reprocessing waste stream as material which is sent to a repository. This assumes that material left in the fuel cycle is not sent to a repository and will be utilised in the future. Whilst WASTE stream results are unrealistic, it is worth considering so that results can be compared to previous studies. WASTE stream only results can also be used to show how the best case, FULL scenario results differ from the overly optimistic WASTE stream only results.

4.3.4 Assessment criteria

Fuel cycles were assessed based on the following performance factors, relative to a reference case:

- Total electricity generated, GWy(e), relative to a once-through MOX fuelled PWR;
- Lifetime of the fuel cycle, relative to MOX fuelled PWR;
- Final TRU fuel cycle inventory sent to a repository, relative to the initial stockpile;
- Lifetime of radiotoxicity, relative to a UOX fuelled PWR;
- Total radiotoxicity 1000 years after disposal, assuming a disturbed repository scenario, relative to the initial stockpile and an open LWR fuel cycle;
- Repository size, relative to the initial stockpile and once-through UOX fuelled PWR;
- Bare sphere critical mass of waste stream sent to a repository, relative to the initial stockpile.

Final inventory, generating capacity, total radiotoxicity and decay heat can all be exported from ORION but post-processing was required for some of the assessment criteria, described below.

For 1000 year radiotoxicity and repository size, results were given relative to two reference scenarios: the direct disposal of the initial stockpiled material; and relative to an open LWR fuel cycle with the same generating capacity as the SFR scenario and the direct disposal of the stockpile. Comparing assessment criteria to the stockpile only represents a fuel cycle scenario where the stockpile is being reduced and any electricity generated is merely a by-product. Comparing results to the stockpile plus open fuel cycle LWRs, represents a fuel cycle where the operation of SFRs as an alternative to LWRs, offsetting the number of LWRs required in the fuel cycle to meet the same generating capacity. In this latter case,ssssssss ORION is only modelling the fast reactor park.

4.3.4.1 Electricity generated

SFR electricity generation was compared to a MOX PWR scenario to see how electricity generated compared to a high TRL, once-through PWR MOX scenario.

4.3.4.2 Final transuranic mass

Final TRU inventory was tabulated after 50 years of cooling. This was reported as the final mass of plutonium, neptunium, americium and curium and compared to the direct disposal of the stockpile to determine the reduction in stockpile size.

4.3.4.3 Radiotoxicity lifetime

The radiotoxicity lifetime is considered to be the time it takes for SNF to decay to the same level as natural uranium used to fuel a PWR generating the same amount of electricity. Natural uranium radiotoxicity is considered a safe level, as it is the raw material being mined and used to fuel a reactor. Radiotoxicity lifetime was used to indicate the inter-generational liability of nuclear waste. Radiotoxicity per GWy(e) was plotted relative to the radiotoxicity of natural uranium for one million years. A UOX fuelled PWR with average burnup of 48 GWd/t was also plotted as a reference case to see how much shorter the lifetime of radiotoxicity was for SFR scenarios. A significant reduction in the lifetime of waste was considered to be an order of magnitude less than a UOX fuelled PWR, that is less than 29,000 years.

There are limitations to this method of estimating the radiotoxicity lifetime. Firstly, the total radiotoxicity of natural uranium should be the radiotoxicity of natural uranium used to fuel the reactors that generated the initial plutonium stockpile. Secondly, the radiotoxicity of waste should also include HLW generated from reactors used to produce the plutonium stockpile. This additional information was not publicly available and was not accurately estimated in the fuel feed assessment. Therefore, it was excluded and results should only be used for the comparative assessment of fuel cycles or as a rough indication of the inter-generational liability of nuclear waste.

4.3.4.4 1000 year radiotoxicity in a disturbed repository

Total radiotoxicity at 1000 years represents the potential hazard in a disturbed repository, where a repository is compromised and there is the potential for large releases 1000 years after the closure of a repository. Under nominal operating conditions there will be no significant release from a repository, however given such long timescales, there is the potential for human intrusion or natural events that could lead to large releases. 1000 years was assumed to be the earliest timescale for a disturbed repository. In addition, 1000 years is an appropriate near-term timescale to require a large improvement in radiotoxicity. The total radiotoxicity represent the total ingested dose of all material in a repository, giving a maximum upper limit to the problem. Ingested dose is considered as it is the most likely exposure pathway, through food and water [111]. Results were given as the 1000 year radiotoxicity, relative to the direct disposal scenarios at the same date. Comparing results to the direct disposal scenario gives a good indication of the relative danger. A reduction in 1000 year radiotoxicity of a half was considered to be a significant improvement in this thesis as the radiotoxicities are high and anything less than half was considered too small. The evolution of radiotoxicity was plotted from 1000 years to 1500 years, to see if results changed significantly over this period given the different final inventories of fuel cycle scenarios.

4.3.4.5 Repository size

The main factor influencing repository size is the decay heat generated by nuclear waste. To obtain an accurate representation of repository size, the host geology must be known and waste canister loading must be optimised to ensure that temperature limitations are not exceeded for the waste package wall and the centreline temperature between canisters. Modelling a repository is not feasible for this study as a UK repository has not been sited and therefore the host geology is unknown. In addition, the optimisation of repository loading and waste canisters was outside of the scope of this study.

The relative size of a repository can be estimated from the cumulative decay heat (CDH), which is the integrated decay heat from time of emplacement to the peak centreline temperature between canisters. Several studies have used integration periods of 500, 1500 and 2000 years [121,124,127]. 1000 years was selected as a middle ground

between UK and international studies, with the sensitivity of the integration period tested in Section 7.3.3.1. It is assumed that material is sent to a repository at the end of the fuel cycle and stored in the interim to allow decay heat to reduce.

The impact of cooling time on relative repository size was considered from 50 to 350 years after the end of the fuel cycle, to allow FPs and ^{244}Cm to decay [130]. Results were given relative to the direct disposal scenarios which were cooled to the same date. Any reduction in repository size would be preferable, resulting in the reduced cost of building and operating a repository. As a result of the low accuracy of the CDH repository size estimation, a size reduction of at least 20% was needed to ensure that there would actually be a reduction in repository size. The accuracy of CDH is the main limitation, as well as the assumption that all waste is stored and only sent to a repository at the end of the fuel cycle.

4.3.4.6 Bare sphere critical mass

Bare sphere critical mass is an indicator of the degradation of the fissile feed vector as a result of irradiation [124]. The final inventory TRU vector was taken from ORION and used to calculate mass required to get $k_{eff} = 1$. The fuel vector, taken from ORION, was modelled using the SERPENT Monte-Carlo code as a bare metallic sphere. The radius of the sphere was increased until $k_{eff} = 1$ with the results given to the nearest 0.1 kg. The degradation of the isotopic vector was used to indicate the impact of the fuel cycle on the isotopics, and the reduced attractiveness of material to a potential proliferator wishing to cause a criticality. This could be considered a small factor in terms of proliferation resistance as it does not account for the weapons usability of the material. There are several limitations to using bare sphere critical mass to assess proliferation resistance as it not account for other intrinsic factors which influence the proliferation resistance of fuel cycle, discussed in Appendix D. Despite these drawbacks, bare sphere critical mass is included in the assessment criteria as it is a measurable quantity that can be used to assess the relative performance of a fuel cycle.

Degradation of fuel vector occurs with more fuel recycling, so the first fuel batches reprocessed will have similar results for critical mass across different scenarios. Therefore the most degraded fuel vector was used from the last batch of reprocessing to rank

the effectiveness of a fuel cycle at degrading the stockpile fuel vector.

4.3.5 Improvement factors

Final results were tabulated as improvement factors over a reference case for each assessment criteria. An improvement factor of one has no improvement over a reference case. An improvement factor greater than one represents an improvement relative to the reference case and an improvement factor less than one represent a dis-improvement relative to a reference case.

A single improvement factor was tabulated for each assessment criterion and for each fuel cycle scenario. This allowed all assessment criteria for all scenarios to be compared side by side. By normalising the improvement factors and applying weightings to each assessment criteria, the improvement factors can be used for decision analysis to determine which fuel cycle scenarios warrant further study.

Weightings are user dependent, as such, results for decision analysis will differ based on the user. Examples of decision analysis have been presented in Chapter 10. However, the aim of the decision analysis chapter is to present the methods so the reader can repeat the work, applying their own weightings.

Chapter 5

Fuel feed assessment

5.1 Introduction

In this chapter, fuel feed refers to the fissile material which is mixed with depleted uranium to fuel a reactor. Fuel feeds for a UK fuel cycle must be assessed as they are not known and they are the starting point for both a fuel cycle modelling and neutronics modelling.

To model a UK fast reactor fuel cycle, reactor fuel feeds must be determined. In the UK, potential fuel feeds are the UK's plutonium stockpile and reprocessed SNF from new build LWRs. The UK has a long history or reprocessing Mangox and AGR reactor fuel which has produced a large stockpile of separated plutonium. In 2013 the UK civil plutonium stockpile contained 99.6 tonnes of UK owned plutonium and 23.4 tonnes of foreign owned plutonium [13]. Reprocessing in the UK is expected to end between 2018 and 2020 [14, 15], leaving an estimated plutonium stockpile of 140 tonnes [1]. Current UK plans also include the construction of new thermal reactors to replace the current reactor fleet. At present, the new build goal is 16 GWe by 2050 [3].

ORION fuel cycle models and ERANOS neutronics models need a representative fuel feed to model a UK SFR and fuel cycle scenarios. The Office for Nuclear Regulation (ONR), Nuclear Decommissioning Authority (NDA) and the Department of Energy and Climate Change (DECC) have published reports which outline the current plutonium stockpile or estimate the stockpile mass at the end of reprocessing. However, information on the isotopic content of the UK's plutonium stockpile is not publicly available [152]. In addition, the final mass and isotopic content of SNF that

might arise from the UK's new build programme is not publicly available. There are inherent difficulties associated with estimating UK new build as the timescales for deployment, type of reactors and number of reactors will be subject to change. As such, there is a lot of uncertainty surrounding the specifics of the UK's new build programme.

To address these problems ORION fuel cycle models were used, with publicly available information, to assess the build-up of potential fuel feeds. Two fuel feeds were assessed, the UK's plutonium stockpile and the potential transuranics built up from reprocessing SNF from new build LWRs. For the UK's plutonium stockpile, publicly available information on reactor operating histories and reprocessing histories were used to estimate the build-up of plutonium. The final mass of plutonium, estimated with ORION, was compared to data published by the ONR and NDA. Due to large uncertainty in the UK's new build programme a basic approximation was used for new build LWRs, using a PWR and constant build rate to meet current new build goals.

This chapter aims to answer the following questions:

1. Does the total plutonium stockpile build-up at a similar rate to that reported annually by the ONR?
2. At the end of reprocessing is the plutonium stockpile mass similar to that reported by the NDA?
3. What is the isotopic vector of stockpiled material at the end of reprocessing (2025) and when this study considers fast reactors to be built (2040)?
4. How much plutonium and MAs will be generated by the new build scenario?
5. What is the isotopic vector of reprocessed new build SNF at the end of reprocessing, and at an average point when fast reactors will utilise it?
6. What are the known limitations or sources of error with the method used to estimate the UK's plutonium stockpile and SNF from new build reactors?

5.2 The UK's plutonium stockpile

There is limited information on the UK's plutonium stockpile. The ONR, NDA and DECC have published reports which outline the current stockpile or estimate the

stockpile at the end of reprocessing [1, 13]. However, these reports do not contain details of the isotopic content of the stockpile. This section outlines the methods used to estimate the UK's plutonium stockpile, the parameters used as well as the limitations and sources of error.

One of the key aspects of modelling the build-up of the UK's plutonium stockpile is americium in-growth. Americium in-growth is the decay of ^{241}Pu to ^{241}Am during the storage of separated plutonium. Americium in-growth is affected by the buffering of fuel prior to reprocessing. A shorter cooling period before reprocessing results in less ^{241}Pu decay prior to reprocessing, leading to more ^{241}Pu in the plutonium stockpile and ultimately resulting in more americium in-growth. Therefore, representing the build-up of fuel and reprocessing is very important in determining the americium content of the plutonium stockpile.

5.2.1 Method

ORION was used to model SNF produced from AGR and Magnox reactors, the cooling and reprocessing of spent fuel. Several parameters were needed in the model: An average burnup and SNF vector for AGR and Magnox fuel; AGR and Magnox reactor operating histories; fuel inventory for AGR and Magnox reactors; minimum cooling time of AGR and Magnox fuel before reprocessing; reprocessing histories of AGR and Magnox fuel through the B205 and THORP plants, without foreign reprocessing contracts. Evaluation of each of these parameters is discussed in the following section and summarised in Table 5.1.

The ORION model, used to simulate the build-up of the UK's plutonium stockpile, is shown in Figure 5.1. The initial feeds represent the annual SNF discharged from AGR and Magnox reactors. This is followed by a minimum period of cooling before the SNF is stored in a buffer ('Mag Buff' and 'AGR Buff'), ready to be reprocessed. Magnox reprocessing ('Mag Rep') models the annual throughput of Magnox fuel through the B205 reprocessing plant. AGR reprocessing ('AGR Rep') models the annual throughput of AGR fuel through the THORP reprocessing plant. Reprocessing plants extract the plutonium and send it to the 'Pu Stock' buffer, sending all other isotopes to 'Repro Waste.' As the fuel cycles in this study are using the UK's plutonium stockpile as a starting point, fission products and MAs produced from AGR and

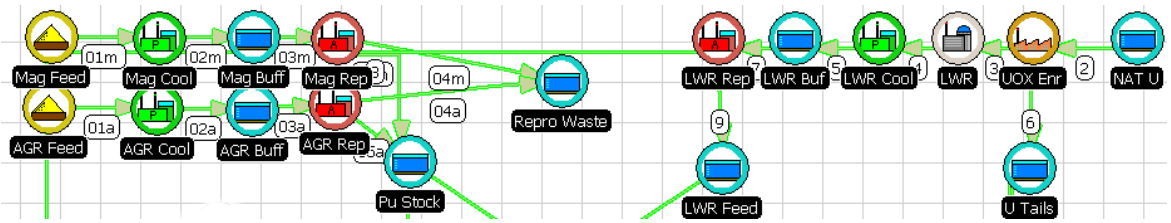


Figure 5.1: ORION models used to estimate the UK plutonium stockpile ('Pu Stock' left hand side) and reprocessed fuel from new build LWRs ('LWR Feed' right hand side).

Magnox spent fuel were ignored in any final analysis.

5.2.2 Parameters

This section outlines the parameters taken from literature which were used to model the build-up of the UK's plutonium stockpile. Parameters used, their source, and other estimates used are summarised in Table 5.1. The limitations of these parameters and potential sources of error are discussed in Section 5.5.

The UK's plutonium stockpile is mostly reprocessed AGR and Magnox fuel. SNF vectors for typical AGR and Magnox burnups are available without the need for neutronics modelling [156]. Information on reactor operating histories is needed to determine how much AGR and Magnox fuel has been produced. Reactor operating histories can be found in the IAEA Power Reactor Information System (PRIS) [5]. Minimum cooling times of AGR and Magnox fuel are publicly available [157, 158] and reprocessing histories can be requested from the NDA as part of the freedom of information act [152].

5.2.2.1 Spent fuel feed

All data used to determine the spent fuel feed is described in Table 5.1. Reactor operating histories for were tabulated by year in terms of unit power ($\text{Reactor}_{\text{GWth}}$) and annual load factor (LoadFactor), summarised in Figure 5.2. Reactor inventories were also tabulated in terms of tonnes heavy metal ($\text{Reactor}_{\text{tHM}}$) along with the average burnup for reactors ($\text{Burnup}_{\text{GWdays/tHM}}$). The total fuel throughput per year (Fuel_{tHM})

Table 5.1: Sources used to estimate the UK's plutonium stockpile.

Parameter	Source	Details
REACTORS		
Reactor operating histories	IAEA-PRIS database [5]	1970–2012 in Figure 5.2. Needed to extrapolate before 1970 and after 2012
Extrapolated reactor histories	AGR and Magnox estimated closure dates [18, 19, 20].	Up to 2024 for AGRs and 2015 for Magnox. Extrapolated load factors were estimated as the average of the previous three years of load factors.
Magnox inventories	NDA FOI [152]	Given as tHM.
AGR inventories	NKS [153, 154] and AGR site visitors centres [155]	Given as tHM.
AGR/Magnox BU and SNF vector	NIREX Report [156]	18 GWd/t for AGRs and 5.6 GWd/t for Magnox. Averaged over lifetime, ignoring changes in burnup with experience. AGR was expected to be a high estimate. Magnox was expected to be a low estimate.
REPROCESSING		
Cooling times	Magnox-Sellafield [157], AGR-NDA [158]	AGR 5 years, Magnox 180 days. An extra year was added to the minimum cooling times to account for transportation time.
Reprocessing histories	NDA FOI [152]	Received as total THORP and total Magnox (B204 and B205) up to 2012, Figure 5.3. Needed to extrapolate to end of reprocessing and account for foreign reprocessing contracts.
Foreign THORP reprocessing	Thorp baseload and post-baseload customers [12] & RWMD [15].	Approximately 50% UK. Assumed to be 50% annually.
Foreign Magnox reprocessing	Italy from PRIS database [5]	Assumes Italian Magnox fuel was reprocessed in the UK.
Extrapolated THORP reprocessing	NDA [14, 15]	THORP is expected to close by 2018, with 3,500 – 5,000 tHM of AGR fuel left over. Assumed that future throughput was the average of previous 3 years.
Extrapolated & adjusted Magnox reprocessing	NDA [15]	B205 is expected to complete reprocessing all Mangox fuel by 2017–2020, also 3,200 tHM of Magnox fuel should be buffered in 2014. To account for the overestimation of Magnox burnup, the B205 throughput was scaled down to 68% of the real value. Assumed that future throughput was the average of previous 3 years.

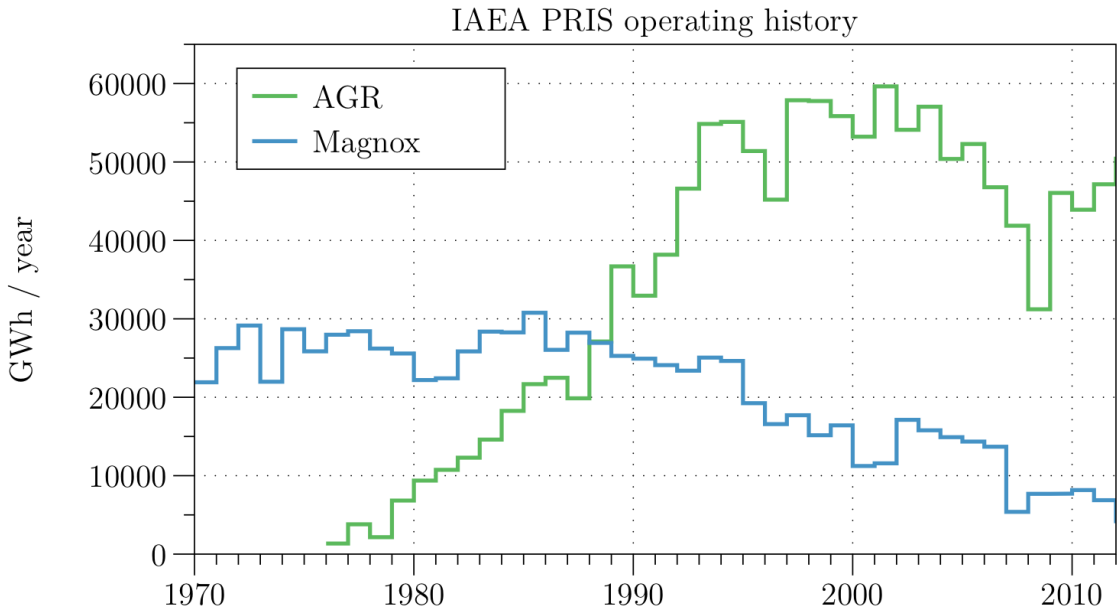


Figure 5.2: Total AGR and Magnox operating histories from the IAEA PRIS database [5] .

for each reactor was calculated as,

$$\text{Fuel}_{\text{tHM}} = \overbrace{365.25 \times \text{LoadFactor}}^{\text{Annual operating capacity}} \times \overbrace{\text{Reactor}_{\text{tHM}} \times \frac{\text{Burnup}_{\text{GWd/tHM}} \times \text{Reactor}_{\text{tHM}}}{\text{Reactor}_{\text{GWth}}}}^{\text{Average daily fuel throughput at full capacity}}, \tag{5.1}$$

and multiplied by the SNF isotopic vector for average burnup fuel to calculate the annual spent fuel generated by a reactor. All AGR and Magnox reactor results were summed by year to give the annual AGR and Magnox fuel feed sent to the cooling ponds in ORION.

5.2.2.2 Reprocessing

To get an appropriate proportion of ^{241}Am in-growth from the decay of ^{241}Pu, a realistic representation of AGR and Magnox reprocessing was needed. Fluctuations in reprocessing throughput have led to large buffers of spent fuel in cooling ponds prior to reprocessing. Any americium in-growth at the cooling stage would be removed by the reprocessing stage. The reprocessing histories of THORP and Magnox were taken from a Freedom Of Information (FOI) request from the NDA [152], Figure 5.3.

All information used to determine the annual reprocessing throughput of UK reactor fuel is described in Table 5.1. Reprocessing plant histories were extrapolated to

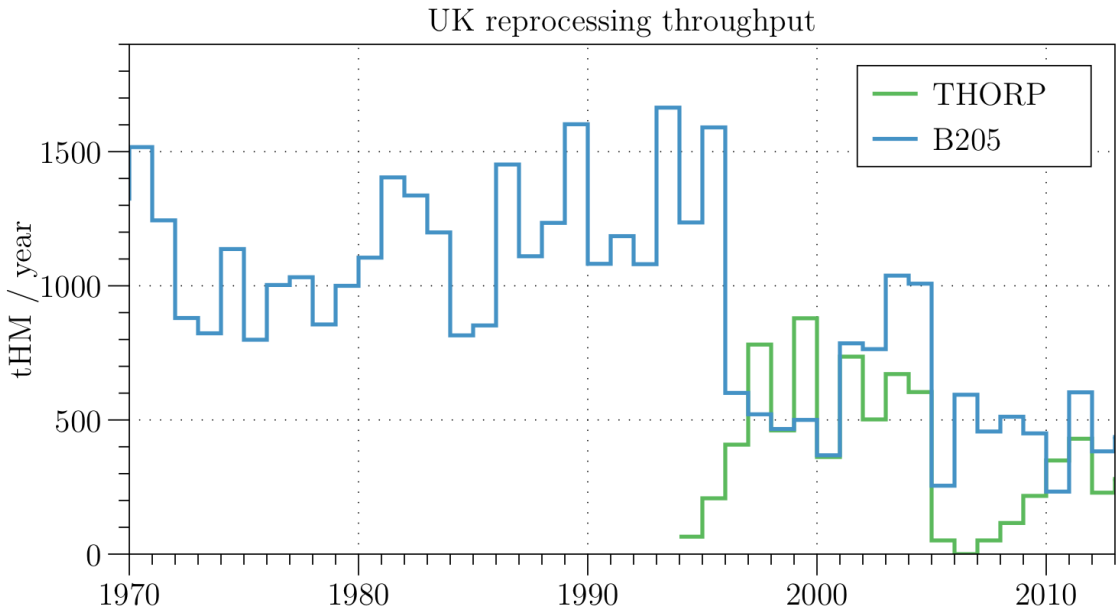


Figure 5.3: Annual throughput of UK reprocessing facilities from the NDA [152].

plant closure dates and scaled down to exclude foreign reprocessing contracts.

One limitation of the fuel feed assessment was that 3,200 tonnes of Magnox fuel should be buffered in 2014, waiting to be reprocessed [15]. Using the raw Magnox reprocessing data resulted in no fuel buffered to be reprocess in 2014. The underestimation of Magnox spent fuel production was presumed to be a result of lower burnup in earlier Magnox reactors, and an overall lower average burnup than the one used in Section 5.2.2.1. Reducing the burnup would lead to a greater annual throughput of Magnox fuel that would need to be reprocessed, which was not represented by the model. To account for the underestimation of Magnox fuel, Magnox reprocessing was scaled down to 68% of the real value. At 68%, all Magnox fuel was reprocessed by 2020, fitting the parameters in Table 5.1.

5.2.3 Results

At the end of reprocessing in 2025 the ORION model calculated the UK's plutonium stockpile to contain 112.6 tHM, 108.2 tHM of plutonium and 4.4 tHM of americium. The ratio of Magnox to AGR plutonium was approximately 4:1. Table 5.2 shows the plutonium vector, and the percentage of americium in-growth.

Table 5.2: Estimated plutonium vector and americium in-growth at the end of reprocessing in 2025 and at start up of SFRs in 2040.

Nuclide	**2025**	**2040**
^{238}Pu	0.24	0.21
^{239}Pu	66.08	66.19
^{240}Pu	25.66	25.97
^{241}Pu	1.69	0.82
^{242}Pu	2.42	2.42
^{241}Am	3.91	4.69

5.2.3.1 Comparison with published data

Published information was used to assess the validity of the estimated UK plutonium stockpile. THORP is expected to close by 2018, with 3,500 to 5,000 tHM of AGR fuel left over, depending on lifetime extension [14, 15]. In ORION 5,900 tHM of AGR SNF was left over. AGR SNF left over is not significantly more than the 3,500 to 5,000 tonnes estimate. The over estimation in ORION could be a results of underestimating AGR average burnup, or differences in the proportion of UK and foreign reprocessing contracts.

As discussed previously, 3,200 tonnes of Magnox fuel should be buffered in 2014, waiting to be reprocessed [15]. This was not the case in the ORION model and was assumed to be a result of early Magnox operation having much lower burnup resulting in higher annual throughput.

The ONR publishes the total separated plutonium stockpile in the UK, as well as the total plutonium stored at reprocessing plants [13]. Table 5.3 compares the UK plutonium stockpile calculated in ORION to the real values published by the ONR from 2008 to 2013. The ORION values are within 6% of the ONR values, however, the real stockpile is increasing faster than the plutonium stockpile calculated in ORION. Difference in the stockpile build-up rate could be a result of the ratio of UK to foreign plutonium reprocessed annually. Alternatively, the burnup of AGR fuel from 2008 to 2013 could be lower in the ORION model than real life, resulting in less plutonium being produced per tonne of SNF reprocessed.

In a 2014 NDA position paper on plutonium management options, the total separated plutonium in the UK at the end of reprocessing was estimated as 140 tonnes,

Table 5.3: Plutonium stockpile estimate compared to ONR published data (determined as total separated plutonium stored in the UK minus foreign owned plutonium) [13].

Year	Estimate (tHM)	ONR (tHM)	Difference (%)
2008	86.8	82.1	5.7
2009	87.9	84.4	4.2
2010	89.2	86.8	2.8
2011	90.9	90.3	0.7
2012	92.6	96.4	-3.9
2013	94.3	99.6	-5.3

including foreign owned plutonium [1]. In a 2008 NDA paper [159] 34 tonnes of plutonium was estimated to belong to foreign bodies by the end of reprocessing, leaving the UK with 106 tonnes of plutonium which is within 7% of the ORION estimate.

To the authors knowledge there has been no published information on the isotopic vector of UK's plutonium stockpile or americium in-growth that could be used to validate the ORION results.

5.3 New build LWRs

16 GWe of new build nuclear power is planned in the UK to replace the current fleet of nuclear reactors. Although the reprocessing of new build SNF has not been planned it is one of the pathways investigated by the UK R&D strategy [3].

It is not possible to accurately represent the build-up of new build LWR SNF as a result of the uncertainty in the operation of 16 GWe of new nuclear reactors over their lifetime. However, it is possible to get a representative model of new build, using typical reactor designs and an average build rate. This section outlines the methods used to model new build reactors and the estimations made in modelling their deployment and operation.

5.3.1 Method

ORION was used to model SNF generated from new build LWRs with a period of cooling before reprocessing. Several parameters were required for the model:

- An average LWR burnup, inventory and SNF vector;
- Build rate of reactors;
- Reprocessing parameters.

Evaluation of these parameters is discussed in the following section and summarised in Table 5.4.

The ORION model used to simulate new build LWRs with reprocessing of spent fuel is shown in Figure 5.1. ORION was used to model the enrichment of uranium ('UOX Enr') and irradiation in an LWR. Isotopic content of irradiated LWR fuel was taken from published LWR SNF data, Table 5.4. The reactor stage is followed by cooling and reprocessing to leave a stockpile of separated material, 'LWR Feed'.

A range of reactors are being considered for the UK new build sites. There are several variables which make an accurate estimation of new build SNF difficult: The number of reactors built, the timescale for their build, lifetime of reactors, load factor and the average burnup of reactors. These variables are subject to change, therefore an accurate calculation of new build SNF would not be worth while. A simple approximation of new build reactors was deemed adequate, using a single reactor design and the current goal of building 16 GWe by 2050.

5.3.2 Parameters

This section outlines the parameters taken from literature as well as estimations that were made with the reasoning explained. The limitations of these parameters and potential sources of error will be discussed in Section 5.5.

At present, only LWRs are being considered for UK new build sites, specifically PWRs (EPR and AP1000) and BWRs (ABWR). For simplicity, only one type of reactor was chosen to represent all new build reactors, a PWR. A single burnup PWR SNF composition was selected to represent new build reactors in ORION. Data used in the ORION model and their sources are tabulated in Table 5.4.

New build reactor build rate was based on the current new build goal of 16 GWe by 2050, which was modelled as 10 EPR sized reactors in ORION, totalling 16.5 GWe. The build rate is shown in Table 5.5. This build rate is ambitious but based on current policy [3, 136].

Table 5.4: Sources used to estimate the build-up of SNF from new build LWRs.

Parameter	**Source**	**Details**
Reactor	AREVA [160] NDA [161]	1.65 GWe EPR-type PWR, 60 year lifetime
Reactor SNF	NDA [161] NIREX [156]	48 GWd/t
Build rate	DECC [3, 136]	First PWR by 2023, 16 GWe by 2050. Assume constant rate of build, one 1.65 GWe PWR every 3 years.
Cooling times	Estimated [81, 86, 114]	Conservative estimate of 10 years. The planned Rokkasho reprocessing plant was designed to accept average burnup of 45 GWd/t with 5 years of cooling. AGR fuel is cooled for a minimum of 5 years for THORP. 10 years is a conservative estimate assuming minimal developments on THORP technology.
Reprocessing	Estimated [88, 130]	Pu only, or Pu+Am. LWR reprocessing assumed to be too near-term for Cm reprocessing.

Table 5.5: Build rate of PWRs to meet the target of 16 GWe by 2050 [3].

Year	**Total # Reactors**	**Capacity (GWe)**
2023	1	1.65
2026	2	3.30
2029	3	4.95
2032	4	6.60
2035	5	8.25
2038	6	9.90
2041	7	11.55
2044	8	13.20
2047	9	14.85
2050	10	16.50

It was assumed that reprocessing would start as soon as PWR SNF was available. By reprocessing fuel as soon as possible allows for its use in SFR fuel cycles as soon as possible. There were two reprocessing options used, the extraction of plutonium alone, or the extraction of plutonium and americium. Curium was not extracted at reprocessing as it was assumed that a near-term reprocessing plant for PWR fuel will not be capable of separating curium.

5.3.3 Results

The estimated stockpile inventory from reprocessed new build reactor fuel was 223.6 tHM or 207.3 tHM at the end of reprocessing in 2130, depending on whether americium was reprocessed. Total masses at the end of reprocessing can be found in Table 5.6, and the isotopic vector of in Table 5.7.

Table 5.6: Estimated stockpile from reprocessed new build fuel at the end of reprocessing in 2130, using ORION.

Element	**Pu (tHM)**	**Pu+Am (tHM)**
TRU	207.3	223.6
Np	0.6	1.3
Pu	195.5	195.5
Am	11.2	26.8

5.4 Results for neutronics models

An average fuel feed vector was required for ERANOS reactor design. Fuel feed vectors used for reactor design can be seen in Table 5.8. For the UK's plutonium stockpile these results were taken from 2040, when the fuel cycle scenarios start to build SFR. Two feed vectors are represented, one with the americium in-growth included and one with americium removed prior to fuel fabrication. The new build PWR SNF vector in Table 5.8 was taken at 2065 instead of at the end of reprocessing in 2120. With SFRs being constructed in 2040 the fuel vector at 2120 was too old with too much americium in-growth to be representative. The 2065 vector was taken as a middle

Table 5.7: Estimated transuranic vector from reprocessed new build fuel at the end of reprocessing in 2130.

Iso	Pu (wt.%)	Pu+Am (wt.%)
^{237}Np	0.27	0.60
^{238}Pu	2.53	2.35
^{239}Pu	56.24	52.14
^{240}Pu	27.98	25.84
^{241}Pu	1.42	1.32
^{242}Pu	6.16	5.71
^{241}Am	5.39	10.18
^{243}Am	0.00	1.77

ground for reactor design, with some plutonium used to fuel an SFR being younger and some being older.

Table 5.8: Estimated fuel feed vectors used for ERANOS reactor design.

	Pu Stockpile		New Build	
Iso	Pu	Pu+Am	Pu	Pu+Am
^{237}Np	-	-	0.03	0.14
^{238}Pu	0.22	0.21	23.18	2.91
^{239}Pu	69.23	66.19	55.83	51.81
^{240}Pu	27.16	25.97	27.85	25.84
^{241}Pu	0.86	0.82	4.47	3.8
^{242}Pu	2.53	2.42	6.11	5.67
^{241}Am	-	4.69	2.53	8.07
^{243}Am	-	-	0.00	1.76

It is worth noting that a reactor designed in ERANOS does not need an accurate fuel feed vector. In ORION, the initial fuel feed vector will change due to decay after reprocessing, to account for this, ORION adjusts fuel enrichments based on ^{239}Pu equivalence calculations. As such, an approximate fuel feed is needed for ERANOS reactor design to get representative cross-sections for use in ORION. The effective cross-sections are not very sensitive to small changes in isotopic fuel vector as discussed in Section 6.3.3.1. As such, a representative rather than accurate fuel vector was needed for reactor design.

5.5 Discussion

This chapter sought to answer several questions which were outlined in the introduction and discussed below.

The UK plutonium stockpile mass was estimated, using ORION, to be 112.6 tHM at the end of reprocessing. The ORION estimates were within 7% of published data on the total mass of the UK's plutonium stockpile. The isotopic content of the UK's plutonium stockpile estimate cannot be verified as there is no publicly available information. The final mass of transuranics from 16.5 GWe of new build reactors was 207.3 or 223.6 tHM, depending on whether americium was reprocessed. The final fuel feed vectors were used for further work designing SFRs in ERANOS, discussed in Chapter 6.

Fuel fuel cycle results and analysis of the direct disposal of stockpiled materials was not discussed in this chapter. Results for the direct disposal of stockpiled material were used as a reference case for comparing fuel cycle scenarios in further work, discussed in Chapters 7, 8 and 9.

One of the aspects of this chapter was to determine the limitations of the fuel feeds estimated. Limitations have been split across two sections due to different limitations on the UK plutonium stockpile and new build SNF.

5.5.1 Limitations: The UK's plutonium stockpile

Fuel cycle scenario studies will use the UK plutonium stockpile as a blended fuel vector of all AGR and Magnox plutonium. In reality, Mangox and AGR plutonium will be separate and canisters of plutonium which will vary based on reactor burnup and the age of the batch. Whilst blending all of the UK's plutonium is unrealistic it is not known how different batches of reprocessed plutonium will be used, but it is assumed that there will be some blending. Without blending of plutonium, enrichment and potentially fuel designs will have to vary from batch to batch, particularly varying from Mangox to AGR plutonium.

As previously discussed, the burnup of 5.6 GWd/t for Magnox fuel is likely to be high and 18 GWd/t for AGR fuel is likely to be low. The average burnup was chosen to represent the average burnup over all AGR and Magnox spent fuel, therefore some

discrepancies are expected as early spent fuel will have lower burnups and more recent spent fuel will have higher burnups. Magnox fuel with a burnup of 3 GWd/t was tested, but generated far too much fuel for reprocessing. As such, the higher burnup for Magnox fuel was selected and Magnox reprocessing was scaled to account for the lower mass of spent fuel generated in ORION. It is understood that using the higher burnup for Magnox fuel will result in a greater proportion of heavier plutonium isotopes and americium in-growth in the final plutonium stockpile.

Underestimating AGR burnup will have the opposite effect to overestimating Magnox burnup, but the influence of AGRs will be smaller as a result of the lower proportion of AGR plutonium in the final stockpile. Whilst this is not ideal, it is a known limitation of this work and the plutonium vector serves as a good basis for comparing reactors and fuel cycles.

The plutonium stockpile estimated in this chapter does not account for smaller plutonium streams, such as plutonium from reprocessing of experimental or prototype reactors. In addition, the UK has already taken ownership of some foreign owned plutonium and may take possession of more [162]. Some of the UK's plutonium stockpile has also become contaminated with chlorides and is not considered usable for fuel. Contaminated plutonium, foreign plutonium, and smaller plutonium streams were assumed to be a small factor and have not been accounted for in this study.

Trying to estimate the UK's plutonium stockpile more accurately would be useful. However, with so many variables and uncertainty in the future, a more accurate fuel feed would not lead to more accurate fuel cycle scenarios in later chapters. The current plutonium stockpile estimate should serve as a representative basis for the comparison of fuel cycle scenarios, not as an accurate representation of the UK's plutonium stockpile. The plutonium stockpile estimate was deemed acceptable as a result of the final inventory masses being in-line with those published by the ONR and NDA.

5.5.2 Limitations: New build SNF

The chosen build rate of one reactor every three years is ambitious, but in line with the 16 GWe by 2050 target. The use of PWRs and BWRs was not accounted for which would vary spent fuel inventories. The ORION model only uses one average burnup

of fuel over the lifetime of the fuel cycle. The burnup of fuel is expected to increase over the lifetime of the fuel cycles. In addition, reactor capacity factors and lifetimes are likely to vary.

One major assumption was that AGR SNF left over after the closure of the THORP reprocessing plant would not be reprocessed. Similarly, SNF from Sizewell B was assumed to not be reprocessed. This material was left out assuming it was disposed of separately.

There are a lot of variables in new build reactors which are unknown and will be subject to change over the lifetime of new build reactors. With a lot of uncertainty a simple model of new build, using a single type of reactor, is acceptable to get a representative build-up of new build SNF for comparing fuel cycle scenarios. Due to the variability in the future of UK new build, an ORION model cannot be an accurate representation of what the UK actually builds and operates. As such, fuel cycle models based on new build SNF in Chapter 9 can only be used as a basis for comparison, rather than an accurate model of UK new build and spent fuel inventories.

New build SNF was reprocessed as soon as it was available. This may not be realistic as it requires the deployment of a reprocessing facility shortly after the closure of THORP. Currently there are no plans to continue reprocessing, therefore the deployment of reprocessing shortly after the closure of THORP is unlikely.

5.6 Conclusions

This chapter estimated the isotopic vector and mass of the UK's plutonium stockpile and the SNF generated from a 16.5 GWe UK new build programme. There was approximately 112.6 tHM of transuranics in the estimated UK plutonium stockpile at the end of reprocessing and 207.3 to 223.6 tHM of transuranics produced from new build reactors.

Estimations of stockpiled materials were made using publicly available information. The main limiting factor in estimating the plutonium stockpile was the average burnups selected to represent AGR and Magnox reactors. However, the total mass estimated for the UK's plutonium stockpile showed good agreement, within 6%, with data published in ONR and NDA reports. It is not possible to accurately represent

the build-up of SNF from new build LWRs as a result of the uncertainty in the operation of 16 GWe of nuclear reactors over their lifetime. Parameters were selected to represent a typical reactor design and an average build rate to give a representative model of UK new build.

The fuel feeds are appropriate for fast reactor fuel cycle studies given the limitations of fuel cycle modelling. The fuel feeds will work well as a basis for comparing the impact of different fuel cycle scenarios. The aim of this work was not to obtain accurate models of a particular UK fuel cycle, but to develop representative models to see how different fuel cycle parameters alter waste performance, as discussed in Chapter 7, 8 and 9.

Key findings:

- There was approximately 112.6 tHM of transuranics in the estimated UK plutonium stockpile at the end of reprocessing and 207.3 to 223.6 tHM of transuranics produced from new build reactors.
- Mass estimates agreed well with published data, within 6%.
- Stockpiles were calculated using ORION so they could be used directly as the fuel feed in SFR fuel cycle models. The isotopic vectors were determined for use in ERANOS models.
- There are known limitations in the accuracy of the models. However, results are representative and sufficient to use as a reference case to compare fuel cycle scenarios.

Chapter 6

SFR neutronics design

6.1 Introduction

This chapter describes the ERANOS neutronics modelling process used in this study to design SFRs. The ERANOS calculation scheme was verified against other neutronics codes and used to optimise SFR designs for each fuel cycle scenario outlined in Section 4.2.

ORION fuel cycle models, used in later chapters, require SFR design parameters as well as effective cross-sections to model an SFR in the fuel cycle. Reactor designs will vary based on three key components, the fuel cycle set up, goals of the fuel cycle scenario, and the fuel feed being used for a reactor.

SFR designs are available in the literature but they are not designed for the specific fuel cycle parameters, fuel cycle goals, or fuel feeds considered in this study. Neutronics software can be used to model an SFR but the validity and run-time of calculation routes were unknown. MOX and metallic (ZR) fuelled reactor models were required for each fuel cycle scenario to compare the relative advantages of high TRL MOX fuel to lower TRL ZR fuel.

To address these problems the ERANOS neutronics code was comparatively assessed against results from an IAEA benchmark of fast reactor deterministic neutronics codes. A reference SFR was modelled in ERANOS using different calculation routes and was validated against the SERPENT Monte-Carlo code. The validity of different ERANOS calculation routes was assessed for SFR design scoping studies. Python

scripts were developed to automate the SFR design process and, where possible, simplifications were made to the calculation route and design methodology to speed up the design process. MOX and ZR fuelled SFR designs were optimised for each fuel cycle scenario accounting for the fuel cycle parameters, goals of the fuel cycle scenario and fuel feeds used. Each reactor design was kept within the domain of allowed design constraints outlined in Section 4.3.2.2. Final design and operating parameters for each SFR design is presented at the end of this chapter, with condensed cross-sections extracted for use in the ORION fuel cycle models presented in Chapter 7, 8 and 9.

This chapter aims to answer the following questions:

1. Does the ERANOS calculation route used give the same k_{eff} and burnup inventory results as other deterministic neutronics codes?
2. How similar are results between ERANOS and SERPENT using the same cross-section library and same reactor geometry?
3. Do SERPENT results change significantly with different cross-section libraries?
4. Are fast running, simplified calculation routes in ERANOS sufficiently accurate for design scoping studies?
5. What are the lowest feasible conversion ratios for reactors in each fuel cycle scenario?
6. How does the choice of MOX or metallic (ZR) fuel influence equilibrium isotopic vectors and conversion ratios?

6.2 Benchmarking and validation

6.2.1 IAEA benchmark

There have been several benchmark studies as part of the IAEA "Coordinated Research Project on Updated Codes and Methods to Reduce the Calculational Uncertainties of the LMFR Reactivity Effects." The one of most interest to this study is Phase 6, "Cores with MOX fuel containing minor actinides" [146]. This allowed ERANOS2.0 to be compared to CEA's ERANOS2.2, as well as other fast reactor deterministic codes, to test whether the calculation routes being used in ERANOS2.0 were appropriate.

Table 6.1: Initial elemental mass in the inner-core regions of the IAEA benchmark problem [146].

Region	**Np (kg)**	**Pu (kg)**	**Am (kg)**	**Cm (kg)**
Inner Fuel (IF)	8.4×10^{0}	1.0×10^{2}	2.1×10^{1}	1.3×10^{0}
Inner Breeding Zone (IBZ)	4.2×10^{-2}	3.3×10^{1}	1.7×10^{-3}	5.6×10^{-5}
Axial Breeder 1 (AB1)	3.5×10^{-3}	4.4×10^{0}	2.4×10^{-6}	1.7×10^{-8}
Axial breeder 2 (AB2)	1.5×10^{-2}	7.2×10^{0}	4.7×10^{-6}	4.9×10^{-8}

6.2.1.1 Methods

Results for k_{eff}, Δk, and fuel depletion were taken from the five contributors to the study and compared to ERANOS. Other results were reported in the benchmark, related to safety analysis work, which were out of the scope of this research. Fuel depletion, presented as change in isotopic mass, was compared for the Inner Fuel (IF) and Inner Breeding Zone (IBZ) of the benchmark problem, as they had the highest TRU inventories, Table 6.1, and received the greatest flux. Although the sample size of results was small, box plots were used to represent the range of results and show whether the author's ERANOS results were within the range of other institutions.

The benchmark had pre-defined material compositions and geometry for a BN-600 core containing TRUs. 2D RZ geometry was used with a diffusion flux solver and one burnup step of 140 Effective Full Power Days (EFPD). Recommendations for mesh sizes ($<$ 3.5 cm) were taken from other IAEA benchmark work [146, 163].

6.2.1.2 CEA Results

CEA took part in this benchmark using ERANOS2.2. Therefore it was expected that the author's results with ERANOS2.0 would be very close to CEA's. Using an RZ diffusion model, the initial k_{eff} was 0.986 compared to 0.988 for CEA with a Δk of 447 pcm compare to 455 pcm for CEA. Using the BISTRO transport model k_{eff} was 0.993 compared to 0.995 for CEA with a Δk of 462 pcm compared to 482 pcm for CEA. The change in isotopic inventories for the inner fuel and inner breeding zone regions of the core were similar between the present authors ERANOS2.0 results and CEA's, varying by less than 4% for plutonium isotopes. Curium, present in relatively low quantities, varied by up to 14%. These results are shown in more detail in Appendix C.1.

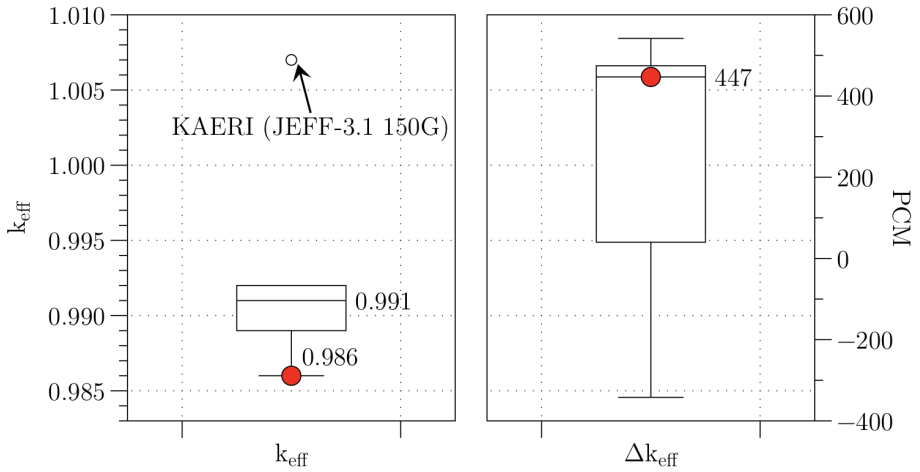


Figure 6.1: Boxplots of the k_{eff} and Δk results from the IAEA benchmark problem in Ref. [146]. All benchmark participant (CEA, FZk, JAEA, KAERI) results are presented with the present author's results plotted in red.

Results between the present author's ERANOS2.0 models and CEA's ERANOS2.2 models showed good agreement. Discrepancies may be a result of CEA using adjusted JEF-2.2 cross-sections.

6.2.1.3 All benchmark results

In the IAEA benchmark CEA, FZK, JAEA and KAERI submitted results using different deterministic neutronics codes with diffusion flux solvers. Different cross-section libraries were also used: FZK using JEFF-3.0, JAEA using JENDL-3.2, and both KAERI and CEA using JEF-2.2 and JEFF-3.1.

The initial k_{eff} and Δk values from these studies are shown in Figure 6.1 with the author's ERANOS (JEF-2.2) results overlaid in red. The change in isotopic mass with burnup was averaged across all results and each institution's results calculated as the percentage deviation from the mean. By representing all inventory results as the deviation from the mean, all major isotopes for a fuel region could be plotted on the same scale for comparison, Figure 6.2. ^{242m}Am isotopes were not plotted because the range in ^{241}Am branching ratios resulted in large variations in ^{242}Am and ^{242m}Am quantities. Curium was not included as the total curium inventories were small, Table 6.1, and lead to large variations in inventories.

Narrow interquartile range and narrow whiskers show that results do not deviate

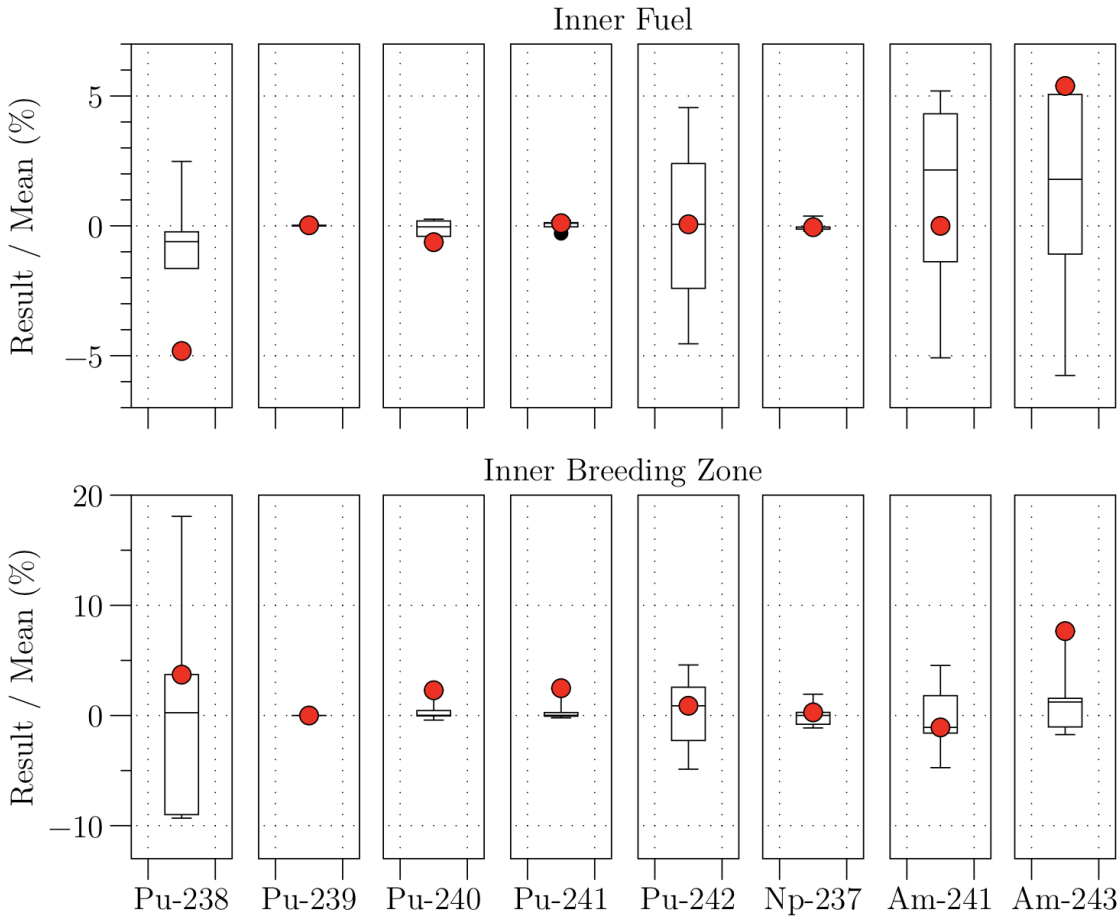


Figure 6.2: Boxplots of the change in isotopic mass with burnup from the IAEA benchmark problem in Ref. [146]. All benchmark participant (CEA, FZk, JAEA, KAERI) results are presented with the present author's results plotted in red. Results are shown as percentage variation from the mean, so all results could be plotted on the same scale.

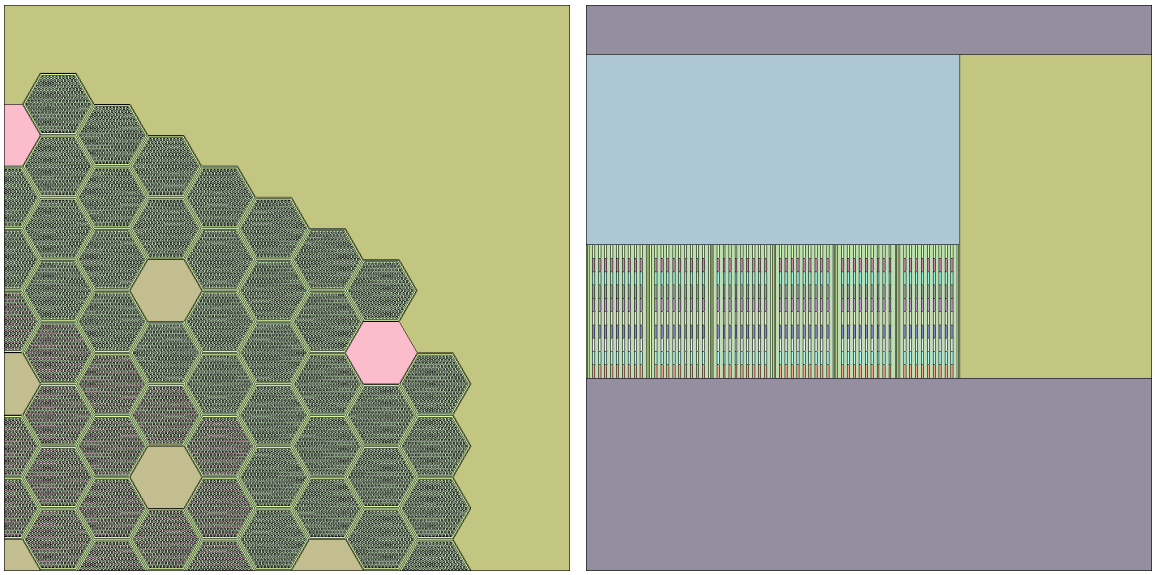
substantially with variations in cross-section and calculation route. Wide interquartile range shows that results vary substantially with cross-section and calculation route. Narrow interquartile range but wide wide whiskers shows most results clustering around the median and with some results deviating substantially. Results outside of the whiskers are outliers.

Results are similar for most k_{eff} and Δk results in Figure 6.1, but clearly vary due to calculation scheme and cross-sections. The author's ERANOS2.0 results were within a few hundred pcm of most results. The ERANOS2.0 k_{eff} result was low compared to all other institution results. The ERANOS2.0 k_{eff} and Δk results were closest to CEA's, therefore it is assumed that the low k_{eff} result was due to ERANOS or the use of the JEF-2.2 cross-section library. The KAERI results are considerably different which is assumed to be a result of their choice of ^{241}Am branching ratio. As shown in Appendix C.3, branching ratios and the quantity of ^{242m}Am can have a large influence on k_{eff}.

In Figure 6.2, ERANOS2.0 results were within 1% of the average for plutonium and neptunium depletion in the inner fuel region, with the exception of ^{238}Pu, where there was a large spread of results and author's ERANOS2.0 result were an outlier. For ^{242}Pu, ^{241}Am, ^{243}Am, the interquartile range was more spread out, but ERANOS2.0 was within the range of other results. For the inner breeding zone, ERANOS2.0 results for ^{240}Pu, ^{241}Pu and ^{243}Am were the upper extremes, but relatively small deviations from the mean. Notable ^{238}Pu had the biggest range of results and seems to vary the most of the plutonium isotopes depending on the calculation scheme and cross-section.

6.2.2 SERPENT-ERANOS validation

A reference SFR, Figure 6.3, from the design optimisation step was used to compare different ERANOS calculation schemes and SERPENT Monte-Carlo methods with different cross-sections. The SERPENT-ERANOS verification served two purposes: benchmarking ERANOS for a reactor design related to the optimisation work, and remodelling the reference design in a different code to ensure there were no user errors in setting up the geometry or materials definitions.



(a) Top down, XY view.

(b) Axial RZ view, showing the fuel and axial burnable zones.

Figure 6.3: SERPENT geometry for reference SFR design..

6.2.2.1 Methods

k_{eff}, Δk and change in inventory with burnup were compared for all solution methods. Results were taken at 0% burnup, fresh core (0 GWd/t), to see if initial k_{eff} results were the same, and compared at 40% burnup (40 GWd/t) and 60% burnup (60 GWd/t) to compare k_{eff} values and the relative change in k_{eff} in each model. 40% burnup (40 GWd/t) and 60% burnup (60 GWd/t) were used to give approximate results for the Beginning Of Cycle (BOC) and End Of Cycle (EOC) of a five batch reloading scheme with no fuel shuffling.

ERANOS2.0 used JEF-2.2 cross-sections, ECCO was used to calculate effective cross-sections of the fuel, condensed to 33 energy groups for whole core flux calculations. Whole core calculations were compared for 2D RZ and 3D HEX-Z geometry with diffusion and transport solvers. SERPENT used continuous JEF-2.2 cross-sections, with identical geometry and burnable zones to ERANOS in HEX-Z geometry, as shown in Figure 6.3.

ECCO cell calculations with homogeneous (hom) and heterogeneous (het) geometry were compared. For whole core calculations in RZ geometry the diffusion solver was used and a transport (S_N) solver, BISTRO. In HEX-Z geometry the diffusion solver and nodal transport solver, VARIANT, were used. The different calculation schemes

Table 6.2: Summary of neutronics methods compared to run times.

Calc	Cell Geom	Core Geom	Flux Solver	Run Time
Hom-RZ-Diff	Homogeneous	2D RZ	Diffusion	10 mins
Het-RZ-Diff	Heterogeneous	2D RZ	Diffusion	25 mins
Het-RZ-BIS	Heterogeneous	2D RZ	S_4 BISTRO	29 mins
Het-HEX-Diff	Heterogeneous	3D Hex	Diffusion	2 hrs
Het-HEX-VAR	Heterogeneous	3D Hex	VARIANT	9 hrs
Serpent (12 cores)				12 hrs

are summarised in Table 6.2, with approximate computing times.

Results were also compared to the same SERPENT model using JEFF-3.1.1 and ENDF/B7 cross-sections. The aim was to determine if the difference between deterministic calculations and Monte-Carlo calculations was of a similar magnitude to the difference between cross-sections.

Equilibrium reactor Additional comparisons were made to look at running a reactor with reprocessing to its equilibrium point. Thirty full core reloads with reprocessing were simulated with the different ERANOS solvers and geometries, to see if equilibrium results diverged. It was expected that discrepancies between models will propagate with each recycle, leading to the quick running ERANOS calculation schemes being less valid for equilibrium reactor optimisation.

6.2.2.2 k_{eff}

Table 6.3 shows k_{eff} and Δk for different ERANOS calculation routes and SERPENT models. Results are given along with their difference, in pcm, to the fastest running ERANOS calculation route (Hom-RZ-Diff) which was used for initial design scoping studies in Section 6.3. Results are only shown for a MOX and metallic (ZR) fuelled, low CR SFR reference cases.

Across ERANOS results, the difference in k_{eff} is typically a few hundred pcm. The largest discrepancies are due to the transport model which give the highest values for k_{eff} and can be up to 1000 pcm greater than equivalent diffusion solutions. Δk was within 400 pcm for all ERANOS models independent of geometry, cell calculation and flux solver. SERPENT JEF-2.2 results differed from ERANOS Hom-RZ-Diff model

Table 6.3: Once-through plutonium burner results for ERANOS and SERPENT models. k_{eff} was taken at 0 GWd/t, 40 GWd/t and 60 GWd/t. Δk was taken from 0 to 60 GWd/t.

Zr Low-CR	$\mathbf{k_{eff}}$ 0%	$\mathbf{k_{eff}}$ 40%	$\mathbf{k_{eff}}$ 60%	**Δk** (pcm)	Difference to Hom-RZ-Diff (pcm)			
Hom-RZ-Diff	1.128	1.053	1.018	9604	-	-	-	-
Het-RZ-Diff	1.128	1.052	1.016	9802	42	-85	-156	198
Het-RZ-BIS	1.139	1.063	1.027	9612	879	883	871	9
Het-HEX-Diff	1.132	1.054	1.017	10003	359	92	-41	399
Het-HEX-VAR	1.139	1.061	1.025	9770	873	755	707	166
SERP (JEF-2.2)	1.136	1.058	1.021	9938	645	435	311	334
SERP (JEFF-3.1.1)	1.144	1.065	1.028	9831	1245	1109	1018	227
SERP (ENDF/B7)	1.145	1.065	1.028	9873	1291	1106	1022	269
MOX Low-CR								
Hom-RZ-Diff	1.153	1.078	1.042	9257	-	-	-	-
Het-RZ-Diff	1.157	1.080	1.044	9380	256	180	133	123
Het-RZ-BIS	1.165	1.088	1.052	9222	871	898	906	-35
Het-HEX-Diff	1.161	1.082	1.045	9571	547	340	232	314
Het-HEX-VAR	1.167	1.089	1.052	9323	1010	960	944	66
SERP (JEF-2.2)	1.156	1.078	1.042	9451	173	38	-21	195
SERP (JEFF-3.1.1)	1.170	1.092	1.054	9439	1273	1159	1091	182
SERP (ENDF/B7)	1.174	1.096	1.058	9337	1547	1553	1467	80

Table 6.4: Initial masses (kg) of the main transuranic isotopes in the inner fuel region of the reference SFR.

	^{238}**Pu**	^{239}**Pu**	^{240}**Pu**	^{241}**Pu**	^{242}**Pu**	^{237}**Np**	^{241}**Am**	^{243}**Am**
Zr Low-CR	1.4	440.9	171.3	5.1	16.4	0.0	0.0	0.0

less than the transport models.

Comparing the two most up to date cross-section libraries available in SERPENT, JEFF-3.1.1 (2007) to ENDF/B7 (2006), results do vary. The difference between JEFF and ENDF cross-section results were relatively small for the ZR model, less than 100 pcm, and larger for the MOX model. Whilst Δk was similar for old JEF-2.2 and new JEFF-3.1.1 results, the k_{eff} values were up to 1000 pcm higher using JEFF-3.1.1.

6.2.2.3 Inventory

Change in SNF inventories were compared for major TRU isotopes across ERANOS calculation schemes and SERPENT models. Results are shown for a MOX and metallic (ZR) fuelled, low CR SFR reference cases in Table 6.4, Figure 6.4 and 6.5. To highlight any discrepancies the change in mass from 0 GWd/t to 60 GWd/t were compared. Initial fuel masses are given in Table 6.4, the largest mass changes are shown in Figure 6.4, and the percentage difference of each model from the Hom-RZ-Diff reference case is shown in Figure 6.5.

All ERANOS results were close together. SERPENT and ERANOS inventories were within a similar range, with the exception of ^{240}Pu which differed by 5–15 kg. ^{242}Pu and ^{237}Np have large proportional changes compared to Hom-RZ-Diff, $\sim$20%, which corresponds to relatively small mass changes over the burnup period. Americium results have good agreement, which was unexpected considering it is not present at reactor start-up.

6.2.2.4 Equilibrium reactor

ERANOS results for once-through burners were close enough to permit the use of the simplest calculation scheme (Hom-RZ-Diff). However, small differences in results may add up to become significant when recycling fuel through a reactor for several

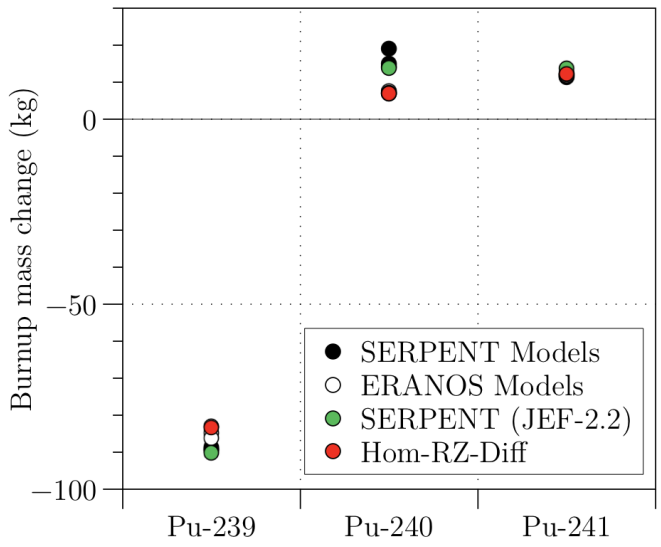


Figure 6.4: Results for the change in isotopic inventory for the three most abundant isotopes in SERPENT and ERANOS models. Black points are SERPENT results with different cross-sections, white points ERANOS results with different calculation routes.

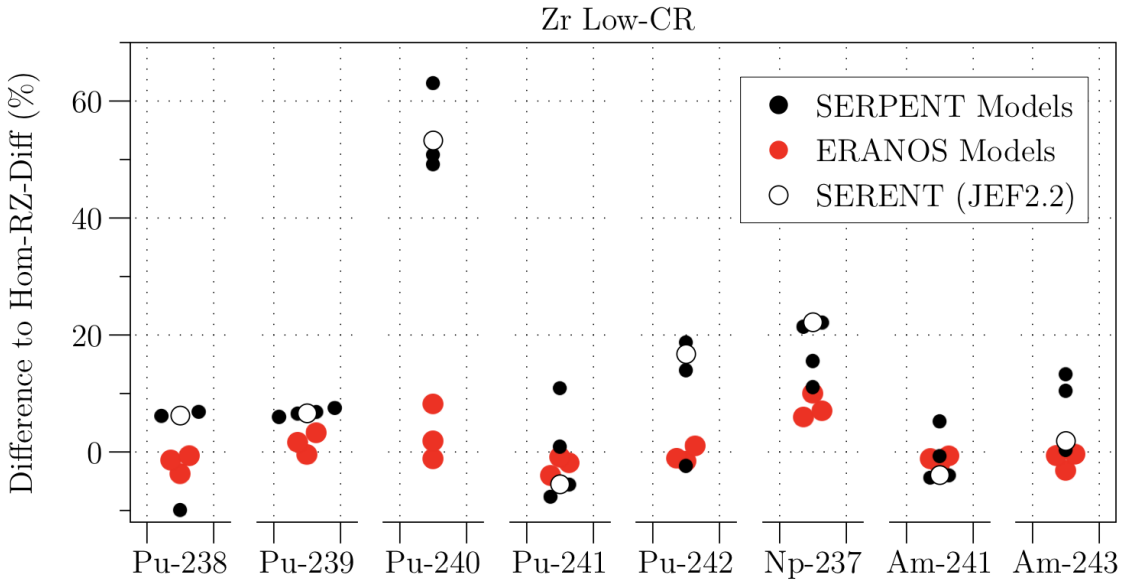


Figure 6.5: Results for the percentage change in isotopic inventory, relative to the Hom-RZ-Diff ERANOS model. Black points are SERPENT results with different cross-sections, red points ERANOS results with different calculation routes.

iterations. A python script was written to recycle all TRUs through an SFR for 30 iteration, with 5 years of fuel cooling, described in Chapter 4. Zr-low (CR=0.86) and MOX-low (CR=0.92) reactors were used.

After 30 iteration k_{eff} and fuel inventory were at an equilibrium point. At equilibrium, k_{eff} and inventories look identical for every subsequent cycle (see Figure C.3). Reaching equilibrium can take between hours and days of computing time, depending on the modelling method, Table 6.5. As such, fast running methods would be best for scoping studies where many design parameters need testing.

Table 6.5: Time taken to reach equilibrium (30 cycles) running different ERANOS routines.

Calculation	**Run Time**
Hom-RZ-Diff	5 hrs
Het-RZ-Diff	13 hrs
Het-RZ-BIS	14 hrs
Het-HEX-Diff	2.5 days
Het-HEX-VAR	12 days
SERPENT (4 cores)*	30 days

*SERPENT results were extrapolated.

Equilibrium ERANOS k_{eff} and Δk results in Table 6.6 differ by a similar amount at equilibrium to the results for a once-through burners in Table 6.3. All results are within a few hundred pcm of each other, with the transport model having the largest difference relative to Hom-RZ-Diff. Therefore, use of Hom-RZ-Diff is still acceptable for design scoping studies.

Final equilibrium spent fuel inventories were taken for all the ERANOS solution methods and compared relative to Hom-RZ-Diff in Figure 6.6. After 30 cycles, the differences in equilibrium spent fuel inventory were relatively small.

Differences in spent fuel inventory would be small compared to the accuracy of fuel cycle models which use one-group cross-sections to model in-core depletion. In addition, fuel feeds in the fuel cycle will change with reprocessing and decay, and one set of one-group cross-section from ERANOS will be used to represent in-core depletion over the lifetime of the fuel cycle. As such, the discrepancies between equilibrium fuel cycle results for different ERANOS models are relatively small when compared to the

Table 6.6: Equilibrium ERANOS results with all transuranics reprocessed. k_{eff} was taken at 0 GWd/t, 40 GWd/t and 60 GWd/t. Δk was taken from 0 to 60 GWd/t.

Zr Low-CR	$\mathbf{k_{eff}}$ 0%	$\mathbf{k_{eff}}$ 40%	$\mathbf{k_{eff}}$ 60%	$\mathbf{\Delta k}$ (pcm)	Difference to Hom-RZ-Diff (pcm)			
Hom-RZ-Diff	1.089	1.042	1.018	6367	-	-	-	-
Het-RZ-Diff	1.086	1.040	1.016	6306	-260	-137	-200	-60
Het-RZ-BIS	1.098	1.050	1.026	6320	704	790	751	-47
Het-HEX-Diff	1.090	1.043	1.018	6452	72	98	-13	85
MOX Low-CR								
Hom-RZ-Diff	1.069	1.026	1.004	6038	-	-	-	-
Het-RZ-Diff	1.068	1.026	1.004	5929	-97	63	12	-109
Het-RZ-BIS	1.077	1.034	1.012	5963	708	812	783	-75
Het-HEX-Diff	1.072	1.029	1.006	6089	230	281	178	51

scale of variability in fuel cycle feeds. However, it is worth noting that equilibrium cross-sections are an accurate way of representing in-core depletion for a fast reactor at equilibrium and at start up, despite differences in fuel feed vector and enrichment, as shown in Section 6.3.3.1.

6.2.3 Discussion

This benchmark section sought to answer several questions which were outlined in the introduction and discussed below.

In the IAEA benchmark problem, ERANOS2.0 results were close to CEA's ERANOS2.2 results and other deterministic codes. Close was considered to be within 5% of the average for the burnup mass change of plutonium isotopes and a few hundred pcm for k_{eff} and δk. In some cases ERANOS results were in the outer range of results. There is a good agreement between ERANOS and SERPENT using the same cross-section library. The difference between recent ENDF and JEFF cross-sections in SERPENT is low when compared to the difference between ERANOS diffusion and transport calculation schemes. The difference between JEF-2.2 (used in ERANOS) and the more recent JEFF/ENDF cross-sections in SERPENT is large for k_{eff}, but small for inventory calculations. Overall, fast running ERANOS calculations are adequate for design scoping work.

For the IAEA benchmark problem, ERANOS2.0 results were in, or close to, the

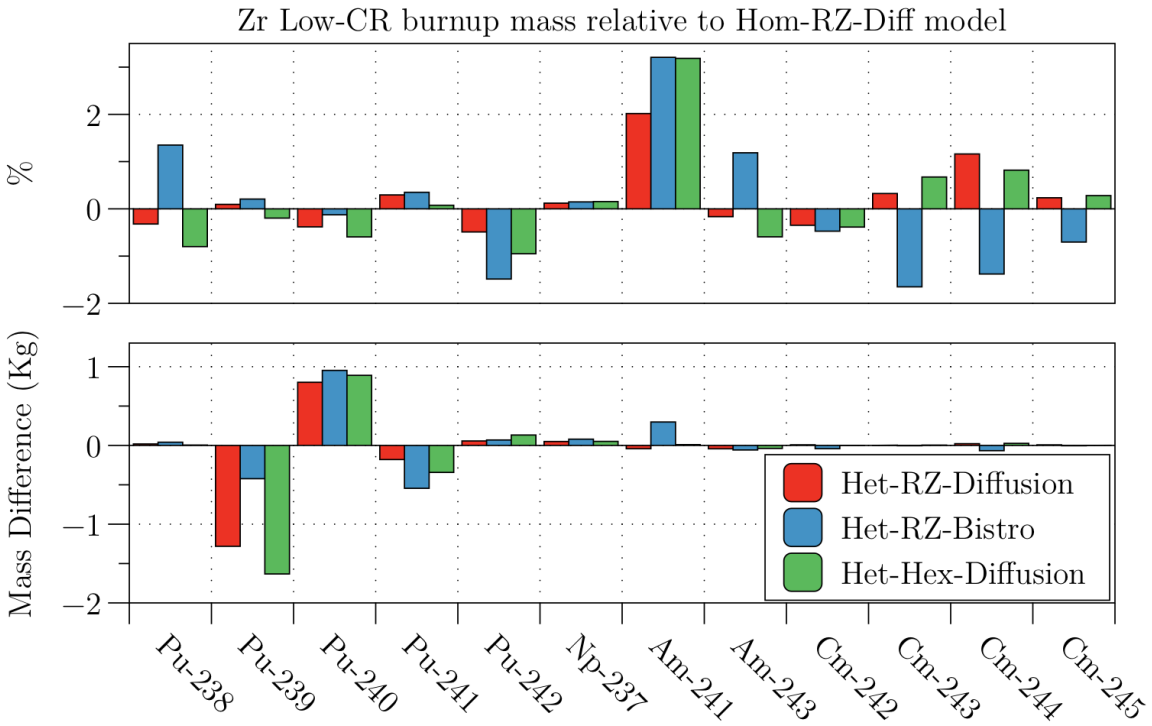


Figure 6.6: Equilibrium SNF compositions for ERANOS models. Results are plotted as the difference to the Hom-RZ-Diff model.

range of results submitted by other institutions. Some ERANOS2.0 results were the upper or lower extremes, but these results were not significantly different to CEA's ERANOS2.2 results. Discrepancies between ERANOS2.0 and other institutions results were assumed to be a result of two main factors, cross-sections libraries and calculation route. Further more, smaller deviations would be due to discrepancies in branching ratios or mesh size.

Although SERPENT and more detailed ERANOS calculation schemes deviated from the Hom-RZ-Diff results, the deviations were not significant enough to influence design studies. However, ERANOS calculation schemes do influence calculation time enough to make slower, HEX-Z models, less preferable. A few hundred pcm between calculation schemes is not significant when the tolerances in the design study are large to ensure reactors are within the range of allowable k_{eff} and Δk values. For depletion calculations, ERANOS and SERPENT results were within an acceptable range of each other, with the exception of ^{240}Pu. Some depletion results varied by a large proportion, however, the real mass differences were relatively small. When considering neptunium and americium, which are not present at reactor start-up, ERANOS and SERPENT

showed very good agreement. The reason for the ^{240}Pu discrepancy is unknown.

There was more than a 1000 pcm difference in k_{eff} between the newest available cross-section library in SERPENT, JEFF-3.1.1, and JEF-2.2 which was the only available cross-section library for ERANOS during this study. The difference in k_{eff} could have an impact on the range of feasible reactor designs. However, for a reactor design, inventory results were similar across different cross-sections in SERPENT.[1] As a result, it was assumed that final inventories for fuel cycle analysis would not be significantly different if more recent cross-section libraries were used with the same reactor geometry. However, there may be a slightly different range of reactor designs that are feasible.

Running reactors to an equilibrium point with different ERANOS calculation routes did not lead to a significant divergence of k_{eff} or inventory results. This was unexpected as final results deviated from the Hom-RZ-Diff model by a similar amount to the once-through burner models.

Overall, Hom-RZ-Diff methods were acceptable for design scoping work. If a reactor design was promising it was re-run using Het-RZ-Diff to ensure that the design was still feasible.

6.3 SFR design

Using the ERANOS calculation scheme determined for best practice in the previous sections, reactor designs were evaluated for different fuel cycles which were selected in Section 4.2, and outlined in Table 6.7.

6.3.1 Method

Each fuel cycle scenario modelled in ORION required an ERANOS neutronics model to generate cross-sections and fluxes that were used to represent the reactor. The different types of scenarios are summarised in Table 6.7, using two fuel feeds: the UK plutonium stockpile, and the UK plutonium stockpile with new build LWR SNF. There are three types of fuel cycle scenario that require different ERANOS modelling techniques, discussed in more detail in Section 4.3.2:

[1] With the exception of ^{240}Pu, previously discussed.

- BURNER scenarios – Operating once-through SFRs with stockpiled material.
- FEED equilibrium scenarios – Operating closed fuel cycle SFRs with reprocessing, using reprocessed material as the primary fresh fuel feed topped up with stockpiled material.
- FULL equilibrium scenarios – Operating closed fuel cycle SFRs with reprocessing, using reprocessed material as the only fuel feed.

Within each of these scenarios there are sub-scenarios which use different fuel cycle cycle and reactor parameters, summarised as:

- Two reactor designs – Metallic (Zr) and MOX fuelled reactor were tested for each scenario.
- Three reprocessed fuel feeds – Reprocessing of plutonium (PU scenarios), plutonium and americium (AM scenario), or all TRUs (TRU scenario).
- Two reprocessing variables – High cooling times and high losses (HI scenario), or low cooling times and low losses (LO scenario), described in Table 4.3.

Each varied parameter has its own fuel cycle scenario that was modelled in ORION and its own SFR reactor that was designed using ERANOS. The once-through BURNER scenario has four sub-scenarios based on reactor design and fuel feed (for the initial BURNER work there were also four high-CR SFRs). The FEED and FULL closed fuel cycle scenarios have twelve sub-scenarios, six based on reactor design and fuel feeds, with two variations based on reprocessing losses and cooling time. The same reactor design was used for HIGH and LOW scenarios, to ensure that a reactor design could cope with variable fuel cycle parameters.

SFR BURNER designs as well as FEED and FULL equilibrium SFRs were optimised using ERANOS. Python scripts were used to vary fuel design and run ERANOS. Scripts tested a range of feasible designs, checking final results to ensure that quantities such as k_{BOC}, k_{EOC} and Δk were within an allowable range, as discussed in Chapter 4. If ERANOS results were within an acceptable range, the model was re-run using more detailed, heterogeneous cell lattice geometry, to re-check that results were within an allowable range. The model parameters and key core characteristics were tabulated in order of CR, with highest or lowest feasible CR reactor selected, depending on the type of fuel cycle being studied.

Table 6.7: Reactor design methods corresponding to fuel cycle studies.

CR	**Feed**	**Fuel Cycle**	**ERANOS reprocessing feed**
Lo	Pu Stock	Once Through FR	-
Hi	Pu Stock	Once Through FR	-
Lo	Pu Stock	Few FR + Rep	FEED: Reprocessed fuel + Stock[1]
Lo	Pu Stock	Max FR	FULL: Reprocessed fuel[2]
Lo	Pu+LWR	Once Through FR	-
Lo	Pu+LWR	Few FR + Rep	FEED: Reprocessed fuel + Stock[1]
Lo	Pu+LWR	Max FR	FULL: Reprocessed fuel[2]

[1] Reprocessed material used as main feed, but TRU's lost due to burnup is replaced with stockpiled material.
[2] As fissile material in the fuel cycle reduces, the number of reactors is reduced to allow for a maximum reduction in inventory.

For reprocessing fuel cycles, equilibrium reactors were optimised rather than start-up reactors. With each recycle the fuels reactivity worth is degraded, requiring a higher enrichment for the same fuel assembly geometry. To ensure that peak allowable fuel enrichments were not exceeded, equilibrium reactors were optimised rather than start-up reactors. Python subroutines ran 30 recycles of the fuel to reach an approximate equilibrium point, Table 6.7. There were two recycling options, the FULL or FEED equilibrium methods: recycling spent fuel, adding enough material from the stockpile to make up for the loss of fissile material during irradiation (FEED scenario); or scaling up the amount of spent fuel to use it as the sole feed material (FULL scenario). The FULL scenario assumes a gradual reduction in reactor fleet as material in the fuel cycle is reduced. Different fuel feeds, cooling times and recycling of different elements were changed based on the fuel cycle scenario. The methods for FEED and FULL equilibrium scripts are described in more detail in Chapter 4.

The following sections outline the selected reactor designs for each fuel cycle scenario. Key design parameters and core characteristics are tabulated: Conversion Ratio (CR), TRU consumption per day (TRU/day), TRU inventory at the beginning of a cycle (BOC TRU), fuel volume percent (Fuel Vol), peak enrichment (TRU/HM), k_{eff} at the beginning of a cycle (k_{eff} BOC), reactivity loss over a cycle (Δk), power peaking factor (Power Peak), peak linear power (Peak Lin) and cycle length (Cycle).

Equilibrium fresh fuel feeds were also tabulated in Appendix C, with MOX and ZR average neutron energy spectrum plotted in Figure 6.7, to show how the energy

Table 6.8: Characteristics of high and low CR SFR's designed as once-through BURNERS of the UK's plutonium stockpile.

BURNER	Zr Lo-CR PU	Zr Lo-CR AM	MOX Lo-CR PU	MOX Lo-CR AM	Zr Hi-CR PU	Zr Hi-CR AM	MOX Hi-CR PU	MOX Hi-CR AM
CR	0.55	0.70	0.42	0.63	0.98	1.06	0.96	1.07
TRU/day (kg)	0.433	0.427	0.54	0.528	0.008	0.009	0.003	-0.002
BOC TRU (kg)	2464	2619	2780	2785	2744	2844	3481	3532
Fuel Vol (%)	23.1	24.5	18.7	18.7	48.9	49.6	49.6	49.6
TRU/HM (%)	30.0	30.0	40.0	40.0	14.4	15.0	16.8	16.8
k_{eff} BOC	1.055	1.055	1.073	1.042	1.001	1.012	1.020	1.006
Δk (pcm)	3288	2978	3210	3017	982	969	1188	715
Batches	5	5	5	5	4	3	5	5
Power Peaking	1.43	1.44	1.49	1.50	1.62	1.60	1.70	1.70
Peak Lin (kW/m)	35.6	35.8	28.4	28.7	40.4	40.1	32.7	32.7
Cycle (EFPD)	184	195	158	158	487	650	415	415

spectrum influences equilibrium fuel vectors.

6.3.2 Once-through SFRs

Table 6.8 describes the SFRs used as once-through BURNERS of the UK's plutonium stockpile. Table 6.9 describes the SFRs used as once-through BURNERS of SNF from UK new build reactors. There are two fuel cycles for each fuel feed considered, one with americium in-growth removed before fuel fabrication (PU) and one which includes americium in-growth (AM). For UK plutonium stockpile BURNERS high-CR reactors were also considered.

Results fit within the physical limitations of the fuel, reactivity, shut-down and thermal limits discussed in Section 4.3.2. Conversion ratios, as given by ERANOS, were lower for MOX fuelled reactors using the plutonium feed, due to the higher enrichment. The addition of americium to the fuel increased the CR.

Overall the lowest CRs for plutonium stockpile BURNERS were 0.42 to 0.7, and for new build SNF BURNERS this was 0.53 to 1.01. The much higher upper limit of new build SNF BURNERS is a result of the fuel feed containing more americium and the plutonium vector which is weighted more towards the heavier isotopes compared to that of the UK plutonium stockpile, Table 5.8. Despite a CR > 1 for some BUNRER reactors, there is still a net reduction of fissile material. The ERANOS conversion

Table 6.9: Characteristics of low CR SFR's designed as once-through BURNERS of reprocessed SNF from new build LWRs.

BURNER	Zr PU	Zr AM	MOX PU	MOX AM
CR	0.64	0.99	0.53	1.01
TRU/day (kg)	0.406	0.394	0.510	0.497
BOC TRU (kg)	2863	3409	3113	3819
Fuel Vol %	26.5	31.7	20.8	25.5
TRU/HM (%)	30.0	30.0	40.0	40.0
k_{eff} BOC	1.063	1.068	1.061	1.059
Δk (pcm)	2941	2015	3013	2138
Batches	5	5	5	5
Power Peaking	1.45	1.47	1.50	1.54
Peak Lin (kW/m)	35.9	36.7	28.7	29.4
Cycle (EFPD)	211	252	176	215

ratio calculation accounts for the contribution of all TRUs. For plutonium stockpile BURNERS, the highest CRs possible were 0.96 to 1.07, this could be increased with the use of breeder blankets, but blankets were not considered as part of this work.

6.3.3 Equilibrium SFRs

Using the methods described previously, reactors were designed using the two fuel feeds and run to equilibrium using the FEED and FULL methods.

SFR parameters and results using the UK's plutonium stockpile as a fuel feed are presented in Table 6.10, with the equilibrium fuel vectors in Table C.3. SFR parameters and results using new build LWR SNF as a fuel feed are presented in Table 6.11, with the equilibrium fuel vectors in Table C.4.

Using the UK plutonium stockpile as the fuel feed, and using the FEED equilibrium method, reactors had a CR of 0.64 to 0.91. Using the FULL equilibrium method, reactors had a CR of 0.73 to 1.05. For the new build LWR SNF fuel feed, using the FEED equilibrium method, reactors had a CR of 0.73 to 1.00 CR, using the FULL equilibrium method reactors had a CR of 0.81 to 1.05.

For the FEED equilibrium methods, the CR increased with the addition of americium (AM), however the TRU consumption per day stayed roughly the same. For plutonium only (PU) fuel, FEED-MOX-PU had a lower CR than FEED-ZR-PU due to

Table 6.10: Characteristics of equilibrium SFR reactors fuelled with the UK's plutonium stockpile.

FEED Scenario	**Zr PU**	**Zr AM**	**Zr TRU**	**MOX PU**	**MOX AM**	**MOX TRU**
CR	0.70	0.85	0.86	0.64	0.90	0.91
TRU/day (kg)	0.376	0.375	0.373	0.468	0.455	0.454
BOC TRU (kg)	3072	3360	3362	3758	3765	3767
Fuel Vol %	28.5	31.2	31.2	25	25	25
TRU/HM (%)	30.0	30.0	30.0	38.4	38.4	38.4
MA/HM (%)	-	1.9	2.3	-	2.9	3.5
k_{eff} BOC	1.054	1.061	1.059	1.065	1.023	1.022
Δk (pcm)	2655	2241	2187	2628	2178	2112
Batches	5	5	5	5	5	5
Power Peaking	1.45	1.47	1.47	1.53	1.56	1.56
Peak Lin (kW/m)	36.2	36.6	36.6	29.3	29.9	29.9
Cycle (EFPD)	227	249	249	212	211	211
FULL Scenario	**PU**	**AM**	**TRU**	**PU**	**AM**	**TRU**
CR	0.73	0.96	0.95	0.8	1.05	1.04
TRU/day (kg)	0.328	0.338	0.343	0.399	0.417	0.426
BOC TRU (kg)	3875	4191	4189	5410	5847	5842
Fuel Vol %	30.7	38.6	38.6	35.8	38.6	38.6
TRU/HM (wt.%)	30	30	30	38.4	38.4	38.4
MA/HM (%)	-	2.6	3.4	-	4.3	5.5
k_{eff} BOC	1.062	1.071	1.080	1.070	1.072	1.089
Δk (pcm)	2043	1609	1562	1931	1353	1318
Batches	5	5	5	5	5	5
Power Peaking	1.48	1.50	1.50	1.58	1.59	1.59
Peak Lin (kW/m)	37.0	37.4	37.4	30.3	30.6	30.5
Cycle (EFPD)	284	307	307	301	326	326

Table 6.11: Characteristics of equilibrium SFR reactors fuelled with the UK's plutonium stockpile and reprocessed SNF from new build reactors.

FEED Scenario	**Zr PU**	**Zr AM**	**Zr TRU**	**MOX PU**	**MOX AM**	**MOX TRU**
CR	0.73	0.94	0.94	0.68	0.99	1.00
TRU/day (kg)	0.362	0.361	0.360	0.450	0.452	0.451
BOC TRU (kg)	3282	3668	3670	3989	4697	4699
Fuel Vol %	30.7	34.0	34.0	26.5	31.2	31.2
TRU/HM (%)	30.0	30.0	30.0	38.4	38.4	38.4
MA/HM (%)	-	2.8	3.3	-	4.0	4.8
k_{eff} BOC	1.058	1.061	1.063	1.056	1.065	1.067
Δk (pcm)	2480	1901	1839	2499	1809	1737
Batches	5	5	5	5	5	5
Power Peaking	1.46	1.48	1.48	1.55	1.57	1.57
Peak Lin (kW/m)	36.4	36.9	36.9	29.6	30.0	30.2
Cycle (EFPD)	244	271	271	244	263	263
FULL Scenario	**PU**	**AM**	**TRU**	**PU**	**AM**	**TRU**
CR	0.81	0.96	0.95	0.81	1.05	1.04
TRU/day (kg)	0.329	0.337	0.342	0.396	0.416	0.429
BOC TRU (kg)	4001	4192	4190	5324	5848	5843
Fuel Vol %	36.8	38.6	38.6	35.1	38.6	38.6
TRU/HM (%)	30.0	30.0	30.0	38.4	38.4	38.4
MA/HM (%)	-	2.7	3.5	-	4.3	5.5
k_{eff} BOC	1.074	1.071	1.080	1.061	1.07	1.089
Δk (pcm)	2039	1605	1560	1948	1347	1316
Batches	5	5	5	5	5	5
Power Peaking	1.48	1.50	1.50	1.58	1.60	1.60
Peak Lin (kW/m)	37	37.4	37.4	30.4	30.7	30.7
Cycle (EFPD)	293	308	308	297	327	327

having a higher enrichment. With the addition of americium (AM and TRU), FEED-ZR-AM and FEED-ZR-TRU had lower CRs than FEED-MOX-AM and FEED-MOX-TRU respectively. As a result of the hard spectrum of FEED-ZR reactors, equilibrium fuels had an MA content of 1.9 to 3.3% where as FEED-MOX reactors had a higher MA content of 2.9 to 4.8%. This was due to the higher ratio of fission to capture for TRU nuclides in the harder neutron spectrum of ZR fuelled reactors.

For the FULL equilibrium method, CRs were higher and TRU consumption rates lower than for the FEED method. FULL methods lead to a greater degradation of the fuel vector. With a lower reactivity worth of the fuel feed a greater fuel inventory was needed to keep $k_{eff} > 1$. Greater fuel inventory lead to a greater mass of fertile ^{238}U. As a result of the degraded fuel vector and greater mass of ^{238}U in the reactors, there was more capture and less of a reduction in TRUs in the FULL SFRs. FULL-MOX reactors resulted in greater degradation of the fuel vector, which lead to high CRs at equilibrium than the equivalent FULL-ZR reactor.

For all FEED scenarios, characteristics such as k_{eff} and MA enrichment were within the limitations previously discussed in Section 4.3.2. For FULL scenarios, with all TRUs reprocessed, some of these limits were exceeded. This was a result of keeping enrichment and fuel volume the same for the AM and TRU scenarios. The aim of keeping fuel design the same was to compare the same reactor set up but with the addition of all TRUs rather than just americium. In keeping AM and TRU reactor designs the same, the initial k_{eff} for FULL-TRU reactors was greater than the limit of 1.08 and the MA proportion for the MOX reactors exceeded 5%. MOX reactors had a higher proportion of higher actinides due to the softer neutron spectrum, shown in Figure 6.7, which resulted in a lower fission to capture ratio.

6.3.3.1 One-group cross-sections

A set of one-group cross-sections from ERANOS will be used to model a reactor in ORION from beginning of life to equilibrium. Enrichment and fuel feed will change over the lifetime of reactor operation. To determine how much effective cross-sections change over this period, ECCO was used to export one-group cross-section for three reactor conditions. The first was an equilibrium reactor, where equilibrium was reached by varying the enrichment after each cycle based on ^{239}Pu equivalence. Next was an

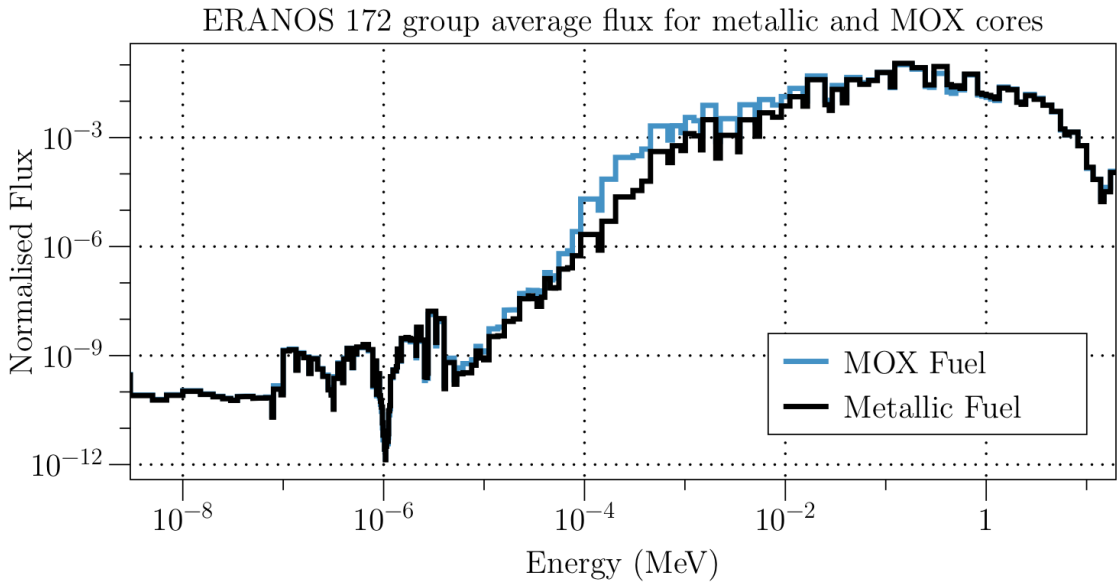


Figure 6.7: Normalised average flux spectrum from ERANOS for a metallic and MOX fuelled SFR.

equilibrium reactor where enrichment was kept the same for each recycle. Finally, was a beginning of life reactor with the low starting enrichment. These results are presented in Table 6.12, and the methods are discussed further in Appendix C.4. Table 6.12 shows that equilibrium effective cross-sections are the same using varied enrichment and constant enrichment methods to reach equilibrium. In addition, the beginning of life reactor has very similar effective cross-sections to the equilibrium reactor, varying by less than 2%.

6.3.4 Discussion

This SFR design section sought to answer several questions which were outlined in the introduction and discussed below.

Low-CRs from 0.42 to 1.05 were achieved using feasible fuel designs, but this was very dependent on fuel feed and recycling strategy. The softer spectrum of MOX fuelled reactors lead to higher CRs over ZR fuelled reactors in reprocessing scenarios, or when americium was included in the fuel. Equilibrium MOX fuelled reactors had a higher MA content than ZR fuelled reactors. FULL-MOX-TRU results had the highest MA content which was greater than 5%.

CRs were the lowest for plutonium only scenarios (PU). With the addition of americium (AM) CRs increased. With more reprocessing, CRs were higher due to the

Table 6.12: One-group effective cross-sections for major isotopes, given as the percentage difference to equilibrium SFR cross-sections where enrichment was changed for each recycling step. Results are presented for the equilibrium cycle, where the enrichment was not changed with each recycle, and at the beginning of life with the low initial enrichment.

BOE using final enrich for each recycling step					
	Total (%)	Elastic (%)	Inelastic (%)	Fission (%)	Capture (%)
U238	0.01	0.01	-0.02	-0.04	0.03
NP237	0.01	0.01	-0.02	-0.03	0.03
PU238	0.01	0.01	-0.02	-0.01	0.03
PU239	0.01	0.01	-0.02	0.00	0.04
PU240	0.01	0.01	-0.01	-0.03	0.03
PU241	0.01	0.01	-0.02	0.01	0.02
PU242	0.01	0.01	-0.02	-0.03	0.03
AM241	0.01	0.01	-0.02	-0.04	0.03
BOL using initial enrich					
U238	0.09	0.22	-0.67	-2.14	0.44
NP237	0.24	0.31	-0.42	-1.22	1.02
PU238	0.22	0.25	-0.32	-1.57	0.93
PU239	0.22	0.30	-0.46	0.03	1.13
PU240	0.37	0.49	-0.23	-1.03	1.52
PU241	0.23	0.23	-0.49	0.35	0.56
PU242	0.28	0.37	-0.29	-1.29	1.15
AM241	0.24	0.29	-0.51	-1.51	0.82

degradation of the feed vector and an increase in ^{238}U in the core. The degradation of the feed vector was more significant for MOX fuelled reactors as they have a softer neutron spectrum, leading to a lower fission to capture ratio in both TRU nuclides and ^{238}U. As a result, CRs increased considerably for MOX fuelled reactors with reprocessing and the inclusion of americium. Despite changes in CR and reactor design, the TRU consumption rate for PU, AM and TRU reprocessing scenarios were roughly the same, however Δk was lower.

When comparing the FULL SFR equilibrium feed vectors and reactor parameters, results were similar. This was independent of the use of the UK's plutonium stockpile or new build LWR SNF as a fuel feed. With more recycling and no external source of material, the equilibrium fuel vector was more dependant on the reactor design than the initial feed vector. As such, the best case reactor design becomes similar, independent of initial fuel feed.

Results for one-group effective cross-sections were compared at BOL and BOE, showing little variation over a reactors lifetime. Therefore one set of cross-sections is adequate to model a reactor in ORION over the entire fuel cycle.

Further work used NNL codes to extract the effective cross-sections and fluxes from ERANOS output files and put them in an ORION usable format for fuel cycle modelling. Converting output from ERANOS to ORION is discussed in Chapter 4 and results from the fuel cycle models are discussed in Chapter 7, 8 and 9.

6.4 Conclusions

In this chapter ERANOS was benchmarked against previous studies to ensure that ERANOS calculation routes were accurate compared to alternative deterministic codes. Reference SFR designs were modelled in ERANOS using different calculation schemes and verified with the SERPENT Monte-Carlo code. The verification ensured that there were no modelling errors and ERANOS gave accurate results for the problem being considered. The benchmarked ERANOS calculation scheme was then used to optimise SFR designs for the fuel cycle scenarios being considered.

ERANOS agrees well with other deterministic codes presented in the literature for an IAEA benchmark, with ERANOS results within 6% of the average for plutonium

mass change. Low CR reference SFR models were used to compare ERANOS and SERPENT, showing going agreement, within 10% for most plutonium masses, with the exception of ^{240}Pu depletion, the reason for which is unknown. However, the differences in depletion were expected to have a small impact on fuel cycle modelling.

The use of more up to date ENDF/B7 and JEFF-3.1.1 cross-section libraries lead to large discrepancies (up to 1500 pcm) in k_{eff} when compared to JEF-2.2 in SERPENT. However, depletion calculation results did not differ substantially. JEF-2.2 was the only available cross-section library in ERANOS. It was assumed that there would be a slightly different envelope of feasible designs if more up to date cross-section libraries were used, but depletion calculation results would not deviate substantially for the same reactor design.

For both BURNER and equilibrium reactors it was determined that the fastest running Hom-RZ-Diff calculation scheme gave sufficiently accurate results, with heterogeneous cell lattice calculations used for final models. For equilibirum reactors Hom-RZ-Diff gave changes in isotopic masses within 3% of more detailed solver methods. The use of hexagonal 3D geometry or transport solvers leads to longer run times and do not significantly improve inventory or k_{eff} results when compared to SERPENT.

For each fuel cycle scenario being studied an SFR was designed with the lowest possible CRs and highest TRU consumption rate, using ERANOS. Designs were within the limiting factors outlined in Chapter 4. Metallic fuelled reactors have a harder neutron spectrum compared to MOX fuelled reactors, which leads to a smaller proportion of MAs in the fuel at equilibrium and lower CRs. Equilibrium one-group reactor cross-sections were compared to show that one set of equilibrium cross-sections is acceptable to model an SFR in ORION over the entire reactor lifetime. This is because the cross-sections do not change significantly from beginning of life to beginning of equilibrium cycle.

For each ERANOS model effective cross-section and fluxes were extracted for use in ORION fuel cycle models in Chapter 7, 8 and 9.

Key findings:

- ERANOS performs as well as other deterministic codes used in the IAEA benchmark.
- ERANOS and SERPENT results show good agreement.
- The use of more recent ENDF/B7 or JEFF-3.1.1 cross-section libraries, compared to JEF-2.2 library in SERPENT, has a significant impact on k_{eff} but a relatively small impact on fuel depletion.
- MOX fuelled reactors at equilibrium have a greater build-up of MAs in the fuel than in metallic fuelled reactors.
- Effective cross-sections do not vary substantially from beginning of life to beginning of equilibrium for an SFR.

Chapter 7

Plutonium disposition using once-through SFRs

7.1 Introduction

This chapter considers the use of SFRs as once-through BURNERS of the UK's plutonium stockpile. Spent fuel from SFRs was considered as the waste stream, sent to a repository, with no further reprocessing in the fuel cycle. Final material inventories were compared to the direct disposal of the initial stockpile and SNF from a MOX fuelled PWR scenario.

The PRISM SFR has been suggested as a once-through BURNER for UK plutonium disposition as part of the UK's plutonium consultation [1, 2]. PRISM is one of the preferred re-use options that has been outlined by the Nuclear Decommissioning Authority (NDA), in addition to PWR MOX and CANDU MOX. An additional option for disposition of the UK's plutonium stockpile is direct disposal in geological repository, however this is not an NDA preferred option. International use of MOX fuelled PWRs means that the impact of PWR MOX on plutonium inventories is well understood. Research on fast reactor fuel cycles to reduce stockpiled material has focused on closed fuel cycles to maximise reduction. The impact of a once-through SFR BURNER scenario on stockpiled materials is excluded from most studies. The lack of information on once-through SFRs for plutonium disposition is significant, considering that it is one of the options for UK plutonium disposition.

To address this problem SFRs, similar to PRISM, were modelled using the ORION

fuel cycle code, irradiating the UK's plutonium stockpile by 2100. Assessment criteria were developed and used to compare the relative merits of: direct disposal of the plutonium stockpile; PWR MOX; and once-through SFRs. Fuel cycles were assessed on: the final inventory of transuranics to be disposed of; total electricity generated; radiotoxicity lifetime; total radiotoxicity at 1000 years; repository size and the bare sphere critical mass of plutonium in the final inventory.

This chapter aims to answer the following questions:

1. Can once-through SFR BURNER scenarios reduce the UK's plutonium stockpile more than once-through PWR MOX?
2. Is the bare sphere critical mass of plutonium in once-through SFR SNF lower than once-through PWR MOX?
3. Will once-through SFR BURNERS have a significant impact on waste performance factors: repository size (> a 20% reduction); 1000 year radiotoxicity (> factor of 2 reduction), or lifetime of waste (> a factor of 10 reduction)?
4. Does cooling prior to repository emplacement have a significant impact on repository size?
5. Do high-CR SFRs perform significantly worse than low-CR SFRs in terms of the assessment?
6. Will high-CR SFRs produce significantly more electricity from the stockpile than low-CR SFRs? Could this electricity offset construction of LWRs and, in turn, offset the total radiotoxicity and repository size, making high-CR SFRs competitive with low-CR SFRs?

7.2 Methods

This section addresses the set up of the fuel cycle model in ORION, the fuel cycle parameters tested, and reviews the assessment criteria used to measure the performance of each fuel cycle. In this chapter a fuel cycle scenario consists of a fuel feed and a reactor specifically designed for the fuel feed. There are eight once-through SFR fuel cycle scenarios and each scenario has its own reactor design.

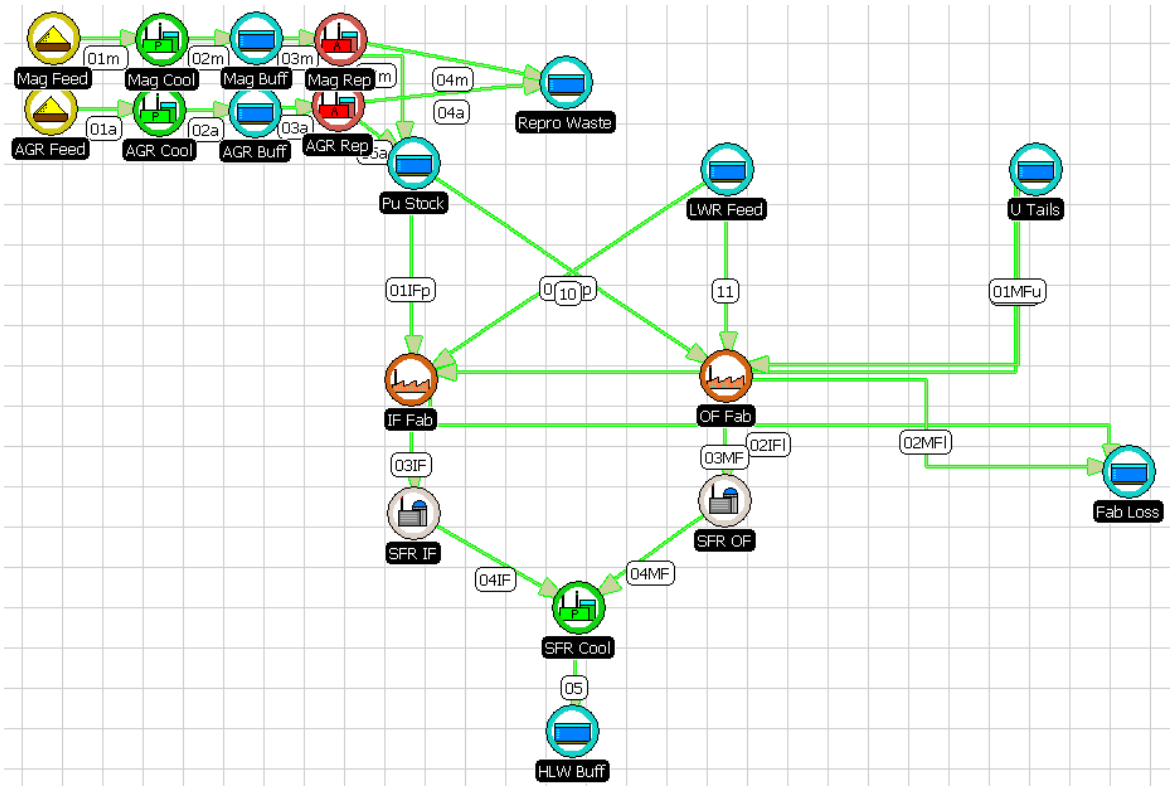


Figure 7.1: ORION model for a once-through SFR BURNER fuel cycle.

7.2.1 ORION model

The once-through SFR fuel cycle set up in ORION is illustrated in Figure 7.1. The fuel feed from AGR and Magnox reactor operating histories was used, as described in Chapter 5, to produce a representative plutonium stockpile in the 'Pu Stock' buffer. 'Pu Stock' was used as the fuel feed with uranium enrichment tails, 'U Tails,' as the carrier feed. Fuel was fabricated for the inner and outer reactor regions and irradiated in 'SFR IF' and 'SFR OF,' using reactor parameters and cross-section from ERANOS, discussed below. Spent fuel was cooled and the final inventory was stored in the 'HLW Buff' buffer.

More detailed discussion of ORION fuel cycle models can be found in Section 4.1.2. More detail on the use of ERANOS to design SFRs can be found in Chapter 6.

7.2.2 Parameters

As defined in this chapter, a BURNER scenario consists of a reactor design and fuel feed, which results in three parameters which can be varied:

- conversion ratio, that is whether the reactor has a high-CR or low-CR;

- fuel matrix, that is, whether the fuel is oxide (MOX) or metallic (ZR) fuel;

- americium in-growth, that is, whether americium is included in the fuel (AM) or whether an additional step to remove americium is included in the fuel cycle scenario (PU).

Note that while reactors are described in terms of high-CR or low-CR, the reactor design for each fuel cycle scenario is different and designed to match the fuel cycle. Therefore CR is used as a reactor descriptor but should not be taken to mean that there are only two reactor designs. There was no reprocessing of SFR fuel in these scenarios, so there were no reprocessing parameters to test. The fuel cycle scenarios were run to 2100 and cooled for a minimum of 50 years prior to disposal.

Reactor parameters and effective cross-sections were taken from the work completed in ERANOS, Chapter 6. The reactor parameters used in ORION are shown in Table 7.1. Each of the reactor models corresponds to a separate fuel cycle modelled in ORION which is described as a re-use fuel cycle scenario in this chapter. In addition to the re-use scenarios, the plutonium stockpile with no reactors was modelled as a direct disposal scenario. The direct disposal scenario was used to compare the impact of reactor scenarios on the plutonium stockpile. In Table 7.1, AM scenarios have the americium in-growth included in the fuel, PU scenarios have americium in-growth removed before fuel fabrication. In each scenario, the total power generated, GWy(e), and the number of reactor modules required to irradiate all stockpiled material was determined using ORION.

Nine re-use fuel cycle scenarios were modelled in total, one using PWR MOX and eight using BURNER SFRs. A stockpile direct disposal scenario was also modelled and used to compare the relative impact re-use scenarios had on final inventory sent to a repository; repository size, and radiotoxicity. The PWR MOX model used cross-sections from an infinite lattice of 100% MOX PWR assemblies. The PWR was not modelled in detail as it was only intended as a high TRL benchmark to compare the performance SFR fuel cycles. The eight BURNER SFR scenarios are described in Table 7.1.

Table 7.1: SFR and PWR MOX parameters used for each of the once-through fuel cycle scenarios. Assumes an 85% capacity factor for SFRs.

BURNER	ZR Lo-CR PU	ZR Lo-CR AM	MOX Lo-CR PU	MOX Lo-CR AM	ZR Hi-CR PU	ZR Hi-CR AM	MOX Hi-CR PU	MOX Hi-CR AM	PWR MOX PU
TRL	6	4	9	5	6	4	9	5	10
CR	0.55	0.70	0.42	0.63	0.94	1.00	0.96	1.09	-
HM (t)	8.75	11.68	7.90	7.90	17.29	17.29	20.74	20.74	37
TRU/HM	29.5	29.8	37.7	38.3	15.5	16.7	16.9	17.0	9.0
GWe	0.38	0.38	0.38	0.38	0.38	0.38	0.38	0.38	0.55
GWt	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.5
# Reac	2	2	2	2	4	4	4	4	2
GWy(e)	35.4	37.7	27.1	28.6	68.6	67.2	59.2	62.1	59.9

7.2.3 Assessment criteria

Fuel cycle scenarios were assessed on the following assessment criteria:

- Total electricity generating capacity, GWy(e);
- Fuel cycle lifetime and the earliest repository date (2150 for every scenario in this chapter);
- Final TRU inventory sent to a repository;
- Lifetime of radiotoxicity;
- Total radiotoxicity at 1000 years, assuming a disturbed repository scenario;
- Repository size estimated by cumulative decay heat (CDH);
- Bare sphere critical mass of final plutonium vector sent to a repository.

Final inventory, generating capacity, total radiotoxicity and decay heat can all be exported from ORION but some post-processing is needed. This is discussed in Chapter 4, with a brief recap below.

The lifetime of radiotoxicity is considered to be the time it takes SNF to decay to the same level as the total natural uranium used to fuel an LWR generating the same amount of electricity. The waste lifetime from each re-use fuel cycle scenario was compared to the same generating capacity of uranium fuelled LWRs, excluding the plutonium stockpile. A significant reduction in the waste lifetime is considered to

be an order of magnitude less than uranium fuelled LWRs [1], that is less than 29,000 years.

The total radiotoxicity 1000 years after disposal represents the potential hazard in a disturbed repository, where 1000 years is assumed to be the earliest timescale for a disturbed repository. A disturbed repository is described as being compromised by a geological event or human intrusion, which could potentially lead to large releases. Repository size is estimated by CDH, which is the integrated decay heat for 1000 years after disposal in a repository. Both CDH and 1000 year radiotoxicity are calculated for a range of cooling times between 50 and 500 years, to see how the decay of short to intermediate lived isotopes influences results over this timescale.

Re-use scenario results for 1000 year radiotoxicity and repository size are presented relative to the stockpile direct disposal scenario. Presenting results relative to the direct disposal scenario shows the relative improvements of re-use scenarios over direct disposal without accounting for other reactors. The operation of SFRs generates electricity (GWy(e)) which could be considered as an alternative to the operation of once-through LWRs to meet the UK's nuclear generation needs. The ability of SFR scenarios to offset once-through LWRs, and thus reduce the amount of LWR SNF generated was considered. As such, 1000 year radiotoxicity and repository size were also presented relative to the stockpile direct disposal scenario, with the addition of LWR SNF corresponding to the same GWy(e) as SFRs in the scenario. Therefore, two sets of 1000 year radiotoxicity and repository size results were presented, one with the offset of LWRs and one without.

Bare sphere critical mass is an indicator of the degradation of the fissile feed vector as a result of burnup. The final inventory TRU vector was taken from ORION and used to calculate mass required to get $k_{eff} = 1$, to the nearest 0.2 kg. The degradation of the isotopic vector is used to indicate the impact of the fuel cycle on the isotopics and the reduced attractiveness of material for potential proliferators.

[1] The waste lifetime for uranium fuelled LWRs is approximately 290,000 years.

7.3 Results

A summary of fuel cycle scenario results have been tabulated in Table 7.4. Summarised results are given as the relative improvement factor over a reference case (e.g. direct disposal or a MOX fuelled PWR) for each of assessment criteria so the relative merits of each scenario can be compared to their TRL.

7.3.1 Masses

The final fuel cycle inventory of transuranics at 2150, after 50 years of cooling, are given in Table 7.2.

PWR MOX reduces the amount of plutonium in the stockpile more than the Low-CR SFR BURNER scenarios. All scenarios increase americium content, however the PWR MOX scenario produces the most americium. The PWR MOX scenario produces more americium than SFR BURNER scenarios as a result of the thermal neutron spectrum in PWRs. Cross-sections in the thermal energy range have a lower ratio of fission to capture than cross-sections in the fast energy range for plutonium, resulting in the build-up of more high mass transuranics [164], particularly ^{241}Am,

$$^{240}\mathrm{Pu} + \mathrm{n} \rightarrow ^{241}\mathrm{Pu} \xrightarrow{\beta\text{-decay}} ^{241}\mathrm{Am}. \tag{7.1}$$

Americium increases the least in BURNER-AM scenarios where americium is included in SFR fuel. The inclusion of americium in SFR fuel does not prevent the build-up of more americium but allows for some transmutation,

$$\begin{aligned} \mathrm{Am}^{241} + \mathrm{n} &\xrightarrow{\text{fission } \sim 14\%} \mathrm{FP} + \mathrm{n} \\ &\xrightarrow{\text{capture } \sim 11\%} \mathrm{Am}^{242m} \xrightarrow{\text{fission}} \mathrm{FP} + \mathrm{n} \\ &\xrightarrow{\text{capture } \sim 75\%} \mathrm{Am}^{242} \xrightarrow{\text{decay } \sim 81/19\%} \mathrm{Cm}^{242}/\mathrm{Pu}^{242}, \end{aligned} \tag{7.2}$$

therefore BURNER-AM scenarios have the lowest final americium inventory. Metallic fuelled, BURNER-ZR scenarios produce less americium than the equivalent BURNER-MOX scenarios as a result of the harder neutron spectrum which leads to a greater ratio of fissions to capture in plutonium nuclei. High-CR BURNER scenarios reduce the plutonium stockpile the least, increasing it in some cases. Plutonium reduction in all Low-CR BURNER scenarios is similar, with the highest and lowest inventories differing by less than 2%.

Table 7.2: Final masses (tHM) of transuranic elements to be sent to a repository after 50 years of cooling in BURNER scenarios.

	Pu Stock	**PWR MOX**	**ZR Lo-CR Pu**	**ZR Lo-CR AM**	**MOX Lo-CR PU**	**MOX Lo-CR AM**	**ZR Hi-CR PU**	**ZR Hi-CR AM**	**MOX Hi-CR PU**	**MOX Hi-CR AM**
TRU	111.80	85.97	98.25	96.63	98.75	97.78	115.79	112.20	116.52	116.68
Pu	105.48	65.71	87.81	88.75	88.37	89.43	104.02	103.80	103.92	107.11
Np	1.20	2.50	1.61	1.27	1.61	1.32	1.69	1.35	1.88	1.45
Am	5.12	17.64	8.82	6.59	8.76	7.02	10.03	6.99	10.68	8.07
Cm	0.00	0.11	0.01	0.02	0.01	0.02	0.05	0.06	0.03	0.04

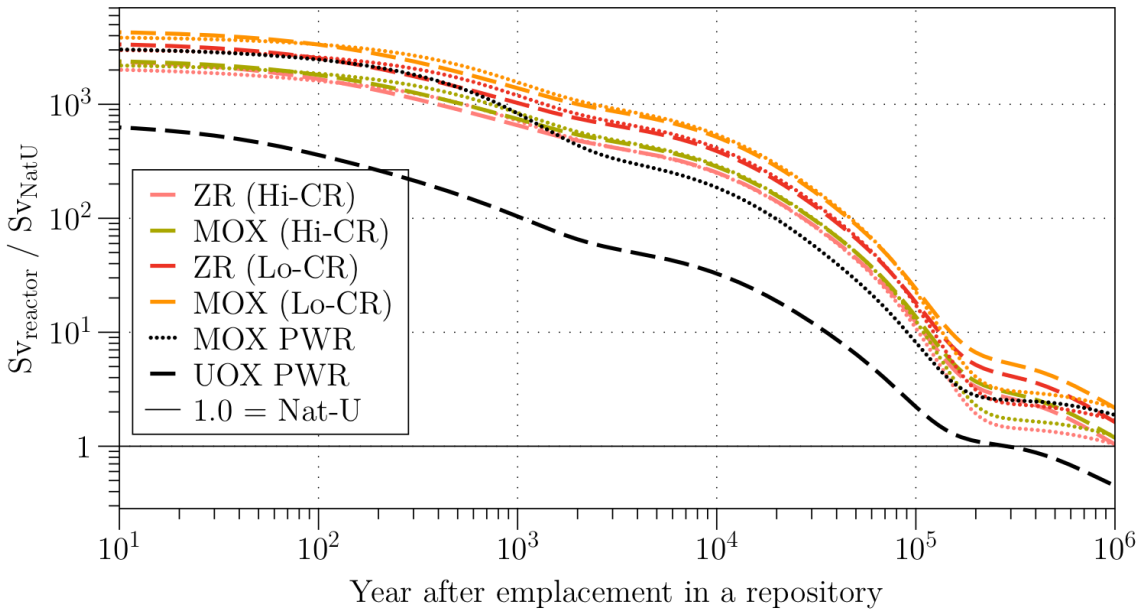


Figure 7.2: Radiotoxicity per GWy(e) for BURNER scenarios after disposal in a repository. Radiotoxicities are given relative to the radiotoxicity of natural uranium required to fuel a UOX PWR. Fine dashed lines represent PU scenarios, thick dashed lines represent AM scenarios.

In terms of the total reduction of TRUs, PWR MOX is much better than once-though SFR BURNER scenarios.

7.3.2 Radiotoxicity

Elemental contributions to radiotoxicity over time are discussed in Section 8.3.2.1. They are not discussed in this chapter as the elemental contributions to once-through BURNER radiotoxicity is similar for all scenarios. The main contributor to radiotoxicity in the first few thousand years is americium, followed by plutonium up to a few hundred thousand years.

7.3.2.1 Radiotoxicity lifetime

The total radiotoxicity per GWy(e) relative to natural uranium for each re-use fuel cycle scenario is presented in Figure 7.2. A UOX fuelled PWR has been included as a benchmark. In this thesis, a significant reduction in the lifetime of radiotoxicity was considered to be an order of magnitude less that UOX PWR, that is, less than 29,000 years.

It is clear that the re-use of the UK's plutonium stockpile in PWRs or SFRs leads to high radiotoxicities. To decay to similar levels as natural uranium will take more than one million years for all re-use scenarios. This decay time is very high compared to PWR UOX fuel, which would take 290,000 years to decay to natural uranium levels. The long lifetime of these scenarios is due to the high radiotoxicity of plutonium, and once-through BURNER scenarios do not significantly reduce the total plutonium inventory in the stockpile.

Radiotoxicity results should ideally include the HLW generated by the AGR and Mangox reactors that produced the plutonium. The total electricity generated by the re-use scenarios should ideally include the electricity generated by Magnox and AGR reactors used to generate the plutonium stockpile. However, additional AGR and Magnox information was not available and the plutonium stockpile assessment in Chapter 5 was not accurate enough to get a good estimate of Mangox and AGR HLW. Without the inclusion of AGR and Magnox reactors, results in Figure 7.2 only give an indication of the lifetime of radiotoxicity to compare scenarios. It is clear that the life-time of radiotoxicity is so long that there is no substantial benefit to a particular BURNER scenario in terms of radiotoxicity.

7.3.2.2 1000 year radiotoxicity

The total radiotoxicity relative to direct disposal scenarios at 1000 years (Sv_{1000}), which is assumed to be the earliest timescale for a disturbed repository, is presented in Figure 7.3. BURNER scenario results are given relative to the stockpile direct disposal scenario, with and without the offset LWR SNF. The change in results over a period of 50 to 500 years shows how the radiotoxicity of different scenarios changes over time. In this thesis a factor of two reduction in 1000 year radiotoxicity is considered to be a significant improvement.

At 50 years PWR MOX and high-CR BURNER-PU scenarios have the highest radiotoxicity, considerably more than the plutonium stockpile, as seen in Figure 7.3, 'SFR / Pu Stockpile'. Low-CR BURNER-AM scenarios have the lowest radiotoxicity, close to that of the stockpile. When LWR SNF offset is included in Figure 7.3 'SFR / (Pu + LWR),' the best and worst scenarios remain the same, however high-CR BURNER and PWR MOX scenarios are closer to low-CR BURNER scenarios as a

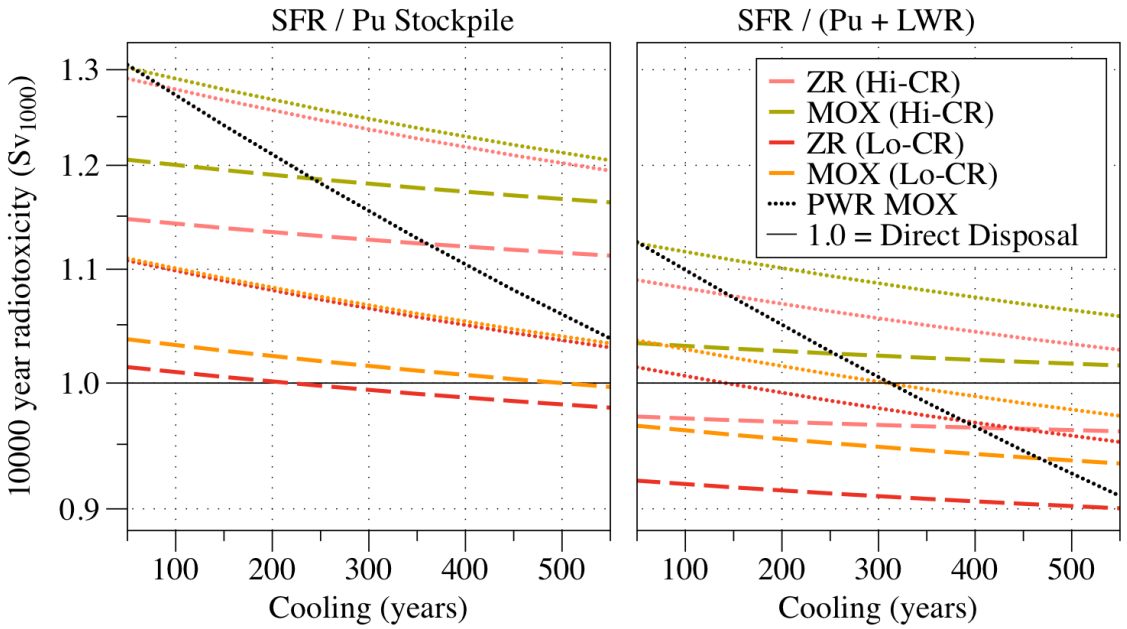


Figure 7.3: 1000 year radiotoxicity (Sv_{1000}) for BURNER scenarios relative to direct disposal scenarios. Results in the left hand figure are given relative to the direct disposal of the UK's plutonium stockpile. Results in the right hand figure are relative to a once-through PWR scenario (the UK's plutonium stockpile plus the same GWy(e) of PWR SNF). Fine dashed lines represent PU scenarios, thick dashed lines represent AM scenarios.

result of greater power generation off setting more LWRs. When the offset of LWRs is considered, the radiotoxicity of low-CR BURNER-AM scenarios is always less than that of the direct disposal scenario.

Over the 500 year timescale in Figure 7.3, the order of results stays approximately the same, that is, the best and worst BURNER scenarios for radiotoxicity increase are unchanged. The radiotoxicity of BURNER-PU scenarios reduces slightly faster than BURNER-AM scenarios over the 500 year period and the radiotoxicity of PWR MOX reduces much faster than all BURNER scenarios. The PWR MOX scenario contains the least plutonium and the most americium. The contribution of plutonium to the radiotoxicity of PWR MOX stays roughly the same from 500 to 2000 years and americium decays away over this timescale, as seen in Figure 7.4. Americium is the major radiotoxic contributor in the PWR MOX scenario up to 1200 years and the plutonium contributions to radiotoxicity are relatively low compared to americium. As a result of the low plutonium contribution and high americium contribution, PWR MOX has the highest radiotoxicity at 1000 years, which decays very quickly to become one of the least radiotoxic scenarios at 1500 years, similar to the BURNER-PU scenarios, as

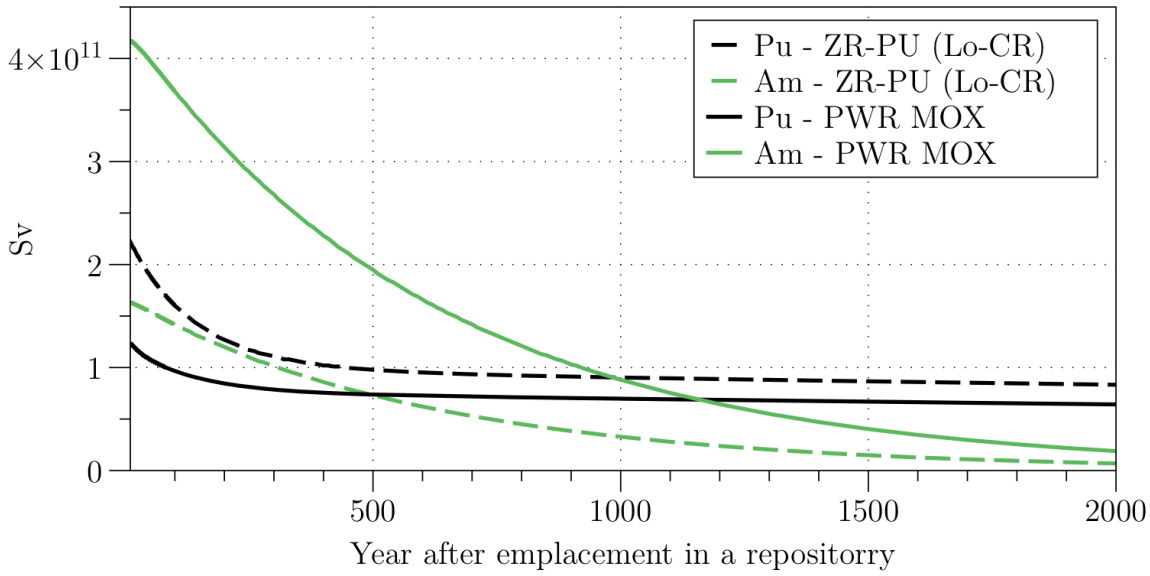


Figure 7.4: Main elemental contributions to total radiotoxicity for 2000 years after disposal of PWR MOX and BURNER-ZR-PU spent fuel.

shown in Figure 7.3.

The differences in 1000 year radiotoxicity between equivalent BURNER-MOX and BURNER-ZR scenarios in Figure 7.3 are a result of the amount of americium generated in each scenario. For low-CR BURNER scenarios, there is no significant benefit in terms of 1000 year radiotoxicity to one scenario over another. The difference between 1000 year radiotoxicity in low-CR BURNER scenarios and direct disposal scenario is within 11%. High-CR BURNER scenarios and PWR MOX scenarios are within 30% of direct disposal scenarios, as shown in Figure 7.3 (12% when offset LWRs are considered).

Overall, the 1000 year radiotoxicity for BURNER scenarios does not vary significantly with different fuel cycle or reactor parameters. Any differences in 1000 year radiotoxicity is a result of different final americium inventories, discussed in Section 7.3.1. Results vary between a 10% decrease in 1000 year radiotoxicity to a 30% increase. A 10 to 30% difference in total radiotoxicity is not significant considering how high the total radiotoxicity is at 1000 years. A much larger variation would be required to produce a meaningful reduction in radiotoxicity.

7.3.3 Repository size

The cumulative decay heat (CDH), of each re-use scenario relative to the stockpile direct disposal scenario, is presented in Figure 7.5. CDH is used to estimate relative repository size. The minimum cooling time was assumed to be 50 years and the longest cooling time considered was 350 years prior disposal. However, results were plotted up to 550 years to see how cooling time and the decay of short to intermediate lived components of SNF influence repository size.

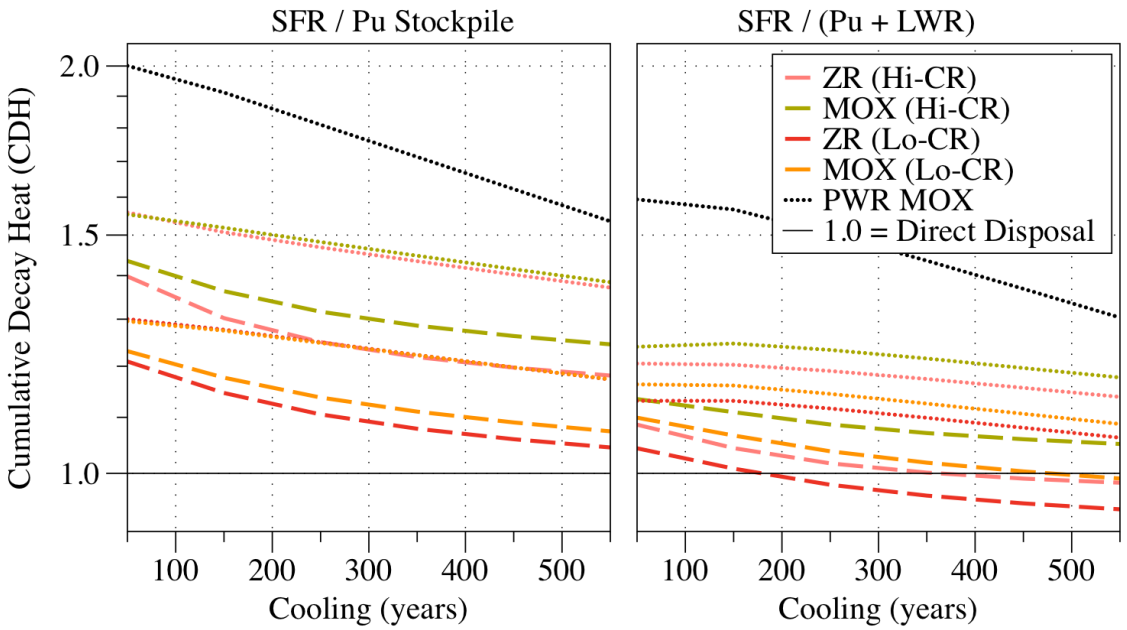


Figure 7.5: Relative repository size for BURNER scanarios estimated by the CDH method and given relative to direct disposal scenarios. Results in the left hand figure are given relative to the direct disposal of the UK's plutonium stockpile. Results in the right hand figure are relative to a once-through PWR scenario (the UK's plutonium stockpile plus the same GWy(e) of PWR SNF). Fine dashed lines represent PU scenarios, thick dashed lines represent AM scenarios.

Comparing re-use scenario repository sizes to the direct disposal of the plutonium stockpile, Figure 7.5, 'SFR / Pu Stockpile', PWR MOX has the largest repository size and low-CR BURNER-AM scenarios have the smallest. However, both PWR MOX and BURNER-AM have larger repositories than the direct disposal scenario. At the shortest cooling time, 50 years, the PWR MOX repository size is twice that of the stockpile disposal scenario. The low-CR BURNER-AM scenario is approximately 20% larger than the stockpile disposal scenario. With 350 years of cooling the repository size of the PWR MOX and low-CR BURNER-AM scenarios are 71% larger and 8% larger

than the stockpile disposal scenario, respectively. The difference between repository sizes is due to the difference in final americium inventories as americium contributes most significantly to decay heat over the 1000 years integrated time period. The cause of different americium inventories in each scenario is discussed in Section 7.3.1.

When comparing re-use scenario repository size to the direct disposal repository size, with the addition of offset of LWR SNF, Figure 7.5 'SFR / (Pu + LWR)', the largest and smallest repositories are PWR MOX and low-CR BURNER-ZR-AM scenarios respectively. With 50 years cooling time the PWR MOX repository size is 60% larger than the direct disposal scenario, and low-CR BURNER-ZR-AM is 4% larger than the direct disposal scenario. With 350 years of cooling prior to disposal PWR MOX is 30% larger and low-CR BURNER-ZR-AM is 6% smaller than the direct disposal scenario. Significantly increased cooling time does not substantially reduce the low-CR BURNER-ZR-AM repository size. The relative repository size for each BURNER scenario, compared to direct disposal, changes when LWR SNF offset is considered, as the relative repository size of each scenario is dependent on the final americium inventory and the total power generated in a scenario. If more power is generated in a scenario, then more LWR SNF is offset reducing the total repository size. As a result, the second best scenario when LWR offset is considered is the high-CR BURNER-ZR-AM scenario which has a very low americium inventory per GWy(e). When the offset of LWRs is considered, all BURNER-AM scenario have smaller repositories than all BURNER-PU scenarios, as BURNER-AM scenarios have lower final americium inventories and generate more power.

As the HLW component of a repository has the largest footprint and therefore the largest cost, larger repositories should be avoided. The PWR MOX scenario is therefore not ideal as it increases repository size more than any other scenario, whereas the low-CR BURNER-AM scenarios increase repository size the least.

If repository size is deemed more important than the re-use of the plutonium stockpile in reactors, then the direct disposal of the stockpile is preferable. If re-use of the stockpile is deemed important whilst minimising the increase in repository size, then low-CR BURNER-AM scenarios are preferable. When the offset of LWRs is considered, BURNER-AM scenarios with prolonged cooling can reduce repository size, however this is a small reduction and can be considered negligible (see Section 7.3.3.1).

Therefore, if the re-use of the plutonium stockpile is deemed important and SFRs are used to offset LWRs, BURNER-AM scenarios can re-use the stockpile with a negligible impact on repository size.

7.3.3.1 Sensitivity of CDH timescale

There is no defined cumulative decay heat integration timescale. It is dependant on waste form design, heat generation and the ability of the host geology to dissipate heat. Previous studies using CDH as an estimate of repository size have used different timescales to integrate decay heat over: 450 years [127], studied by NNL, UK; 1450 years, studied by INL, USA [114,121], and 1900 years in German studies [124]. There is a possibility that if the CDH timescale is altered, a repository size that was estimated to decrease would increase relative to the direct disposal scenario. The sensitivity of the estimated repository size to the selected integrated CDH timescale was investigated to determine the maximum potential increase or decrease in repository size. This was used determine the maximum repository size increases or decreases relative to the direct disposal case under all reasonable CDH timescales. This allowed the author to set a minimum repository size reduction that ensured a reduction in repository size given different CDH periods. The tested CDH periods were 500, 1000, and 1500 years.

The sensitivity study results for an early ORION study of BURNER scenarios, are presented in Figure 7.6, which should not be confused with other results presented in this chapter. Two sample cases were used to give a range of results: PWR MOX which has a large increase in near-term decay heat relative to the direct disposal scenario and BURNER-ZR-AM which has similar decay heat relative to the direct disposal scenario. BURNER-ZR-AM repository size does not vary significantly with CDH period when compared to the direct disposal scenario. PWR MOX varies substantially with CDH period when compared to the direct disposal scenario. For PWR MOX, the difference in repository size between 500 year and 1500 year CDH integration period is approximately 20%. To ensure that a repository size is definitely larger or smaller than the direct disposal scenario it was decided that a 20% increase or decrease in relative repository size was needed.

The integrated decay heat timescale chosen should be based on the time it takes

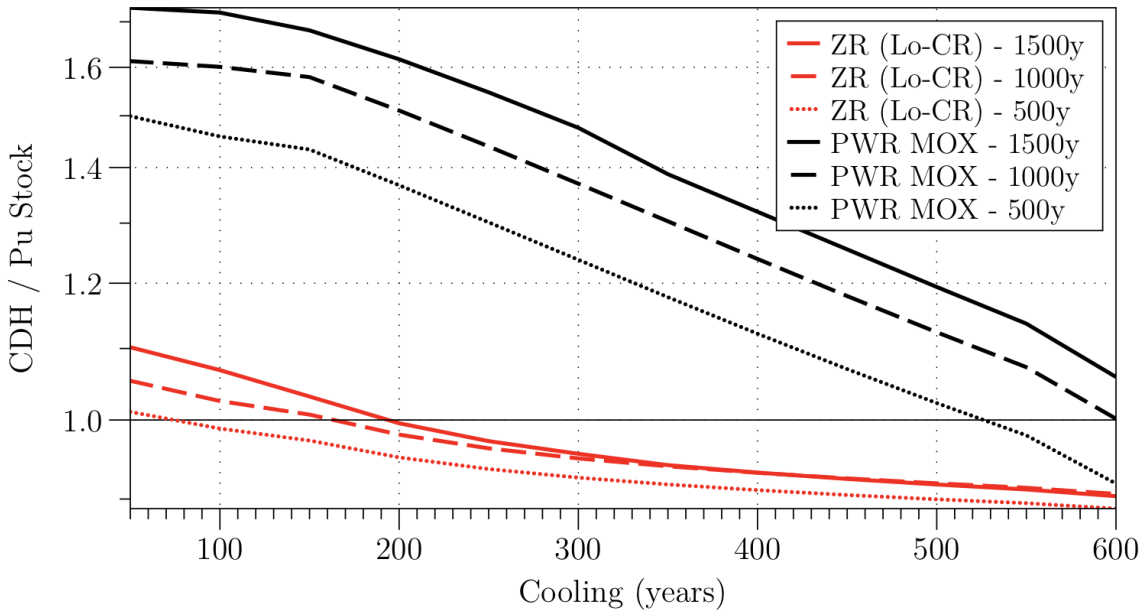


Figure 7.6: Sensitivity of relative repository size estimate to CDH integration period. Sample results show the trend for low-CR BURNER-ZR-AM and PWR-MOX repository size over 600 years of cooling. CDH was integrated over 500 years, fine dashed line; 1000 years, thick dashed line, and 1500 years, solid line.

to reach peak repository temperature [114] as a result of geology and repository loading. As there is no repository design for the UK, deciding a timescale for decay heat integration is difficult. The UK specific NNL value of 450 years [127] was considered, however it is a substantially shorter timescale than the international CDH timescales. As no explanation was provided for this short timescale it was rejected for this work. The 1000 year timescale for decay heat integration was chosen as it was an intermediate value between the UK and international studies. However, as discussed, an improvement factor difference of at least 20% is required to be considered a significant change, as there are many outstanding unknowns in UK repository design, such as geology and loading.

7.3.4 Bare sphere critical mass

The bare sphere critical mass for the plutonium vector in the final inventory, if reprocessed, is presented in Table 7.3.

The greatest improvement in bare sphere critical mass occurs for PWR MOX. The thermal spectrum of PWRs leads to a substantial degradation in the plutonium vector.

Table 7.3: Bare sphere critical mass (kg) for plutonium vector in final inventories of BURNER scenario SNF, after 50 years of cooling.

	Pu Stock	**PWR MOX**	**Zr PU**	**Zr AM**	**MOX PU**	**MOX AM**
Low-CR	12.8	14.8	13.6	13.5	13.6	13.7
High-CR	-	-	13.4	13.3	13.4	13.3

Low-CR SFR BURNERS do not significantly alter the critical sphere mass and high-CR SFR BURNERS reduce it even less. One pass through an SFR BURNER leads to a very small reduction in both plutonium mass and has very little impact on the isotopic vector. For fuel in an SFR, ^{238}U and all TRUs undergo fission, with a high ratio of fission to capture in the fuel. For fuel in a PWR MOX assembly, ^{239}Pu and ^{241}Pu fission reactions are most common, in other plutonium nuclei capture is more likely. The lower ratio of fission to capture in PWRs than SFRs, leads to a more significant degradation of the PWR isotopic vector.

7.4 Discussion

A summary of results for the key assessment factors can be found in Table 7.4. Numerical improvement factors over reference scenarios were tabulated to compare the fuel scenarios across all of the assessment factors. This chapter sought to answer several questions which were outlined in the introduction and discussed below.

PWR MOX reduces the total stockpile mass more than the best SFR BURNER scenario. PWR MOX has an improvement factor of 1.30 for total transuranic inventory, compared to 1.16 for the best case SFR BURNER scenario. The final plutonium vector from PWR MOX is more degraded than SFR BURNER scenarios, with an improvement factor of 1.86 for PWR MOX compared to 1.28 for the best case SFR BURNER scenario. There is no significant improvement in radiotoxicity or repository size for any scenario. Low-CR BURNER-ZR-AM scenario has the only improvement in repository size, which is 1.04, and the largest improvement in 1000 year radiotoxicity, 1.10, which requires 350 years of cooling (50 years of cooling is included in Table 7.4, 350 years is not) and includes LWR offset. Low-CR SFR BURNER scenarios generally performed better than the PWR MOX scenario in terms of radiotoxicity and

Table 7.4: Using the UK's plutonium stockpile as a fuel feed, the BURNER fuel cycle scenario improvement factors are presented relative to a [reference case] for each assessment criteria. Improvement factors greater than one show a positive improvement. Improvement factors less than one show a dis-improvement.

		Pu Stock	**PWR MOX**	**Zr Lo-CR PU**	**Zr Lo-CR AM**	**MOX Lo-CR PU**	**MOX Lo-CR AM**	**Zr Hi-CR PU**	**Zr Hi-CR AM**	**MOX Hi-CR PU**	**MOX Hi-CR AM**
TRL		-	10	6	4	9	5	6	4	9	5
GWy(e) [PWR MOX]		-	1.00	0.59	0.63	0.45	0.48	1.15	1.12	0.99	1.04
Mass [Stock]	TRU	1.00	1.30	1.14	1.16	1.13	1.14	0.96	-1.00	0.96	0.96
CDH [Stock]	50y	1.00	0.50	0.77	0.83	0.77	0.81	0.64	0.71	0.65	0.69
CDH [Stock+LWR]	50y	1.00	0.63	0.88	0.96	0.86	0.91	0.83	0.92	0.81	0.88
Sv_{1000} [Stock]	50y	1.00	0.76	0.90	0.99	0.90	0.96	0.78	0.87	0.77	0.83
Sv_{1000} [Stock+LWR]	50y	1.00	0.88	0.99	1.09	0.96	1.04	0.92	1.03	0.89	0.97
Sv Lifetime [UOX LWR]		-	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10
Critical mass [Stock]		1.00	1.86	1.28	1.25	1.27	1.26	1.06	1.06	1.06	1.02

repository size. In all assessment criteria high-CR SFR BURNER scenarios performed worse than the equivalent low-CR scenario. However, if the offset of LWRs is included then the difference between low-CR BURNER and high-CR BURNER results is not significant as roughly twice the electricity is generated, leading to a larger LWR offset.

The PWR MOX scenario has many advantages over SFR BURNER scenarios. It has a higher TRL and reduces the stockpile mass and plutonium stockpile attractiveness more than once-through SFR BURNER scenarios. The stockpile is reduced less in SFR scenarios as there is more plutonium production as a result of ^{238}U capture in a fast reactor. In a fast reactor more ^{241}Am and ^{238}U fission occurs than in a PWR, which contributes to the total burnup. The attractiveness of the final stockpile is reduced more in the PWR MOX scenarios as the ratio of fission to capture is lower than in an SFR, resulting in a greater proportion of heavy plutonium isotopes and a lower proportion of ^{239}Pu.

All advantages that SFR BURNER scenarios have over the PWR MOX scenario is a result of the lower final americium inventory. SFR BURNERS have lower 1000 year radiotoxicity and repository sizes than the PWR MOX scenario. However, SFR BURNERS have minimal advantages over direct disposal for 1000 year radiotoxity and repository sizes. Americium increases in all scenarios, but increases the least in SFR BURNER-AM scenarios, which includes, americium in the fuel, as some transmutation occurs. As americium has the greatest impact on radiotoxicity and decay heat, the inclusion of americium in fuel is vital to achieve the lowest radiotoxicity and repository size results for SFR BURNER scenarios. However, as americium increases in all re-use scenarios, the radiotoxicity and repository size of re-use strategies is generally greater than direct disposal. Americium is also the reason why metallic fuelled scenarios, BURNER-ZR, perform marginally better than BURNER-MOX scenarios in terms of radiotoxicity and repository size. The harder neutron spectrum in metallic fuelled reactors leads to a greater fission to capture ratio in ^{240}Pu and therefore less of a build-up of ^{241}Am. A PWR's thermal neutron spectrum results in a very low ratio of fission to capture in ^{240}Pu, producing a large amount of ^{241}Am, which is why PWR MOX has the largest repository size and greatest radiotoxicity at 1000 years.

There are several limitations of the work presented in this chapter. As previously discussed, the calculation of repository size has several limitations and to ensure a

real increase or decrease in repository size, relative to the direct disposal scenario, there must be an increase or decrease in improvement factor of more than a 20% . The radiotoxicity lifetime is used only as an indication of lifetime, as it does not account for AGR and Magnox HLW or electricity generation. The use of bare sphere critical mass as an indicator of material attractiveness, whilst relevant, is only one factor in proliferation resistance related to the ability to cause a criticality. It does not account for the weapons usability of material. In addition, the attractiveness of a material stockpile for proliferation should include several other intrinsic factors: the mass of material needed; the ability to handle material in terms of dose and shielding requirements, and the processing requirements such as the extraction of FPs, or reduction of oxides to a metal. These aspects are discussed in Appendix D, and concluded that for a sophisticated state, such as the UK, intrinsic barriers are not significant barriers to impede proliferation.

7.4.1 Recommendations and further work

The NDA's preferred options for UK plutonium disposition are re-use scenarios [1]. Two of the credible re-use strategies outlined by the NDA are a MOX fuelled PWR and the PRISM SFR (once-through BURNER SFR). The results presented here are relevant to the UK's plutonium disposal strategy as PWR MOX and SFR BURNER scenarios have been modelled to compare their relative merits. Several parameters were varied in SFR BURNER reactor design to assess the important parameters in SFR BURNER design. The significant results that separate scenario performance are TRL, final inventory mass, repository size and bare sphere critical mass. The impact on radiotoxicity lifetime and 1000 year radiotoxicity is sufficiently small to not influence fuel scenario selection. Different priorities in UK nuclear policy will lead to different scenarios being favourable. If the re-use of plutonium is a priority and repository size is not a priority then PWR MOX is the best scenario as a result of its very high TRL. If maximum stockpile reduction and reducing the attractiveness of the stockpile are priorities then PWR MOX is the best scenario to meet these goals. If the re-use of plutonium and repository size are priorities then low-CR SFR BURNER-AM scenarios are the best option, as plutonium is re-used with a minimal impact on repository size. It is important that americium is included in SFR fuel and a low-CR

SFR design is used to minimise the increase in repository size. The choice of metallic or MOX SFR fuel has little impact, so higher TRL MOX fuel would be preferable. If re-use is no longer a preferred option and stockpile reduction is not a priority, then the direct disposal of the UK's stockpile would be the best option as it has the lowest repository size and avoids the expensive construction and operation costs of an SFR or PWR MOX reactor.

According to Eurobarometer polls, the three main factors that influence public perception of nuclear power are: terrorism concerns; waste disposal and proliferation, in that order [4]. The most important factor, terrorism, cannot easily be prevented by fuel cycle scenarios but waste disposal and proliferation can. Separated plutonium, such as the UK's plutonium stockpile, is a large proliferation concern. To tackle proliferation concerns re-use of the UK's plutonium stockpile, which puts plutonium out of reach in a highly active matrix, would be preferable. PWR MOX would be the best option to reduce proliferation concerns, reducing the attractiveness of the stockpile more than alternative scenarios. In addition to reducing the proliferation risk, the high TRL of PWR MOX means the deployment timescale would be short. In terms of waste, all scenarios studied in this chapter have very long waste lifetimes and a minimal impact on 1000 year radiotoxicity and repository size. To increase public support for nuclear power the lifetime of waste, 1000 year radiotoxicity and repository size should be minimised which is not possible with once-through re-use scenarios. There is significant evidence that large improvements in waste disposal factors can be achieved by using closed SFR fuel cycles dedicated to burning TRUs [113, 114, 115, 121, 124, 135, 165]. Previous studies have shown improvements in waste lifetimes and repository sizes of an order of magnitude over the direct disposal of stockpiled material. Closed SFR fuel cycles dedicated to burning the UK's plutonium stockpile will be considered in Chapter 8.

7.5 Conclusions

This chapter modelled once-through fuel cycles, used to irradiate the UK's plutonium stockpile without further reprocessing. Scenario aims were to reduce the stockpile size and minimise the burden on a repository. PWR MOX scenarios are represented in

the literature but once-through SFR scenarios are not. Once-through SFR BURNER scenarios were compared to once-through PWR MOX scenarios as there have been no previous comparison in the literature. Both scenarios are under consideration for UK plutonium stockpile disposition and comparing their relative performance may prove useful in selecting a preferred option.

Eight SFR re-use scenarios were considered, where CR and fuel composition were varied. Fuel composition describes the fuel matrix, that is MOX or metallic (ZR) fuel and whether americium in-growth was included or removed. These scenarios were compared to a PWR MOX scenario and the direct disposal of the current plutonium stockpile. All once-through SFR and PWR scenarios considered in this chapter increase the repository size relative to direct disposal. The PWR MOX and low-CR SFR BURNER scenarios reduced the total plutonium stockpile by up to a factor of 1.3 and 1.16 respectively. High-CR once-through SFR BURNER scenarios increase the total plutonium stockpile, with an improvement factor of 0.96.

From all scenarios considered in this chapter, the PWR MOX scenario reduced the total plutonium stockpile mass more than any once-through SFR BURNER scenario. However, SFR BURNER scenarios had smaller repository sizes and a lower 1000 year radiotoxicity than the PWR MOX scenario, with improvement factors as high as 1.09 for SFR BURNERS compared to 0.88 for PWR MOX. Relative to direct disposal of the plutonium stockpile, all scenarios increased the repository size and 1000 year radiotoxicity. The best case SFR BURNER scenario, in terms of waste performance, had the lowest final inventory of americium, favouring low-CR, metallic fuelled SFRs with americium included in the fuel feed, BURNER-ZR-AM. When the power generated by re-use in a scenario is considered to offset LWR operation, high-CR BURNER scenarios generate enough electricity to become competitive with low-CR BURNER scenarios, in terms of repository size, with improvement factors of 0.92 compared to 0.96 for low-CR BURNERS. Given prolonged cooling and the offset of LWRs, the low-CR BURNER-AM scenarios show small but not significant improvements over the direct disposal scenario in terms of repository size and 1000 year radiotoxicity. Therefore BURNER-AM scenarios are the only scenario considered that can re-use the plutonium stockpile and prevent an increase in repository size.

Results in this chapter are relevant to the UK as SFR BURNERS, such as the

PRISM reactor, and PWR MOX scenarios are under consideration for UK plutonium disposition. This chapter shows that PWR MOX has a higher TRL and will reduce the stockpile mass and attractiveness of stockpiled material more than once-through SFR BURNERS. However, PWR MOX will increase the repository size compared to direct disposal of the UK's plutonium stockpile. SFR BURNER scenarios, on the other hand, increase the size of a repository but not significantly and less than the PWR MOX scenario. To make SFR BURNER scenarios competitive, americium should be included in the fuel and a low-CR reactor must be used. This requires high fuel enrichment, close to the limitations of the fuel. However, the choice of metallic or MOX SFR fuel has little impact on disposal outcomes and the higher TRL of MOX fuel is favourable. The NDA's preferred options for plutonium disposition are re-use strategies over direct disposal. Therefore the choice between PWR MOX or a low-CR SFR BURNER-AM scenario is related to the relative importance given to TRL, final mass, material attractiveness, and repository size.

Based on the literature, waste disposal and proliferation are the two main public concerns, related to the nuclear industry, that can be influenced by the fuel cycle. To minimise proliferation concerns, plutonium disposition using PWR MOX in the shortest feasible timescale is preferable. Improving public perception regarding waste disposal requires significant reductions in repository size and radiotoxicity which the once-through BURNER scenarios considered in this chapter are unable to provide (however, they do perform better than PWR MOX). Greater improvements in waste performance would be possible with a closed fuel cycle and more SFRs, which are recommended for further study. As such, closed SFR fuel cycles are considered in Chapter 8 to determine the impact they have on waste performance and the relative merits of developing closed fuel cycles over once-through SFR BURNER fuel cycles.

The key limitations of the results presented in this chapter are the approximation method used for repository size and radiotoxicity lifetime. Repository size estimated by the cumulative decay heat method is a useful estimate for comparing different scenarios but does not accurately model the loading of a repository. The key limitations of the fuel cycle assessment are the lack of quantitative assessment of proliferation, development needs and cost. Proliferation resistance of the fuel cycle is an important factor and is not accounted for in the assessment criteria. TRL was used to infer the

development requirements of the fuel cycle and is a general approximation that does not fully capture the large risk and cost of lower TRL technology. Equally, potential bottle necks in technology development have not been assessed.

Key findings:

- Direct disposal leads to a smaller repository and lower 1000 year radiotoxicity than any once-through scenario.
- Once-through fuel cycle scenarios have a minimal impact on radiotoxicity lifetime.
- PWR MOX reduces plutonium stockpile mass and attractiveness more than SFR BURNER scenarios.
- PWR MOX increases repository size more than any SFR BURNER scenario.
- The best case SFR BURNER scenario has a similar repository size to the direct disposal scenario. It requires a low-CR reactor design and the inclusion of americium in the fuel (BURNER-AM).
- There is little difference between metallic fuelled (ZR) and MOX fuelled SFRs in terms of assessment criteria, therefore higher TRL MOX fuel is preferable.
- The choice between PWR MOX and a once-through BURNER-AM scenarios depends on the relative importance assigned to TRL, stockpile reduction and repository size.
- High-CR SFRs used as plutonium stockpile BURNERS generate enough electricity to offset LWRs and, as a result, become competitive in terms of repository size with low-CR BURNER scenarios. However, low-CR BURNERS still perform marginally better.

Chapter 8

Plutonium disposition using SFR closed fuel cycles

8.1 Introduction

This chapter considers the use of SFRs in a closed fuel cycle, and their ability to reduce the UK's plutonium stockpile.

The PRISM SFR has been suggested as a once-through BURNER for UK plutonium disposition as part of the UK's plutonium consultation [1, 2]. A PRISM-type SFR fuel cycle was modelled in Chapter 7 and had a limited impact on the plutonium stockpile by 2100. Previous studies have looked at fast reactor closed fuel cycles to reduce stockpiled material. Such studies have focused on high TRU content fuels in advanced reactors and fuel cycle concepts to maximise stockpile reduction [122,124,129]. Advanced concepts have low TRLs, but have the potential to achieve significant stockpile reductions in short timescales, less than 300 years. Most research on closed SFR fuel cycles for stockpile reduction assumes the phase out of nuclear power. The ability of SFRs to reduce a stockpile and offset thermal reactors is excluded from most studies.

Reprocessing and a greater number of SFRs in the reactor fleet would improve plutonium stockpile reduction over a once-through BURNER scenarios. To quantify this, reprocessing was included in SFR fuel cycle scenarios and SFR deployment was extended to irradiate all plutonium by 2150.

In previous studies of stockpile reducing fuel cycle scenarios, low TRL fuel cycles are

used with advanced reactor concepts [122, 124, 129]. Low TRL fuel cycles in previous studies use ADS reactors or very high TRU loading with short reprocessing times to give a short timescale for stockpile reduction whilst placing a significant burden on technology development. There is a significant gap in the literature with respect to the effects of higher TRL fuel cycle concepts on stockpile reduction and timescales. To address this gap, a range of fuel cycle and SFR parameters were tested, ranging from high TRL to low TRL concepts. High and low TRL fuel cycle scenarios were assessed to determine how they impacted on timescales and total plutonium stockpile reduction.

The ability of SFRs to offset LWRs, and reduce the stockpile, is significant in a UK context where new build LWRs are being considered along side plutonium stockpile reduction. This chapter specifically looks at how SFRs affect stockpile reduction in a scenario where nuclear power generation is continued. With specific nuclear generating targets in the UK, the operation of SFRs would mean fewer new build LWRs would be required to meet the 16 GWe 2050 target, thus reducing the amount of LWR SNF to be disposed of. To address this, SFR fuel cycles were compared to two reference cases: the stockpile direct disposal scenario, and the stockpile direct disposal scenario with the addition of the same generating capacity of once-through LWRs.

This chapter aims to answer the following questions:

1. How much of an improvement will running SFRs with reprocessing until 2150 (FEED scenarios) have over once-through BURNERS?

2. Does the reprocessing of curium have an impact on results up to 2150?

3. Given reprocessing until a maximum stockpile reduction is achieved (FULL scenarios), what is the maximum improvement over a direct disposal scenario, and over what timescale?

4. How do the WASTE stream only results compare to equivalent FULL scenarios?

5. Do plutonium recycling scenarios (PU scenarios) have any impact on radiotoxicity or repository size compared to direct disposal scenarios?

8.2 Methods

This methods section will discuss two things, the set up of the fuel cycle model in ORION and the fuel cycle variables that were tested. In addition, the assessment criteria used to measure the performance of each fuel cycle is summarised in Section 7.2.3. ORION was used to test a range of reactor designs developed in ERANOS for a closed SFR fuel cycle. Each varied parameter has its own fuel cycle scenario which was modelled in ORION.

8.2.1 ORION model

The closed fuel cycle set up in ORION is illustrated in Figure 8.1. The fuel feed from AGR and Magnox reactor operating histories was used, as described in Chapter 5, to produce a representative plutonium stockpile in the 'Pu Stock' buffer. 'Pu Stock' was used as the fuel feed with uranium enrichment tails, 'U Tails,' as the carrier feed. Fuel was fabricated for the inner and outer reactor regions and irradiated in 'SFR IF' and 'SFR OF' using reactor parameters, cross-section and fluxes from ERANOS, discussed below. Spent fuel was cooled and reprocessed in 'SFR Rep,' sending the recycled elements to 'Rep Feed' and waste to the 'HLW Buff' buffer. When there is enough fissile material available, the 'Rep Feed' buffer becomes the primary fuel feed, ahead of 'Pu Stock.' If there is a lack of material to fabricate fuel from 'Rep Feed' it is topped up with material from 'Pu Stock.' More detailed discussion of ORION fuel cycle models can be found in Section 4.1.2.

Two types of fuel cycle scenario were modelled, with sub-scenarios described in the following section:

- FEED scenarios – Operating enough SFRs to irradiate all stockpiled material by 2150, Figure 4.5;
- FULL scenarios – Operating several generations of SFRs, reducing the number of reactors in each generation due to the reduction of fuel in the fuel cycle, Figure 4.5. The number of SFRs was reduced in each generation until the maximum stockpile reduction was achieved and no more reactors could be operated.

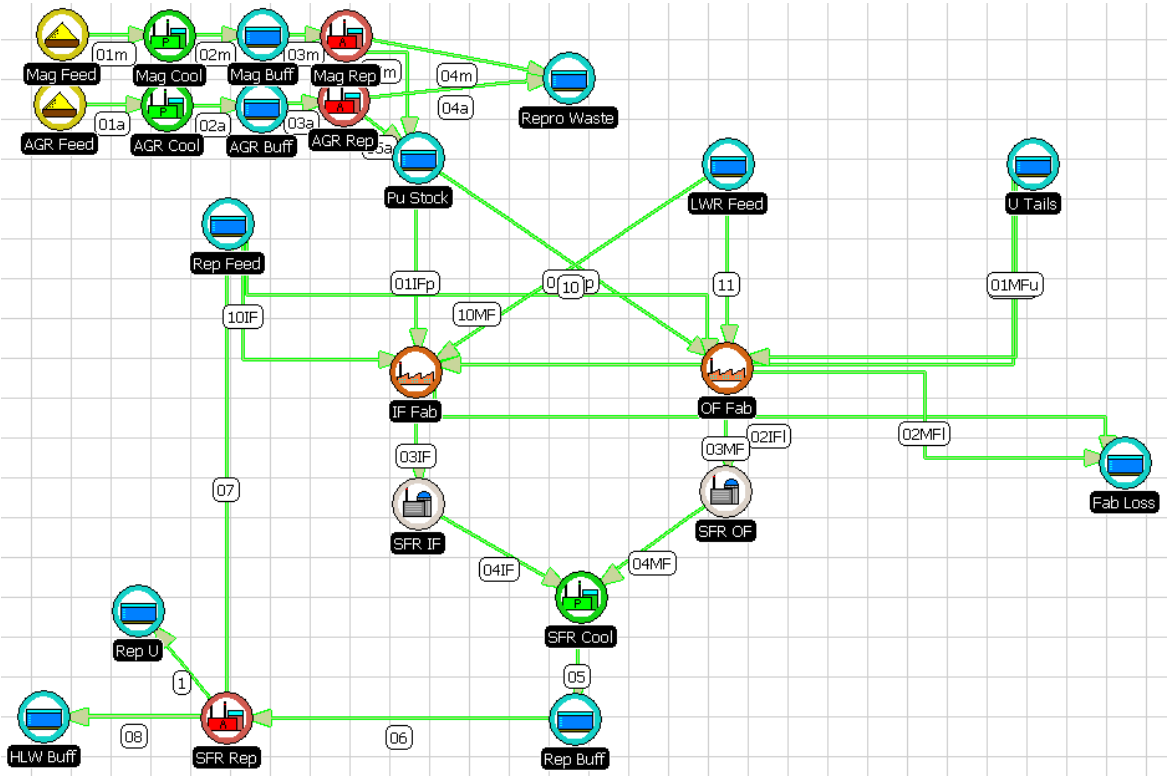


Figure 8.1: ORION model for a closed SFR fuel cycle using the UK's plutonium as a fuel feed.

8.2.2 Parameters

The fuel cycle variables tested were based on different reactor designs and reprocessing options. A total of 24 fuel cycle scenarios were modelled using:

- Two timescales – FEED scenarios modelled until 2150, and FULL scenarios were modelled until material ran out (end dates in Table 8.1);
- Three reprocessed fuel feeds – Removal of americium in-growth and reprocessing of plutonium (PU scenarios), plutonium and americium reprocessing (AM scenario), or all transuranic reprocessing (TRU scenario).
- Two reprocessing variables – High cooling times and high losses (HI scenario), or low cooling times and low losses (LO scenario), Table 4.3;
- Two reactor designs – Metallic (Zr) and MOX fuelled reactors were designed and tested for each scenario. Reactor parameters and self-shielded cross-sections were taken from the work completed in ERANOS, Chapter 6. The reactor parameters used in each ORION scenario are shown in Table 8.1 for FEED and FULL scenarios.

Table 8.1: SFR parameters used for FEED and FULL fuel cycle models with the UK's plutonium stockpile as a fuel feed. Assumes an 85% capacity factor for SFRs.

FEED Scenario	**Zr PU**	**Zr AM**	**Zr TRU**	**MOX PU**	**MOX AM**	**MOX TRU**
TRL	6	4	3	7-8	5	3
CR	0.70	0.85	0.86	0.64	0.90	0.91
TRU/HM (%)	30.0	30.0	30.0	38.4	38.4	38.4
MA/HM (%)	-	1.9	2.3	-	2.9	3.5
GWe	0.38	0.38	0.38	0.38	0.38	0.38
GWt	1.0	1.0	1.0	1.0	1.0	1.0
Cycle length (EFPD)	227	249	249	212	211	211
Lo GWy(e)	164.4	210.9	214.8	147.9	182.5	189.3
Hi GWy(e)	116.6	136.3	135.7	102.4	113.0	115.6
FULL Scenario	**PU**	**AM**	**TRU**	**PU**	**AM**	**TRU**
TRL	6	4	3	7-8	5	3
CR	0.73	0.96	0.95	0.8	1.05	1.04
TRU/HM (%)	30	30	30	38.4	38.4	38.4
MA/HM (%)	-	2.6	3.4	-	4.3	5.5
GWe	0.38	0.38	0.38	0.38	0.38	0.38
GWt	1.0	1.0	1.0	1.0	1.0	1.0
Cycle length (EFPD)	284	307	307	301	326	326
Lo GWy(e)	348.8	503.9	542.6	310.1	426.4	387.6
Lo End Date	2340	2355	2415	2340	2350	2350
Hi GWy(e)	271.3	368.2	387.6	232.6	290.7	290.7
Hi End Date	2400	2400	2460	2340	2400	2460

Each fuel cycle scenario was modelled in ORION, with reactor parameters and self-shielded cross-sections taken from the work completed in ERANOS, Chapter 6.

The FEED and FULL closed fuel cycle scenarios have twelve sub-scenarios, six based on reactor design and fuel feed, Table 8.1, with two variations based on reprocessing losses and cooling time. A stockpile direct disposal scenario was also modelled to compare the relative impact SFR scenarios had on the final inventory sent to a repository, repository size and radiotoxicity.

8.3 Results

A summary of fuel cycle scenario results have been tabulated in Table 8.5. Summarised results are given as the relative improvement factor over a reference case (e.g. direct disposal, PWR MOX) for each of assessment criteria so the relative merits of each scenario can be compared to their TRL.

8.3.1 Masses

The final fuel cycle inventory of TRUs sent to a repository, after 50 years of cooling, are given in Table 8.2 for each fuel cycle scenario.

The greatest overall TRU mass reductions were in scenarios where there was more recycling of TRUs through a low CR SFR. Each pass through an SFR leads to fission and transmutation, therefore more recycling through an SFR leads to a larger overall reduction in stockpiled material. LOW scenarios reduce the stockpile more than HIGH scenarios as a result of shorter cooling times in LOW scenarios which, leads to less fissile material being held up in cooling buffers, and more fissile material available to fuel more reactors. Therefore, more SFRs are operated in LOW scenarios than HIGH scenarios and the total stockpile is recycled through SFRs a greater number of times. For LOW scenarios, three years of fuel is held in the fuel cycle (1 year for cooling, 1 year for reprocessing and fabrication, and 1 year in reactor), for HIGH scenarios thirteen years of fuel is needed (10 year for cooling, 2 year for reprocessing and fabrication, and 1 year in reactor). HIGH scenarios also lead to more americium building up than the equivalent LOW scenarios, as a result of longer cooling times allowing more ^{241}Pu to decay to ^{241}Am prior to irradiation in a reactor.

Considering the FEED scenarios, plutonium reduction is maximised in FEED-PU scenarios. FEED-MOX scenarios reduce the plutonium stockpile more than equivalent FEED-ZR scenarios due to higher enrichments and lower fuel volumes in FEED-MOX SFRs (meaning less fertile ^{238}U in the fuel), resulting in a lower conversion ratio. FEED-PU scenarios reduce the plutonium stockpile more than equivalent FEED-AM or FEED-TRU scenarios, as a result of a lower fuel volume which reduces the mass of ^{238}U and therefore reducing breeding. Americium in the fuel in FEED-AM and FEED-TRU scenarios also leads to the generation of more curium which decays to plutonium.

Table 8.2: Final masses (tHM) of transuranic elements to be sent to a repository after 50 years of cooling in FEED, FULL and WASTE scenarios using the UK's plutonium stockpile as a fuel feed.

	Zr Lo-PU	**Zr Lo-AM**	**Zr Lo-TRU**	**MOX Hi-PU**	**MOX Hi-AM**	**MOX Hi-TRU**	**Zr Lo-PU**	**Zr Lo-AM**	**Zr Lo-TRU**	**MOX Hi-PU**	**MOX Hi-AM**	**MOX Hi-TRU**
FEED Scenario												
TRU	58.09	51.87	50.02	55.92	48.67	46.96	75.55	72.72	70.82	74.53	70.74	69.95
Pu	39.48	45.17	44.13	35.60	41.66	40.68	56.89	64.20	62.86	54.00	61.32	60.96
Np	2.73	1.05	0.44	2.80	0.98	0.44	2.89	1.30	0.79	3.08	1.33	0.83
Am	15.73	5.04	5.04	17.36	5.30	5.33	15.71	6.99	6.97	17.38	7.84	7.95
Cm	0.15	0.61	0.41	0.17	0.72	0.51	0.07	0.24	0.19	0.07	0.25	0.22
FULL Scenario												
TRU	43.07	21.34	8.71	45.26	25.81	16.03	56.22	30.84	18.38	59.90	36.22	23.1
Pu	13.23	17.48	7.65	12.57	21.47	13.56	20.92	24.63	15.37	22.32	28.83	19.17
Np	8.14	1.39	0.13	8.54	1.44	0.20	11.25	2.23	0.43	10.29	2.62	0.54
Am	21.44	0.94	0.76	23.87	1.57	1.97	23.88	3.12	2.27	27.12	4.12	3.13
Cm	0.26	1.53	0.16	0.29	1.33	0.29	0.17	0.86	0.31	0.17	0.66	0.30
WASTE Stream Only												
TRU	33.45	14.03	2.28	36.60	14.04	2.22	37.70	9.35	3.44	39.73	9.19	3.56
Pu	4.06	10.97	1.99	4.40	11.16	1.93	3.02	5.94	2.48	2.99	5.58	2.49
Np	8.11	1.34	0.08	8.51	1.34	0.07	11.21	2.03	0.27	10.25	2.34	0.31
Am	21.01	0.19	0.17	23.40	0.22	0.19	23.30	0.52	0.54	26.32	0.62	0.64
Cm	0.26	1.53	0.04	0.29	1.33	0.02	0.17	0.86	0.14	0.17	0.66	0.12

Curium decay to plutonium has a relatively small effect on final plutonium mass over the FEED scenario timescale. Curium is generated from capture in americium and decays to plutonium, most notably via [164],

$$^{243}\mathrm{Am} + \mathrm{n} \rightarrow ^{244}\mathrm{Am} \xrightarrow{\beta\text{-decay}} ^{244}\mathrm{Cm} \xrightarrow{\alpha\text{-decay}} ^{240}\mathrm{Pu}. \tag{8.1}$$

Removing curium at the reprocessing step of FEED-AM scenarios, leads to ^{244}Cm being sent to the waste stream where it decays to ^{240}Pu, therefore FEED-AM scenarios reduce the plutonium stockpile slightly less than the equivalent FEED-TRU scenario. Americium content is similar in equivalent FEED-AM and FEED-TRU scenarios, but americium increases significantly more in FEED-PU scenarios. FEED-PU scenarios remove americium at reprocessing which removes the potential for transmutation and leads to a greater americium inventory at the end of the fuel cycle.

Considering the FULL scenarios, there are two main differences compared with the FEED scenarios: (1) FULL-TRU scenarios reduce americium more than the equivalent FULL-AM scenarios.[1] (2) Plutonium reduction is larger in FULL-TRU scenarios than FULL-PU scenarios as curium is not removed from the fuel cycle, so there is no decay of curium to plutonium in the reprocessing waste stream. In addition, FULL-TRU scenarios make more fuel available than FULL-PU scenarios by reprocessing all TRUs, therefore more reactors can be operated and there is more fuel recycling. FULL-PU scenarios are better than FULL-AM scenarios at reducing plutonium in the stockpile because americium is removed. In FULL-AM scenarios curium is generated through capture in americium and removed at reprocessing to decay to plutonium. In FULL-PU scenarios americium is removed at reprocessing, therefore minimising curium build-up and negating the effect of curium decay. FULL-PU scenarios are not as effective as FULL-TRU scenarios at reducing plutonium as a result of less reactor operation and therefore less recycling of the stockpile through SFRs. Also, to a lesser extent, some curium is still produced in FULL-PU scenarios, which is not recycled in reactors, leading to some plutonium being generated from curium decay than in the equivalent FULL-TRU scenario. FULL-Zr scenarios reduce the stockpile more effectively than the equivalent FULL-MOX scenario. Lower CRs in FULL-Zr reactors lead to a more effective reduction of the stockpile, but more significantly, the lower TRU inventory of

[1] With the exception of the MOX-LO-TRU scenario as the total generating capacity is lower than for the MOX-LO-AM scenario

FULL-ZR reactors leads to more fuel being available to operate reactors, resulting in more recycling through SFRs and a better overall reduction in the stockpile.

Considering the FULL scenario WASTE streams only, results are in the same order as FULL scenarios but stockpiles are lower in the WASTE streams as the final fuel buffer has been excluded. However, the WASTE-HI-TRU scenarios generate more americium in the waste stream than WASTE-HI-AM scenarios. This is a result of more recycling, as a result of increased total fuel available, leading to a greater build-up of americium in the waste stream. Similarly, as a results of less recycling in WASTE-HI scenarios, the amount of plutonium in the waste stream for WASTE-HI scenarios is lower than for WASTE-LO scenarios.

8.3.2 Radiotoxicity

8.3.2.1 Total radiotoxicity

The total radiotoxicity for once-through SFR BURNERS in Chapter 7 was not discussed as the total radiotoxicities were similar for each of the BURNER scenarios. Although total radiotoxicity is not used as one of the assessment criteria it is important to understand how different closed fuel cycle scenarios will influence the total radiotoxicity, shown in Figure 8.2. To obtain a better indication of what contributes most significantly to radiotoxicity over different timescales, the elemental contributions to total radiotoxicity were plotted in Figure 8.3 for FULL scenarios with different element recycling strategies. FULL scenarios were selected as an example as they show large discrepancies between PU, AM and TRU scenarios, based on the large discrepancies in final elemental mass.

In Figure 8.2 it is worth noting that all HI and LO scenarios are separated due to differences in final TRU inventories, discussed in Section 8.3.1. WASTE stream results are included for completeness but are not discussed here as the analysis is the same as Section 8.3.2.2, with the separation of results based on what is reprocessed and the build up of losses from reprocessing.

Considering the FEED scenarios in Figure 8.2, radiotoxicity starts off higher than the plutonium stockpile and drops below the plutonium stockpile after a few hundred to a few thousand years. Equivalent FEED-AM and FEED-TRU scenarios are very close,

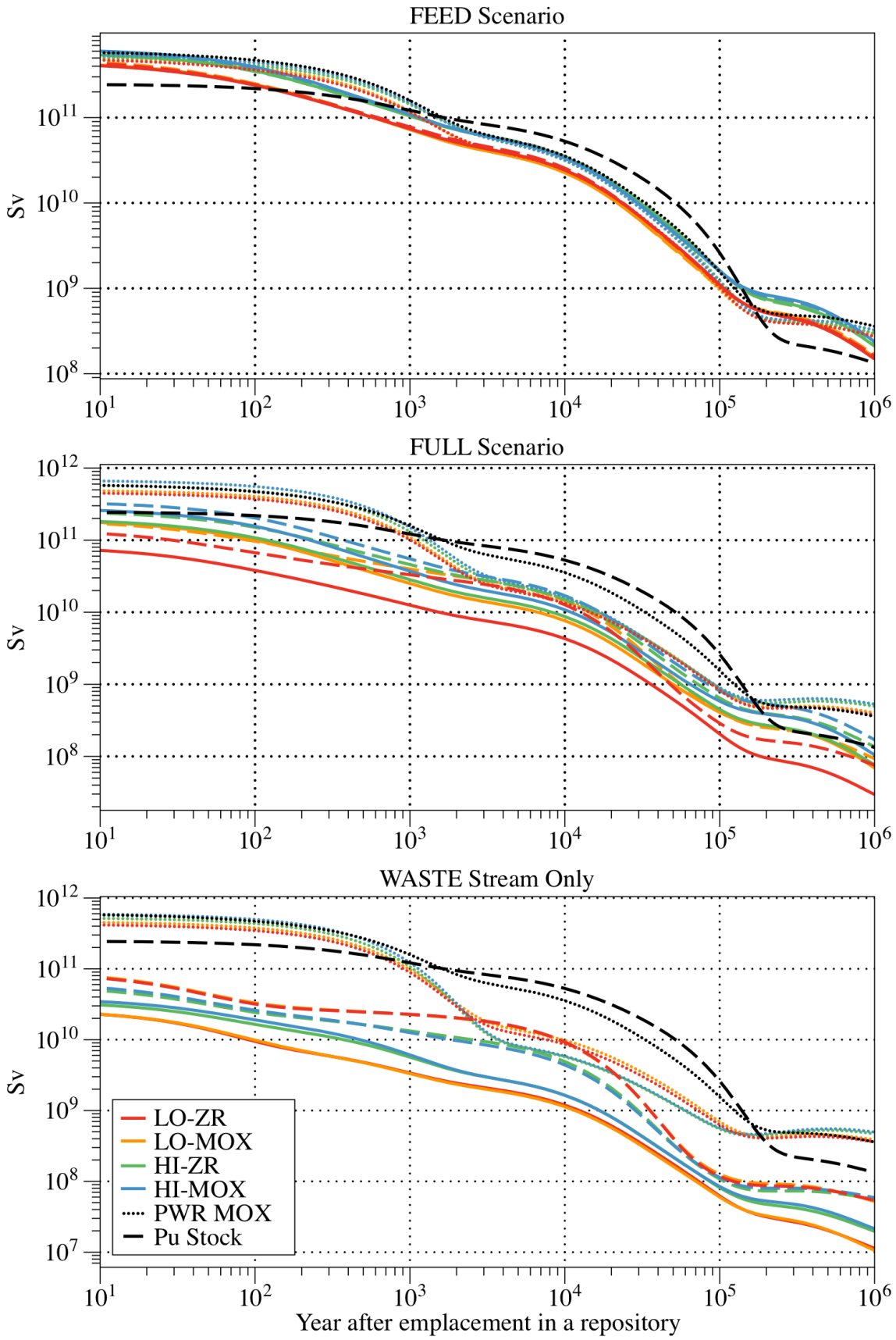


Figure 8.2: Total radiotoxicity in a repository for FEED, FULL and WASTE scenarios using the UK's plutonium stockpile as a fuel feed. The fine-dashed lines represent PU scenarios, long-dashed lines represents AM scenarios and solid line represents TRU scenarios.

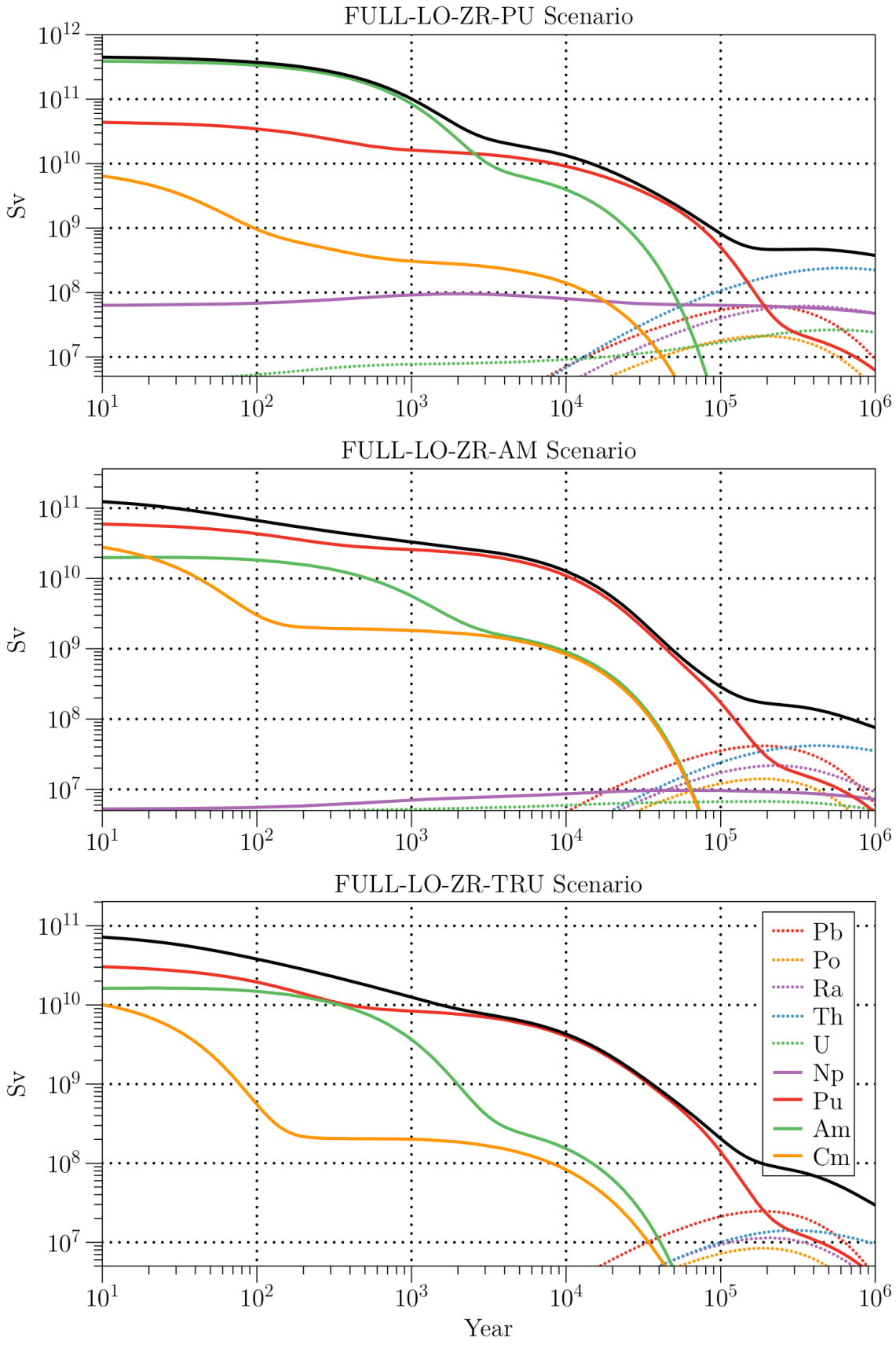


Figure 8.3: Elemental contributions to the total radiotoxicity of FULL scenarios.

as are equivalent FEED-ZR and FEED-MOX scenarios. FEED-PU scenarios have higher initial radiotoxicities than the equivalent FEED-AM or FEED-TRU scenarios but decay to similar levels after approximately 2000 years.

Considering the FULL scenarios in Figure 8.2, radiotoxicity tends to stay below that of the plutonium stockpile with the exception of FULL-PU scenarios which have a greater radiotoxicity than the plutonium stockpile for the first 1000 years. FULL-PU scenarios have a low plutonium inventory but very large americium inventory which dominates radiotoxicity in the first 1000 years, as shown in Figure 8.3. The distribution of FULL-AM and FULL-TRU scenarios in the first 1000 years in Figure 8.2 is a result of their different ^{241}Am and ^{240}Pu inventories. After 1000 years, all FULL-TRU scenarios have lower radiotoxicities than all FULL-AM scenarios as a result of lower plutonium inventories in FULL-TRU scenarios, which dominates radiotoxicity after americium has decayed away, as shown in Figure 8.3. The reason for the differences between FULL-AM and FULL-TRU is due to differences in plutonium mass, as discussed in Section 8.3.1.

8.3.2.2 Radiotoxicity lifetime

Figure 8.4 shows radiotoxicity per GWy(e) relative to natural uranium for each fuel cycle scenario. The time it takes for the radiotoxicity to drop below that of natural uranium is shown in Table 8.3, described as the lifetime of radiotoxicity. The radiotoxicity lifetime is a function of final transuranic inventory and the power generated by a scenario. The fine-dashed lines in Figure 8.4 represent PU scenarios, long-dashed lines represent AM scenarios and the solid lines represent TRU scenarios. In this thesis, a significant reduction in the lifetime of radiotoxicity was considered to be an order of magnitude less that UOX PWR, that is, less than 29,000 years.

Considering the FEED scenarios in Figure 8.4, results are very close and only the FEED-LO scenarios have radiotoxicities that drop below the level of natural uranium before UOX PWR. Plutonium content has the biggest impact on radiotoxicity over the 100,000 year timescale. All FEED-LO-ZR results for radiotoxicity lifetime are lower than FEED-LO-MOX results, for example FEED-LO-ZR-PU has a shorter radiotoxicity lifetime than FEED-LO-MOX-TRU. This is surprising as FEED-LO-MOX-TRU has a lower final plutonium inventory and higher generating capacity which should

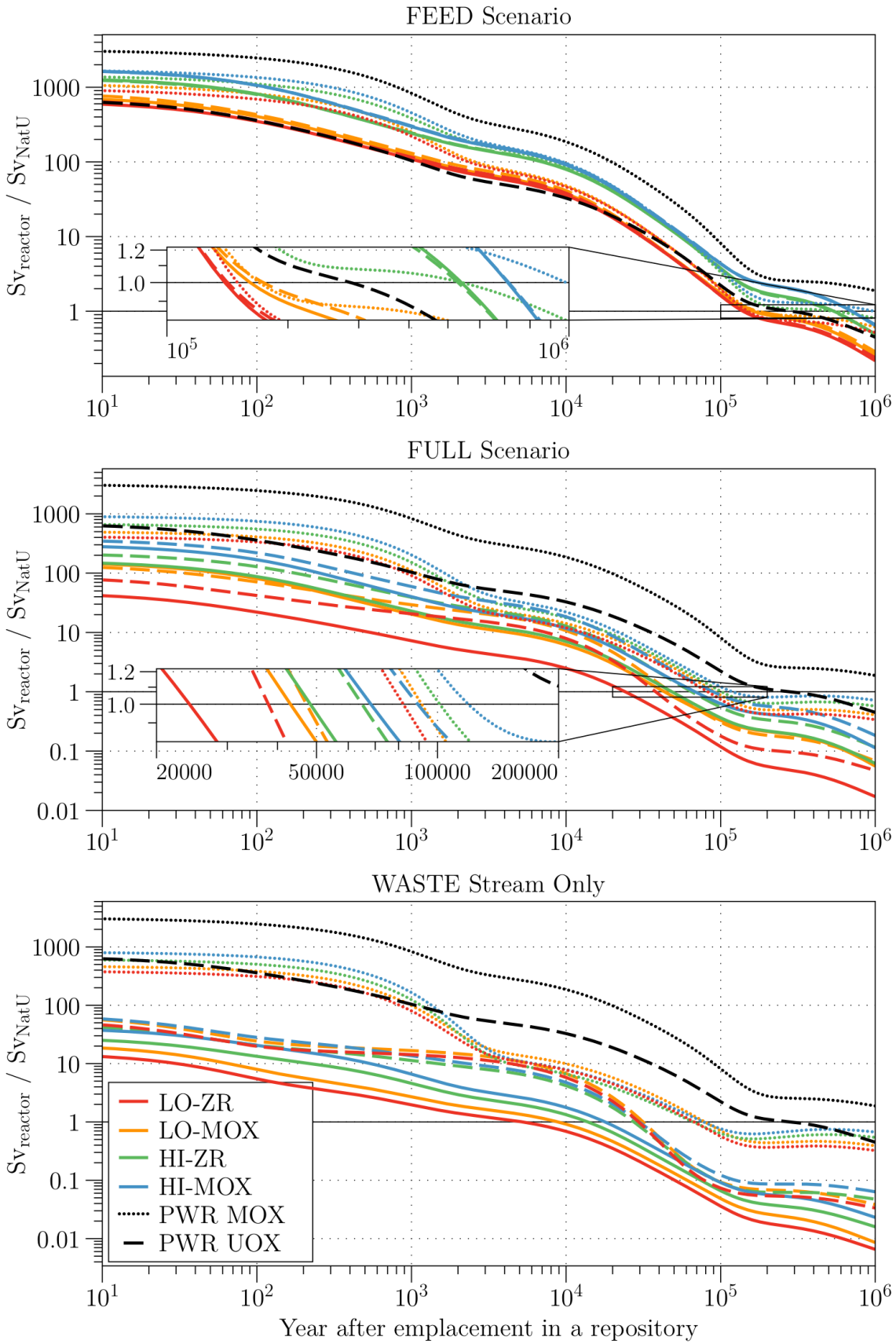


Figure 8.4: Radiotoxicity per GWy(e) for FEED, FULL and WASTE scenarios after disposal in a repository. Radiotoxicities are given relative to the radiotoxicity of natural uranium required to fuel a UOX PWR. Fine dashed lines represent PU scenarios, thick dashed lines represent AM scenarios, solid line represents TRU scenarios.

Table 8.3: Years taken for the radiotoxicity of final inventories to decay to the same radiotoxicity as natural uranium for FEED, FULL and WASTE scenarios using the UK's plutonium stockpile as a fuel feed. UOX PWR SNF takes 290,000 years.

	Zr PU (years)	**Zr AM (years)**	**Zr TRU (years)**	**MOX PU (years)**	**MOX AM (years)**	**MOX TRU (years)**
FEED						
LO	150,000	140,000	140,000	170,000	170,000	160,000
HI	540,000	530,000	540,000	980,000	710,000	700,000
FULL						
LO	82,000	38,000	24,000	91,000	47,000	43,000
HI	100,000	65,000	48,000	120,000	89,000	68,000
WASTE Stream Only (for FULL scenario)						
Low	67,000	30,000	5,200	79,000	32,000	8,900
High	66,000	27,000	14,000	77,000	30,000	18,000

correspond to a shorter lifetime. FEED-LO-Zr-PU has a shorter lifetime than FEED-LO-MOX-TRU because, over the 100,000 to 200,000 year timescale, plutonium decays to the same level as lead–thorium, as shown in Figure 8.3. Lead–thorium isotopes contribute more to radiotoxicity in FEED-LO-MOX-TRU scenarios than FEED-LO-ZR-PU scenarios, as shown in Figure 8.5, resulting in a lower FEED-LO-ZR-PU radiotoxicity at 200,000 years.

Considering the FULL scenarios in Figure 8.4, the radiotoxicity lifetime is lower than UOX LWR for FULL scenarios. FULL-PU scenarios have the longest radiotoxicity lifetime, despite lower plutonium inventory due to much lower generating capacities. Similarly, FULL-HI scenarios have longer radiotoxicity lifetimes than FULL-LO scenarios and FULL-MOX scenarios have longer lifetimes than FULL-ZR scenarios. Separation between FULL-HI and FULL-LO, as well as FULL-MOX and FULL-ZR is a result of differences in the final inventories and power generated. One example comparison of interest is the FULL-LO-ZR-AM scenario, which has a shorter radiotoxicity lifetime than the FULL-LO-MOX-TRU scenario, despite the FULL-MOX-LO-TRU scenario having approximately 22% less plutonium (which dominates radiotoxicity in the 30,000 to 50,000 year time-cale) as the FULL-LO-ZR-AM scenario generates approximately 23% more power, resulting in a shorter radiotoxicity lifetime.

Considering the WASTE stream only results from FULL scenarios in Figure 8.4,

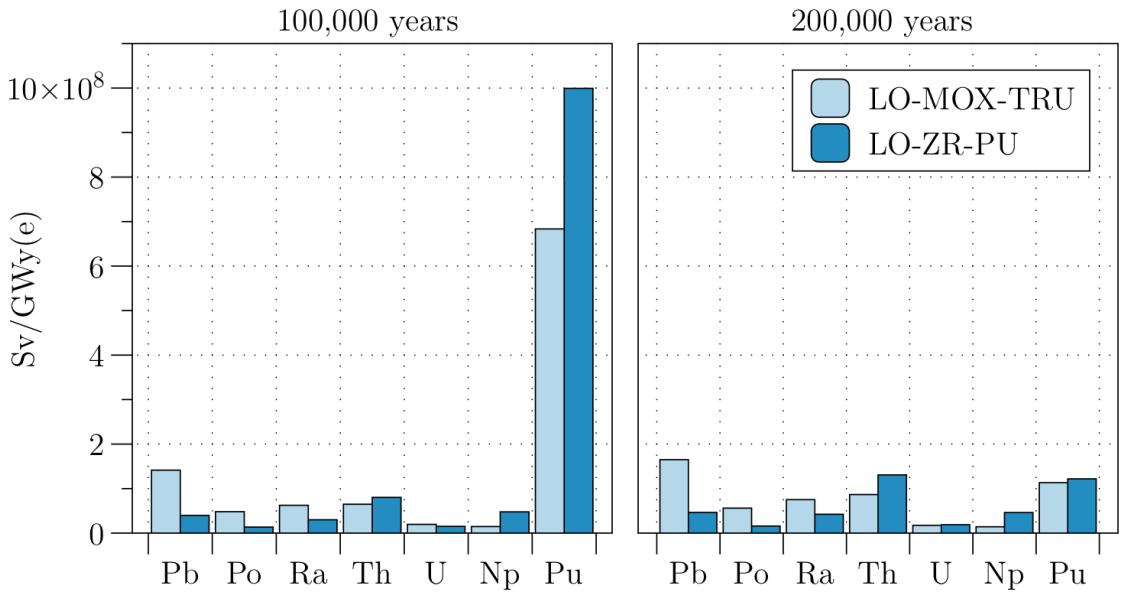


Figure 8.5: Comparison of the major elemental contributors to radiotoxicity at 100,000 and 200,000 years for FEED scenarios.

radiotoxicity lifetimes are shorter than the FULL scenario as a result of the final fuel buffer being excluded from analysis. All WASTE-TRU results have shorter radiotoxicity lifetimes than WASTE-AM results and all WASTE-AM results have shorter radiotoxicity lifetimes than WASTE-PU results. The order of WASTE lifetimes is a result of the total mass of transuranics sent to the waste stream at reprocessing. By reprocessing more transuranics in the AM and TRU scenarios, less transuranic material is sent to the waste stream. WASTE-LO radiotoxicity lifetimes were expected to be shorter than WASTE-HI lifetimes as a result of lower reprocessing losses. However, WASTE-LO-AM lifetimes are longer than the WASTE-HI-AM lifetimes, as a results of increased plutonium in the final WASTE-LO-AM inventory. The increased plutonium content in the WASTE-LO-AM inventory is due to less power generation in WASTE-HI-AM scenarios, resulting in less reprocessing and less curium production which is sent to the waste stream. Curium is removed in WASTE-AM scenarios at reprocessing and allowed to decay to plutonium in the reprocessing waste stream.

For most cases in Figure 8.4, few conclusions can be made as the lifetime of results are similar orders of magnitude and beyond the 29,000 years considered to be a significant reduction in radiotoxicity lifetime.

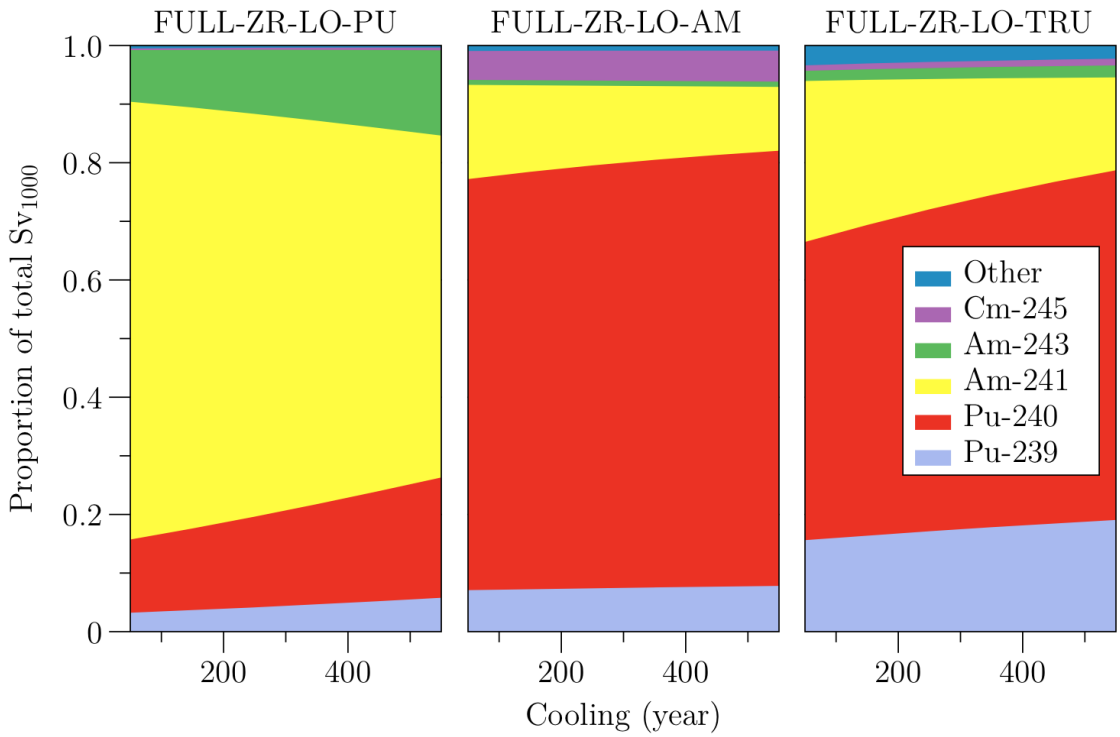


Figure 8.6: Main isotopic contributions to the 1000 year radiotoxicity of FULL scenarios.

8.3.2.3 1000 year radiotoxicity

Figure 8.7 shows the total radiotoxicity relative to direct disposal scenarios at 1000 years (Sv_{1000}), which is assumed to be the earliest timescale for a disturbed repository. Scenario results are given relative to the stockpile direct disposal scenario, with and without the offset of once-through LWR SNF. The change in results over a period of 50 to 500 years shows how the radiotoxicity of different scenarios changes over time. The total radiotoxicity at 1000 years is high, as shown in Figure 8.2, therefore anything less than a factor of two reduction in total radiotoxicity could be considered as negligible. As such, in this thesis, a factor of two reduction in 1000 year radiotoxicity is considered to be a significant improvement factor.

As seen in Figure 8.6, ^{240}Pu is the most significant contributor to 1000 year radiotoxicity, followed by ^{241}Am and ^{239}Pu, unless very high levels of americium are present in the final inventory, for example the FULL-PU scenario. ^{240}Pu is more dominant in AM scenarios than TRU scenarios due to the removal of curium at reprocessing in AM scenarios which leads to the decay of ^{244}Cm to ^{240}Pu in the reprocessing waste stream, as discussed in Section 8.3.1.

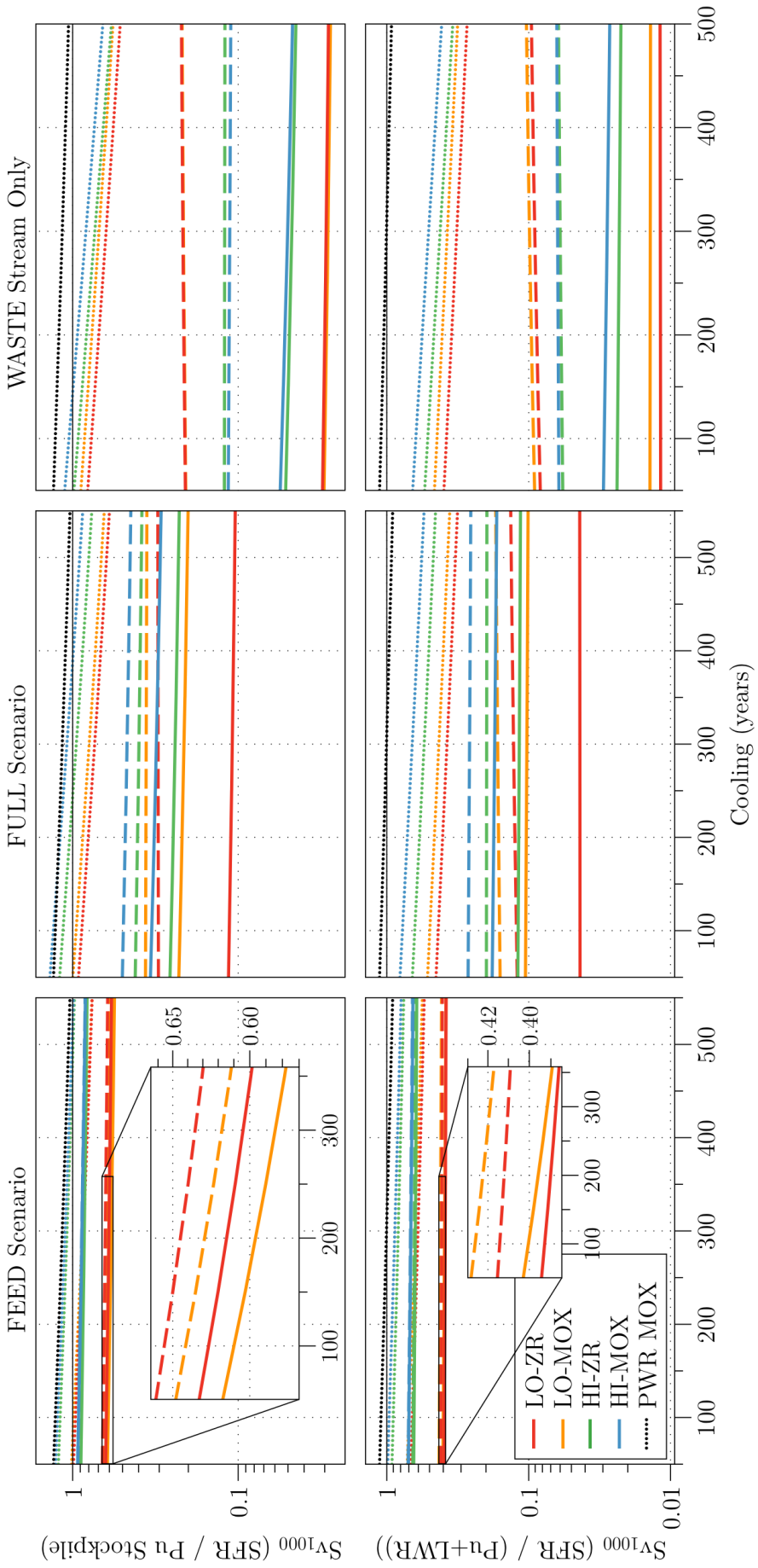


Figure 8.7: 1000 year radiotoxicity (Sv_{1000}) for FEED, FULL and WASTE scenarios relative to the direct disposal scenarios. Results in the upper figures are relative to direct disposal of the UK's plutonium stockpile. Results in the lower figures are relative to a once-through PWR scenario (the UK's plutonium stockpile plus the same GWy(e) of PWR SNF). Fine dashed lines represent PU scenarios, thick dashed lines represent AM scenarios, solid line represents TRU scenarios.

Considering the FEED scenarios in Figure 8.7, FEED-LO-AM and FEED-LO-TRU scenarios have the greatest reduction in 1000 year radiotoxicity as they have the lowest final inventory of plutonium and americium. FEED-MOX scenarios have a greater reduction in 1000 year radiotoxicity than FEED-ZR scenarios as a result of lower plutonium inventory, however, the overall difference between FEED-MOX and FEED-ZR 1000 year radiotoxicity is small. In addition, when once-through LWR offset is considered, FEED-ZR scenarios have a greater reduction in 1000 year radiotoxicity compared to FEED-MOX scenarios because FEED-ZR scenarios generate more electricity than the equivalent FEED-MOX scenario, offsetting more LWRs. FEED-TRU 1000 year radiotoxicities are lower than the equivalent FEED-AM scenarios as a result of smaller plutonium inventories at the end of the fuel cycle. FEED-HI scenarios have greater radiotoxicities than the equivalent FEED-LO scenarios due to a smaller reduction in inventory and longer cooling times, resulting in more americium in-growth.

Considering the FULL scenarios in Figure 8.7, the FULL-AM and FULL-TRU scenarios have much lower 1000 year radiotoxicities than all FEED scenarios. However, FULL-PU scenarios have similar 1000 year radiotoxicities to the FEED-PU scenarios (lower when once-through LWR offset is considered), as any reduction in plutonium radiotoxicity is offset by an increase in americium. As americium is the major contributor to radiotoxicity in FULL-PU scenarios, the relative radiotoxicities decay much faster over time than the FEED-PU scenarios as a result of the decay of americium, Figure 8.6. FULL-ZR scenario 1000 year radiotoxicities are lower than the equivalent FULL-MOX scenarios due to more effective stockpile reduction. FULL-ZR scenarios are more effective at reducing the 1000 year radiotoxicity, to the point that the FULL-HI-ZR-TRU results are close to FULL-LO-MOX-TRU results, despite a longer cooling period. It is also worth noting that the FULL-ZR-AM results are close to the FULL-MOX-TRU results, getting similar improvements in 1000 year radiotoxicity without the need for curium reprocessing.

Considering the WASTE stream only results from FULL scenarios in Figure 8.7, all 1000 year radiotoxicity results are improved over the FULL scenarios, as a result of excluding the final fuel inventories. WASTE-PU results are still similar to FULL-PU scenarios as the build-up of americium in the reprocessing waste stream contributes

most significantly to the 1000 year radiotoxicity of FULL-PU and WASTE-PU scenarios. WASTE-ZR and WASTE-MOX results for 1000 year radiotoxicity are close together as reprocessing losses are the same for WASTE-ZR and WASTE-MOX. The only difference between the WASTE-ZR and WASTE-MOX 1000 year radiotoxicity results is due to more reprocessing in the WASTE-ZR, leading to more material sent to the reprocessing waste stream and a greater build-up of higher actinides in MOX scenarios due to the softer neutron spectrum. As previously mentioned, the WASTE-HI-AM 1000 year radiotoxicity results are lower than the WASTE-LO-AM results which is unexpected. Greater reprocessing losses in WASTE-HI-AM is outweighed by more reprocessing and a more significant build-up of higher actinides in WASTE-LO-AM, as discussed in the previous section.

Overall, the 1000 year radiotoxicity is significantly reduced if americium is recycled (AM and TRU scenarios). Any differences in 1000 year radiotoxicity is a result of different final plutonium and americium inventories. Reprocessing of curium is advantageous in most cases as it prevents the decay of ^{244}Cm to ^{240}Pu in the reprocessing waste stream. Curium decay is a significant contributor to 1000 year radiotoxicity in FULL scenarios. However, curium decay is not a significant contributor to 1000 year radiotoxicity over the timescales of FEED scenarios, due to the minimal build-up of curium in the fuel cycle by 2150.

8.3.3 Repository size

Figure 8.9 shows the cumulative decay heat (CDH) of each scenario relative to the stockpile direct disposal scenario, with and without the offset of once-through LWRs. CDH is used to estimate relative repository size. The minimum cooling time was assumed to be 50 years and the longest 350 years before disposal. However, results were plotted up to 550 years to see how cooling time and the decay of short to intermediate lived components of SNF influence repository size. In this thesis a 20% reduction in repository size is considered significant, see Section 7.3.3.1.

For repository size, the relative performance of scenarios to one another is similar to 1000 year radiotoxicity as ^{241}Am and ^{240}Pu have the most significant contributions to radiotoxicity and decay heat over the 1000 year timescale. However, CDH is integrated over 1000 years so shorter lived isotopes, notably ^{244}Cm and ^{238}Pu also contribute

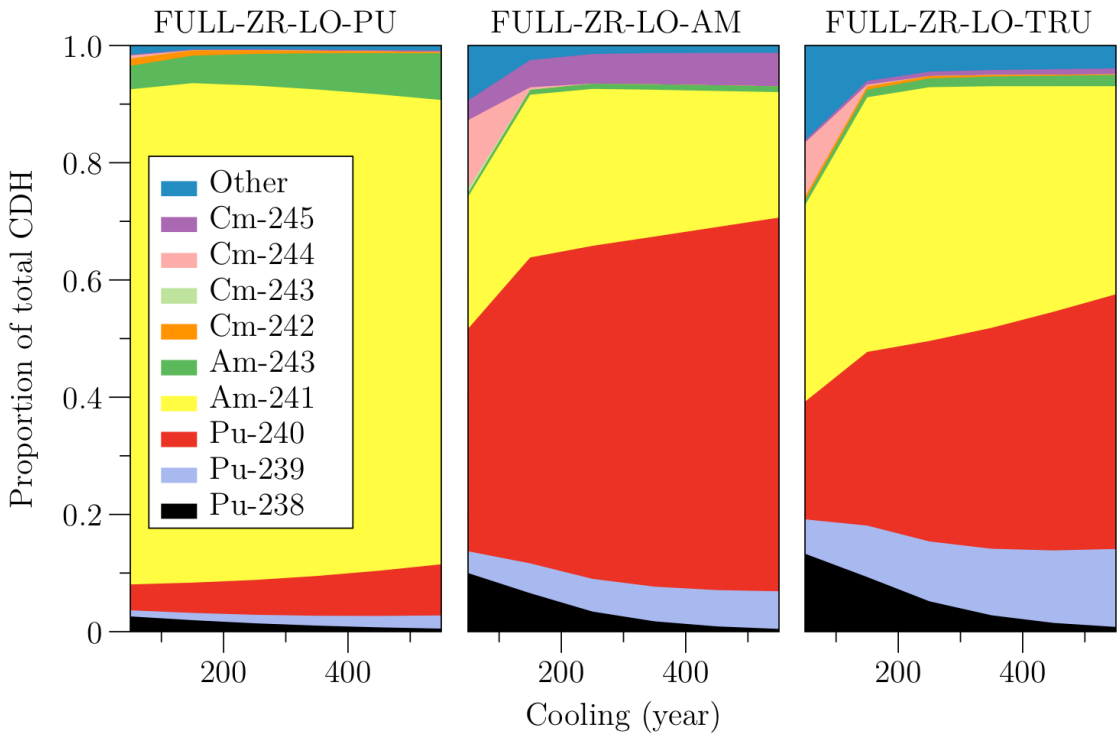


Figure 8.8: Main isotopic contributions to the CDH of FULL scenarios.

to repository size, and ^{241}Am contributes more significantly. Figure 8.8 shows the contribution of the most significant isotopes to repository size, with ^{244}Cm, ^{238}Pu and ^{241}Am having a more significant contribution than to radiotoxicity ins Figure 8.6. The contribution of ^{244}Cm, ^{238}Pu and other short lived isotopes to repository size is significantly reduced with 150 years of cooling, see Figure 8.8. It is worth noting that in FULL-AM scenarios ^{240}Pu is the major contributor to repository size, not ^{241}Am.

Considering the FEED scenarios in Figure 8.9, the FEED-LO scenario repository sizes are very close, with the exclusion of FEED-PU scenarios. When including the offset of once-through LWRs, FEED-LO scenarios are separated as a result of their different power generation. When considering the offset of LWRs, the FEED-LO-TRU and FEED-LO-AM scenarios have a repository size more than half that of the direct disposal scenario. Compared to plutonium stockpile disposal alone, FEED-LO-TRU and FEED-LO-AM scenarios, with 50 years of cooling, have repository sizes similar to direct disposal. However, with 150 years of cooling FEED-LO-TRU and FEED-LO-AM scenarios have significantly smaller repository sizes than the direct disposals scenario, that is more than 20% smaller.

Considering the FULL scenarios in Figure 8.9, the FULL-LO-ZR-TRU scenario has

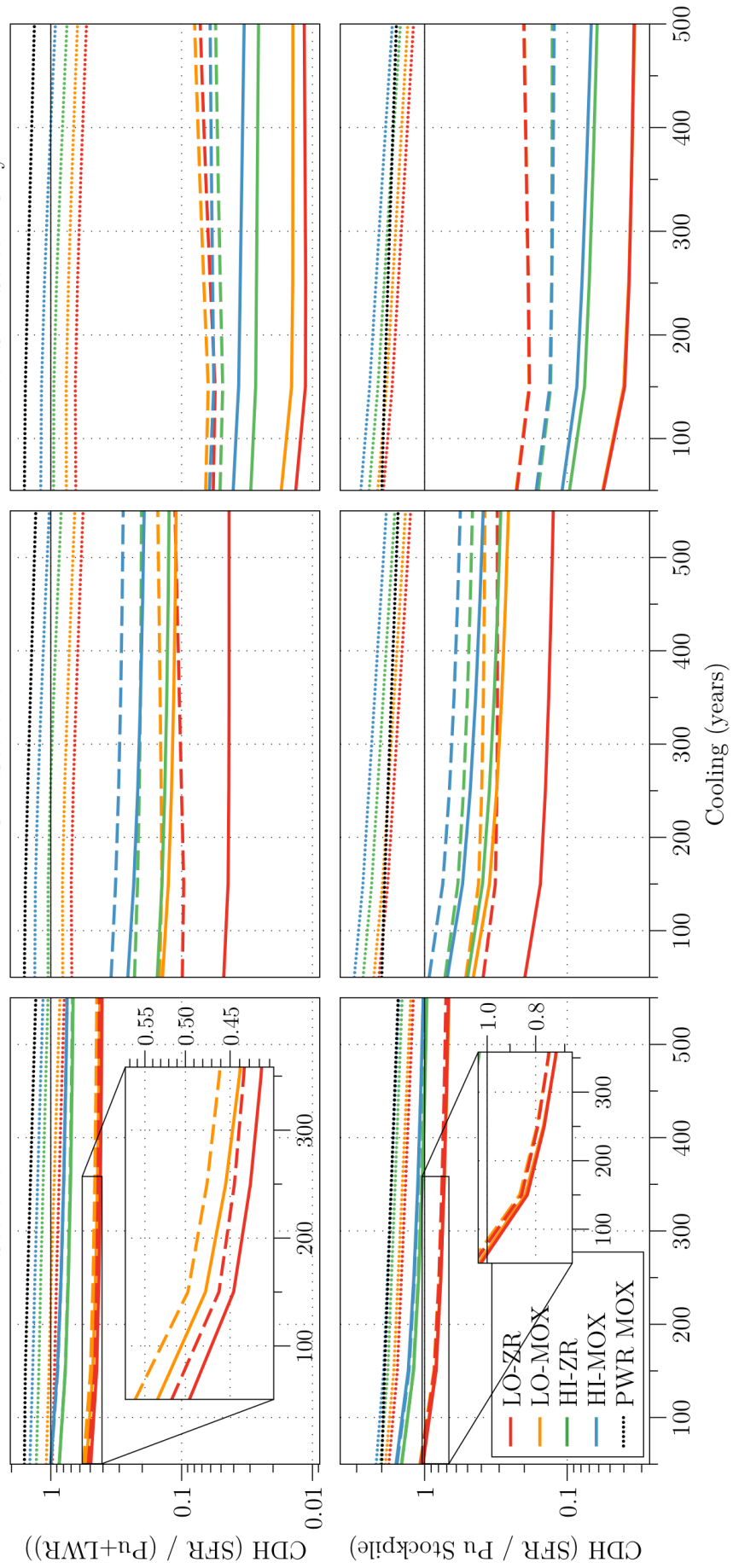


Figure 8.9: Relative repository size for FEED, FULL and WASTE scenarios estimated by the CDH method and given relative to direct disposal scenarios. Results in the lower figures are relative direct disposal of the UK's plutonium stockpile. Results in the upper figures are relative to a once-through PWR scenario (the UK's plutonium stockpile plus the same GWy(e) of PWR SNF). Fine dashed lines represent PU scenarios, thick dashed lines represent AM scenarios, solid line represents TRU scenarios.

Table 8.4: Bare sphere critical mass (kg) for the equilibrium reprocessing feeds of FEED and FULL scenarios using the UK's plutonium stockpile. Initial stockpile critical mass is 12.8 kg.

	Zr PU	**Zr AM**	**Zr TRU**	**MOX PU**	**MOX AM**	**MOX TRU**
FEED						
Low	16.3	16.4	16.2	17.0	16.9	16.6
High	16.2	15.8	15.8	16.8	16.3	16.4
FULL						
Low	17.9	17.1	16.4	17.4	17.6	17.6
High	18.6	17.7	17.6	19.3	18.0	17.6

the smallest repository size. FULL-ZR scenarios have smaller repository sizes than the equivalent FULL-MOX scenarios. A significant result is the FULL-LO-ZR-AM and FULL-HI-ZR-TRU scenarios having similar or smaller repository sizes than the FULL-LO-MOX-TRU scenario. The FULL-LO-MOX-TRU scenario performs poorly as it has a greater plutonium and americium inventory than the FULL-LO-ZR-AM and FULL-HI-ZR-TRU scenarios. With cooling, the FULL-LO-MOX-TRU repository size improves over the FULL-LO-ZR-AM scenario as a result of the decay of ^{241}Am and the lower overall heat contribution of ^{240}Pu in the FULL-LO-MOX-TRU scenario, Figure 8.8. When considering FULL-PU scenarios the repository size is greater than the direct disposal scenarios as a result of the large quantity of americium in the waste stream.

Considering the WASTE stream only results from FULL scenarios in Figure 8.9, the trend is the same as described for 1000 year radiotoxicity in Section 8.3.2.3. WASTE-TRU results have the lowest repository size, followed by WASTE-AM and WASTE-PU results.

8.3.4 Bare sphere critical mass

The bare sphere critical mass for reprocessed fuel streams at the end of each fuel cycle scenario is presented in Table 8.4. It is worth noting that this indicates the peak degradation of the fuel vector at the end of the scenario. The degradation after a few recycles will be less.

There is very little to differentiate between the bare sphere critical masses of different scenarios. Although there is some variation in critical mass, this variation is small. For FEED scenarios, the FEED-LO scenarios degrade the final vector more than the FEED-HI scenarios as a result of more recycling through a reactor. For FULL scenarios, the FULL-HI scenarios degrade the final vector more than the FULL-LO scenarios because the longer cooling times of the FULL-HI scenarios have more of an impact on the final fuel vector than the amount of recycling. Longer cooling times in FULL-HI scenarios, compared to FULL-LO scenarios, leads to the loss of fissile ^{241}Pu due to decay.

FULL-PU scenarios have a more significant degradation of the fuel vector than the FULL-AM or FULL-TRU scenarios. This was unexpected as FULL-AM and FULL-TRU scenarios have MAs in the fuel and less plutonium. The fuel feeds for FULL-TRU and FULL-AM scenarios have a higher proportions of ^{239}Pu than FULL-PU scenarios, of the order of 1%. The presence of americium and curium in the FULL-AM and FULL-TRU scenarios has a negative impact equivalent to less than 0.5% of ^{239}Pu. The ^{239}Pu equivalence was estimated using ERANOS equivalence coefficients from a metallic fuelled reactor, for example, the ^{239}Pu equivalence of americium and curium in the FULL-ZR-TRU reprocessing stream was equal to -0.33% of ^{239}Pu.

8.4 Discussion

A summary of results for the key assessment criteria can be found in Table 8.5 for FEED and FULL scenarios. Improvement factors over reference scenarios were tabulated for ease of comparison. This chapter sought to answer several questions which were outlined in the introduction and discussed below.

Final TRU inventory mass results are not discussed in depth as repository size, radiotoxicity lifetime and 1000 year radiotoxicity are considered more important effects resulting from the final mass of transuranics. Bare sphere critical mass is also not discussed in depth as the scenarios have a relatively small influence on this factor when compared to PWR MOX. Results for 1000 year radiotoxicity and repository size are only improved if americium is recycled, in AM and TRU scenarios. As a result of the poor performance of PU scenarios, they are excluded from most analysis in

Table 8.5: Using the UK's plutonium stockpile as a fuel feed, the FEED, FULL and WASTE fuel cycle scenario improvement factors are presented relative to a [reference case] for each assessment criteria. Improvement factors greater than one show a positive improvement. Improvement factors less than one show a dis-improvement.

		Zr Lo-PU	Zr Lo-AM	Zr Lo-TRU	MOX Lo-PU	MOX Lo-AM	MOX Lo-TRU	Zr Hi-PU	Zr Hi-AM	Zr Hi-TRU	MOX Hi-PU	MOX Hi-AM	MOX Hi-TRU
TRL		6	4	3	7	5	3	6	4	3	8	5	3
FEED Scenario													
GWy(e) [PWR MOX]		2.94	3.77	3.84	2.65	3.26	3.39	2.09	2.44	2.43	1.83	2.02	2.07
Mass [Stock]	TRU	1.92	2.16	2.24	2.00	2.30	2.38	1.48	1.54	1.58	1.50	1.58	1.60
CDH [Stock]	50y	0.56	0.94	0.97	0.53	0.93	0.96	0.49	0.69	0.69	0.46	0.64	0.63
CDH [Stock+LWR]	50y	1.02	1.94	2.02	0.93	1.78	1.87	0.78	1.16	1.16	0.69	1.00	0.99
Sv_{1000} [Stock]	50y	1.01	1.51	1.58	0.98	1.54	1.62	0.82	1.10	1.12	0.79	1.07	1.07
Sv_{1000} [Stock+LWR]	50y	1.48	2.41	2.54	1.39	2.33	2.48	1.09	1.52	1.55	1.01	1.41	1.42
Sv Lifetime [PWR UOX]		1.93	2.07	2.07	1.71	1.71	1.81	0.54	0.55	0.54	0.30	0.41	0.41
Critical Mass [Stock]		1.27	1.28	1.27	1.33	1.32	1.30	1.27	1.23	1.23	1.31	1.27	1.28
FULL Scenario													
GWy(e) [PWR MOX]		6.24	9.01	9.71	5.55	7.63	6.93	4.85	6.59	6.93	4.16	5.20	5.20
Mass [Stock]	TRU	2.60	5.24	12.84	2.47	4.33	6.98	1.99	3.63	6.08	1.87	3.09	4.83
CDH [Stock]	50y	0.48	2.57	5.05	0.44	1.95	2.20	0.37	1.39	2.00	0.32	1.07	1.44
CDH [Stock+LWR]	50y	1.45	10.12	20.98	1.23	6.77	7.14	0.95	4.36	6.51	0.75	2.89	3.88
Sv_{1000} [Stock]	50y	1.08	3.30	8.74	1.00	2.75	4.38	0.83	2.39	3.87	0.73	2.00	2.95
Sv_{1000} [Stock+LWR]	50y	2.22	8.30	23.02	1.94	6.29	9.48	1.51	5.03	8.39	1.24	3.74	5.53
Sv Lifetime [PWR UOX]		3.54	7.63	12.08	3.19	6.17	6.74	2.90	4.46	6.04	2.42	3.26	4.26
Critical Mass [Stock]		1.40	1.34	1.28	1.36	1.38	1.38	1.45	1.38	1.38	1.51	1.41	1.38
WASTE Stream Only													
GWy(e) [PWR MOX]		3.34	7.97	49.04	3.05	7.96	50.36	2.97	11.96	32.50	2.81	12.17	31.40
Mass [Stock]	TRU	0.51	4.44	17.94	0.47	4.39	17.76	0.41	6.27	10.39	0.36	6.06	9.21
CDH [Stock]	50y	1.55	17.43	74.54	1.31	15.26	57.75	1.05	19.67	33.78	0.84	16.30	24.77
CDH [Stock+LWR]	50y	1.23	4.80	32.32	1.12	4.82	33.01	1.02	8.25	19.35	0.89	8.71	17.95
Sv_{1000} [Stock]	50y	2.53	12.07	85.14	2.17	11.01	71.51	1.85	17.39	41.92	1.52	16.33	33.64
Sv_{1000} [Stock+LWR]	50y	4.33	9.67	55.77	3.67	9.06	32.58	4.39	10.74	20.71	3.77	9.67	16.11

this section, except when specifically referred to. In this section, waste performance is measured by the assessment criteria for repository size, radiotoxicity lifetime, 1000 year radiotoxicity and final transuranic inventory. Fuel cycles with long reprocessing timescales and no curium reprocessing are described as high TRL fuel cycles. Fuel cycles with short cooling times or, curium reprocessing, are described as low TRL fuel cycles.

FEED Vs BURNER scenarios Once-through BURNER scenario results taken from Chapter 7 are compared to FEED closed fuel cycle scenario results in this section. FEED scenarios with americium inclusion (FEED-AM and FEED-TRU) generally have improved waste performance over BURNER scenarios. FEED-PU scenarios have larger repositories and greater 1000 year radiotoxicities than BURNER scenarios. Americium build-up is greater in FEED-PU scenarios than BURNER scenarios because americium is removed at reprocessing and builds up in the reprocessing waste stream. The greater americium content of FEED-PU inventories leads to much greater radiotoxicities and decay heat over a 1000 year time period.

FEED scenario final inventory improvement factors are 1.30 to 2.38, which is better than all BURNER scenarios and PWR MOX, 1.30. All FEED scenarios have shorter radiotoxicity lifetimes than BURNER scenarios, because the final plutonium inventory of FEED scenarios is much smaller. FEED-LO scenarios have shorter radiotoxicity lifetimes than the UOX PWR reference case. With 50 years of cooling the 1000 year radiotoxicity improvement factor for the best case BURNER scenario (BURNER-AM) is 0.99 (1.09 with LWR offset), compared to FEED scenario improvements of 1.07 to 1.58 (1.42 to 2.54 with LWR offset). With 50 years of cooling, the BURNER-AM scenarios increased repository size, with an improvement factor of 0.93 (0.96 with LWR offset), FEED scenarios reduced repository size with an improvement factor of 0.63 to 1.03 (0.99 to 2.02 with LWR offset). As such, FEED-AM and FEED-TRU scenarios can have a larger or smaller repository size than the best case BURNER scenarios, depending on fuel cycle parameters of the FEED scenario.

FEED-HI scenarios lead to larger repositories than FEED-LO scenarios. FEED-HI scenarios have longer cooling times which result in fewer reactors being operated over the fuel cycle and more decay of ^{241}Pu to ^{241}Am prior to reprocessing. As a

result of long cooling times in FEED-HI scenarios, final americium inventories are greater than BURNER-AM scenarios, resulting in larger repositories. However, with 150 years of cooling, americium in FEED-HI scenarios decays substantially and repositories are smaller than BURNER-AM scenarios as a result of americium decay and lower plutonium inventories in the FEED-HI scenarios. As a result of greater power generation in FEED scenarios compared to BURNER scenarios, when the offset of once-through LWRs is considered, repositories are always smaller for FEED scenarios than BURNER-AM scenarios.

For FEED scenarios to have an improvement in waste performance over BURNER scenarios there are five requirements: the operation of reprocessing facilities, short cooling times, the recycling of americium in the fuel, operating reactors until 2150 rather than 2100, and operating 2 to 4 times as many SFRs.

It is worth noting that FEED-TRU scenarios have small improvement in waste performance compared to FEED-AM scenarios. Curium reprocessing in FEED-TRU scenarios reduces the TRL of the fuel cycle without providing any substantial benefits over FEED-AM scenarios in terms of waste performance. Waste performance improvements over the timescale of FEED scenarios, to 2150, are dependent on reprocessing cooling times but not the recycling of all TRUs. Similarly the choice of metallic or MOX fuel in FEED scenarios has a limited impact on waste performance. As such, MOX fuel, which has been more widely used, would be preferable.

FULL scenarios FULL scenarios can have significant improvements over BURNER and FEED scenarios. However, FULL scenarios need operating until 2350 – 2460, operating 5 to 10 times more SFRs than the BURNER scenarios. This long timescale may not be realistic, as current UK nuclear policy does not extend past 2050, but warrants studying to determine the maximum stockpile reduction and the impact it has on waste performance.

There are a range of results between the best performing FULL scenario, FULL-ZR-LO-TRU, and worst performing FULL scenario, FULL-MOX-HI-AM. FULL scenarios have an improvement on final transuranic inventory of 3.09 to 12.84, waste lifetime improvement factors of 3.26 to 12.08, 1000 year radiotoxicity improvement factors of 2.00 to 8.74 (3.74 to 23.02 with LWR offset), repository size improvement factors of 1.07

to 5.05 (2.89 to 20.98 with LWR offset), and bare sphere critical mass improvements of 1.24 to 1.36. It is also worth noting that the FULL-ZR-AM scenarios have improvement factors similar or greater than FULL-MOX-TRU scenarios for 1000 year radiotoxicity and repository size, without the need for curium reprocessing.

For FULL scenarios, FULL-ZR reactors have better improvement factors compared to FULL-MOX reactors which is primarily caused by the lower TRU inventory of FULL-ZR reactors. The greater TRU inventory of FULL-MOX reactors, resulting in less fuel availability to operate more reactors. Therefore FULL-MOX scenarios have less reactors operating over the fuel cycle scenario, and shorter fuel cycles timescales, compared to the equivalent FULL-Zr scenario, leading to less recycling and less stockpile reduction. The amount of fuel available to operate reactors has more of an impact on FULL scenario results than the CR of the reactor. This is a significant limitation of this chapter and means that FULL-ZR and FULL-MOX scenario results differ due to the TRU inventory of reactors and not the neutronics performance of the different fuels. The total stockpile reduction of FULL scenarios is dependent on the TRU inventory of the last reactor operated, successively smaller reactors could be operated to achieve better stockpile reduction. However, there will be a balancing act between the fuel cycle timescale and the total stockpile reduction required when considering the operation of smaller SFRs.

Fuel cycle timescale and acceptable stockpile reduction levels are goals that should be outlined before choosing a fuel cycle scenario. A suggestion for further work is to develop MOX and Zr reactor models with similar inventories to see how the neutornics behaviour and design limitations, such as peak enrichment, will impact on FULL scenarios.

WASTE Vs FULL scenarios WASTE-TRU improvement factors were approximately 3.5 to 8 times the equivalent FULL-TRU scenarios, and WASTE-AM improvements are approximately 1.5 to 5.5 times the equivalent FULL-AM scenarios. WASTE results are quoted in many studies [113, 114, 115, 121], giving very large improvement factors based on only the waste stream from reprocessing being sent to a repository. Results in this chapter show that WASTE results have significantly greater improvement factors than FULL scenarios, even though FULL scenarios require 300 to 400

years of reactor operation. As such, WASTE results give an unrealistic impression of the potential performance of transmutation scenarios.

PU scenarios FEED-PU and FULL-PU scenarios, where only plutonium is recycled, reduce final inventories and have a small reduction on radiotoxicity lifetime but have a negative impact on repository size and 1000 year radiotoxicity. The increased recycling in the longer running FULL-PU scenarios leads to greater 1000 year radiotoxicity and larger repositories than FEED-PU scenarios as a result of more americium build-up. FULL-PU scenarios have improvement factors for 1000 year radiotoxicity of 0.73 to 1.08 (1.24 to 2.22 with LWR offset) and improvement factors for repository sizes of 0.32 to 0.48 (0.75 to 1.45 with LWR offset). The only advantages of FULL-PU scenarios in terms of improvement factors are the slightly shortened radiotoxicity lifetime, 2.42 to 3.54, and total inventory reduction, 1.87 to 2.60. Compared to the plutonium stockpile direct disposal scenario, 1000 year radiotoxicity and repository sizes are similar for FULL-PU and PWR MOX scenarios. The only significant improvements of FULL-PU scenarios over PWR MOX is the waste lifetime, which takes more than 200 years of extra reactor operation. However, the electricity generated by FULL-PU scenarios is 4.2 to 6.2 times more than PWR MOX which results in better improvement factors when the offset of LWRs is considered. However, if electricity generation and LWR offset is an important factor, high-CR plutonium recycling fuel cycles would be considered to maximise LWR offset, rather than low-CR fuel cycles discussed in this chapter. As such, there are no real benefits to low-CR fuel cycles over PWR MOX. High-CR SFR fuel cycles were considered in the UK's R&D pathways [21].

There are minimal benefits of long-term operation of low-CR, plutonium recycling fuel cycles when compared to PWR MOX. PWR MOX has a much higher TRL and effectively reduces the stockpile inventory in a short timescale without vastly increasing repository size.

Maximising improvement factors Shorter cooling times of LOW scenarios improve waste performance. Shorter cooling times have two advantages: Reduced americium in-growth and less material stored in cooling buffers. Reduced americium in-growth is advantageous as americium is a major contributor to decay heat and radiotoxicity over the first 1000 years of a repository. Less material in cooling buffers is

advantageous as additional fuel is available to operate more reactors. More reactors operated results in more recycling of the stockpile through SFRs and a more substantial reduction in the stockpile. The best method for reducing cooling time is to develop pyro-reprocessing techniques which can reprocess high activity fuel which has been cooled for a short period.

With a lot of recycling in the FULL scenarios, the inclusion of all MAs in the FULL-TRU scenarios over just americium in the FULL-AM scenarios improves waste performance as a result of two main factors: the transmutation of neptunium and curium; and the amount of material available for fuel is increased, as neptunium and curium are not removed at reprocessing. ^{244}Cm has a large contribution to decay heat and radiotoxicity. The build-up of ^{240}Pu in the reprocessing waste stream of FULL-AM scenarios is large due to the removal of ^{244}Cm at reprocessing which decays to ^{240}Pu. In FULL-AM scenarios ^{240}Pu is the major contributor to repository size. Reprocessing all transuranics in FULL-TRU scenarios ensures that ^{240}Pu does not build-up in the reprocessing waste stream, and improves waste performance as a result of transmutation and increasing the amount of fuel available for reactor operation.

8.4.1 Comparison to literature

Direct comparison between this work and the literature is not straight forward as all fuel cycles are unique. However, for the FULL-LO-TRU scenarios, there are two similar studies which investigate stockpile reduction, comparing results to a stockpile direct disposal scenario: A Japanese study investigated reducing 430 tHM of LWR SNF TRUs with SFRs and similar fuel cycle parameters to the FULL-LO-TRU scenario in this work, operating SFRs until 2300. The Japanese scenario improved repository loading by a factor of 4.7 and a waste lifetime by a factor of 10 [122]. A German scenario similarly considered the reduction of approximately 150 tHM of LWR SNF TRUs,[2] but used ADS fast reactors to reduce timescales, reducing the repository size by a factor of 4 by 2100 [124]. In comparison FULL-LO-MOX-TRU and FULL-LO-Zr-TRU scenarios in this chapter improved repository size by a factor of 2.20 and 5.05, and waste lifetime by a factor of 6.74 and 12.08. FULL-LO-TRU results have similar improvement factors compared to the German and Japanese scenario, and the

[2]Read from figure

timescales are similar to that of the Japanese scenario.

The German and Japanese studies considered low TRL fuel cycles, using very low-CR reactors (ADS or SFRs with high MA loading) and fuel cycles which reprocess all TRUs with short cooling times and low reprocessing losses, similar to the FULL-LO-TRU scenarios. It is of interest to see how these results change given higher TRL fuel cycle scenarios with long cooling times, high reprocessing losses, and recycling of plutonium and americium only, FULL-HI-AM scenarios. FULL-LO-TRU to FULL-HI-AM scenarios have already been discussed in this section, but in summary, the improvement factors for radiotoxicity lifetime, 1000 year radiotoxicity and repository size were approximately 4 to 6 times smaller for FULL-HI-AM scenarios and scenarios take up to 110 years longer. Whilst most waste performance factors are still improved for FULL-HI-AM scenarios, the repository size has an insignificant improvement over direct disposal. Final repository size for the FULL-MOX-HI-AM scenario is very similar to the direct disposal of the stockpile. To commit to fuel cycles which takes more than 300 years of reactor operation, a significant improvement in repository size would be preferable. Therefore, to maximise stockpile reduction and get significant improvements in waste performance, FULL-LO-AM or FULL-LO-TRU scenarios are the only options.

Other studies of relevance, discussed below, consider the operation of LWRs and SFRs, with SFRs using LWR SNF as a fuel feed. These studies compare the offset of LWRs and the ability of SFRs to reduce waste by 2100–2150. Although SFRs utilising LWR SNF is not modelled in this chapter, the plutonium stockpile could be considered as a pre-made thermal reactor waste feed, with SFRs being used to offset additional LWRs and reduce the plutonium stockpile by 2150. That is, essentially FEED scenarios with the offset of once-though LWRs included. Previous studies of relevance are a USA scenario, comparing 1000 year radiotoxicity and repository size to a once-through LWR fuel cycle (same as LWR offset in this chapter). In the USA scenario, all TRUs are reprocessed and reactors are operated to 2100, leading to a improvement of 1.6 in repository size and improvement 1.6 in 1000 year radiotoxicity [113,114,115,121]. The equivalent scenarios in this chapter were operated until 2150, FEED-LO-TRU, with slightly larger reductions in repository size of 1.87 to 2.02 and 1000 year radiotoxicity,

2.48 to 2.54. There are many differences between the USA scenario and FEED-LO-TRU scenario that will result in greater improvements in the FEED-LO-TRU scenario. The most significant differences between FEED-LO-TRU and the USA scenarios are the timescale of the scenarios and the exclusion of thermal reactor MAs and FPs in FEED-LO-TRU. Another similar study to FEED scenarios with LWR offset, is a French scenario considering the improvement of operating SFRs with LWR up to 2150, relative to a once-through LWR scenario [129]. With plutonium and americium reprocessed (similar to FEED-LO-AM scenarios), the French scenario reduced the repository size by a factor of 2, with the inclusion of all MAs this increases to 2.5 [129]. The equivalent scenarios in this chapter, FEED-LO-AM and FEED-LO-TRU, were operated over the same timescale and reduced the repository size by a factor of 1.78 to 1.94 for the FEED-LO-AM scenario and a factor of 1.87 to 2.02 for FEED-LO-TRU scenario. Results in this chapter show a smaller improvements in repository size than the French scenarios. The most significant differences between FEED-LO scenarios and the French scenarios will be a result of the difference in fuel feed.

Many studies have looked at the impact of WASTE streams only on repository size. WASTE stream results are most significantly influenced by reprocessing losses. The WASTE stream results looks at reprocessing waste from the FULL scenario in this chapter. As a result of the long timescales in FULL scenarios Cs and Sr have decayed significantly by the fuel cycle assessment date. Results from a USA scenario, with 99.9% of Cs and Sr removed, have repository size improvements of 10 to 43 for AM scenarios and 10.5 to 225 for TRU scenarios, depending on losses [113, 114, 115, 121]. In this chapter, WASTE-AM scenarios have a repository size improvement of 15.26 to 17.43 and WASTE-TRU scenarios have a repository size improvement of 24.8 to 74.5.

Overall, results in this chapter follow the same trends as those in the literature, but represent the unique UK situation. Several gaps in the literature, such as high-TRL SFR fuel cycles for stockpile reduction have been filled to illustrate the dependence of stockpile reduction on short cooling times and advanced fuel cycle technology.

8.4.2 Recommendations and further work

Based on Eurobarometer polls, the three main factors that influence public perception of nuclear power are: terrorism concerns, waste disposal and proliferation, in that

order [4]. The most important factor, terrorism, can not easily be prevented with fuel cycle scenario design, however, waste disposal and proliferation can.

Separated plutonium, such as the UK's plutonium stockpile, is a large proliferation concern. To tackle proliferation concerns, the re-use of the UK's plutonium stockpile, putting plutonium out of reach in a highly active matrix, would be preferable without any further reprocessing. The best option for reducing proliferation concerns is PWR MOX as discussed in Chapter 7, all scenarios in this chapter consider reprocessing which increases the proliferation risk. If reprocessing is a priority for a UK fuel cycle, then the most proliferation resistant recycling scheme is the inclusion of all TRUs. Inclusion of all TRUs at reprocessing increases the fuels radioactive dose requiring significant shielding to handle the fuel as curium is a spontaneous neutron emitter. Difficulties associated with fuel handling increases the intrinsic proliferation resistance of the fuel cycle, as discussed in Appendix D.

Considering the public perception of waste, re-use options outlined by the NDA [1] for UK plutonium disposition, discussed in Chapter 7, do not have a significant impact on waste performance. In the UK R&D pathways, closed SFR fuel cycles were considered, prioritising electricity generation with SFR fuel cycles reducing the size of a repository by 30% over once-through LWR fuel cycles [21]. Options currently outlined by the UK for plutonium stockpile disposition, and R&D pathways for large nuclear power generation, do not reduce the burden of nuclear waste significantly and do not aim to improve public perception of nuclear waste. Fuel cycles scenarios studies in this chapter could improve public perception of waste. The most effective way to improve waste performance is with FULL scenarios where radiotoxicity lifetime can be reduced by a factor of 12.1, repository size by a factor of 5.1 and 1000 year radiotoxicity by a factor of 8.7. These improvements are significant and may improve public perception of nuclear waste, however they require a closed fuel cycle until 2350–2460. In a reasonable timescale, such as 2150 in the FEED scenarios, the radiotoxicity lifetime can be reduced by a factor of 2.1, repository size by a factor of 1.0 and 1000 year radiotoxicity by 1.62. The waste performance factors for FEED scenarios are smaller than for FULL scenarios and may not be large enough to affect public perception. In the present author's opinion, only FULL scenarios can improve waste performance enough to have an impact on public perception. However, FULL scenarios

are dependent on cooling times. Moving from a FULL-LO-TRU scenario to a FULL-HI-TRU scenario results in a reduction in waste performance factors by 2 to 4 times, making the fuel cycle less favourable. As such, it is likely that only low TRL fuel cycles with short cooling times and curium reprocessing, FULL-LO-TRU, would have an impact on the public perception of waste and proliferation resistance. The cost of developing technology for FULL-LO-TRU scenarios to improve public perception on waste and proliferation, may in turn have a negative impact on public perception. Similarly the need to operate a closed fuel cycle to 2350–2460 may also have a negative impact on public perception.

Depending on decision maker priorities, different fuel cycle scenarios would be preferable in the UK. If improving the public perception of waste and proliferation resistance is a priority then FULL-LO-TRU scenarios would be a priority. If priorities include new build LWRs whilst reducing the burden of new build LWRs on a repository, FEED-LO-AM scenarios would be preferable. FEED-LO-AM scenarios do not commit to a long-term SFR fuel cycle but can off-set new build LWRs whilst reducing repository size and radiotoxicity. If new build reactors, the burden on a repository, and proliferation resistance are priorities then FEED-LO-TRU scenarios would be preferable. FEED-LO-TRU scenarios have the same advantages as FEED-LO-AM scenarios but have a lower TRL and improved intrinsic proliferation resistance. If reducing dependence on natural uranium is a priority then transitioning to SFR scenarios is the UK R&D roadmap would be preferable [21]. By transitioning to a 100% SFRs fuel cycle with CR = 1 the demand on natural uranium can be capped.

Any scenarios in this chapter which have significant improvements in waste performance over direct disposal, will have relatively small improvements in waste performance if the addition of 16 GWe of new build SNF is included in the direct disposal scenario. Work in Chapter 9 considers extending the BURNER, FEED and FULL scenarios to include SNF from new build reactors. The improvement factors for FULL scenarios are plotted with and without the addition of new build LWR SNF in Figure 9.2.

8.5 Conclusions

This chapter describes the modelling of closed SFR fuel cycles to reduce the size of the UK's plutonium stockpile. Two scenarios were considered that aimed to improve waste performance: irradiate all stockpiled material by 2150 (FEED scenarios) and a closed fuel cycle running for as long as possible to maximise reduction of the stockpile (FULL scenarios). Waste performance is measured by the assessment criteria for repository size, radiotoxicity lifetime, 1000 year radiotoxicity and final transuranic inventory. Fuel cycles with long reprocessing timescales and no curium reprocessing are described as high TRL fuel cycles. Fuel cycles with short cooling times or curium reprocessing are described as low TRL fuel cycles.

Closed fuel cycles operated until 2150, FEED scenarios, can have benefits over BURNER scenarios (Chapter 7), however, this is heavily dependent on fuel cycle parameters. Long-term operation of a closed fuel cycle for maximum stockpile reduction, FULL scenarios, have a significant improvement on waste performance criteria. The potential impact of FULL scenarios is heavily dependent on fuel cycle parameters with the largest improvements in waste performance coming from low TRL fuel cycles. The largest improvement in waste performance is a result of fuel cycles that maximise recycling of the stockpile, the best way to do this is to maximise the availability of fuel to operate more reactors. Maximising fuel availability requires short cooling times and the recycling of all transuranics. To achieve significant improvement in waste performance, americium must be reprocessed (AM or TRU scenarios). Low-CR closed fuel cycles that only recycle plutonium (PU) have a negative impact on 1000 year radiotoxicity and repository size, negating any other positive attributes the fuel cycle may have.

FEED scenarios must perform significantly better than the best BURNER scenarios (BURNER-AM) to justify the cost in developing a closed fuel cycle. Only the FEED-LO-AM and FEED-LO-TRU scenarios achieve significantly better waste performance than BURNER-AM scenarios, with improvement factors as high as 2.02 and 2.54 for repository size and 1000 year radiotoxicity, when LWR offset is considered. For FEED scenarios to have significantly better waste performance than BURNER

scenarios, three main factors are required: Short cooling times, the inclusion of americium in the fuel, and operating 2-4 times as many SFRs than BURNER scenarios. As a result of short cooling times and americium recycling, the FEED-LO-AM and FEED-LO-TRU scenarios have low TRLs and the fuel cycle development requirements are high. Higher TRL fuel cycles with long cooling times (FEED-HI), or plutonium only reprocessing (FEED-PU) achieve small, or negative, improvements in waste performance when compared to BURNER-AM scenarios. Therefore, the choice between a BURNER-AM scenario or FEED-LO closed fuel cycle comes down to the importance of waste performance criteria compared to the cost of developing reprocessing and a lower TRL fuel cycle. The choice of metallic (ZR) or MOX fuel does not have a significant impact on waste performance over the timescale of BURNER or FEED scenarios.

This study has illustrated that the waste performance of stockpile reduction scenarios is dependent on fuel cycle parameters. Previous studies have focused on maximising stockpile reduction by operating low-CR SFR fuel cycles for 300 years, using low TRL fuel cycles with short cooling times and recycling all transuranics (FULL-LO-TRU). Over 300 years may be an unrealistic timescale to consider a fuel cycle scenario, but it warrants academic study to determine the maximum improvement in waste performance. Similarly, FULL-LO-TRU fuel cycle scenarios were modelled in the literature and in this study, but in a UK context, and achieved similar reductions in repository size (2.20 – 5.05) and waste lifetime (6.74 – 12.08) over the direct disposal scenario. More feasible, higher TRL fuel cycles were also considered in this study using long cooling times, and plutonium and americium recycling (FULL-HI-AM). The use of higher TRL scenarios for stockpile reduction scenarios is under represented in the literature. The higher TRL, FULL-HI-AM scenarios were operated for over 110 years longer than FULL-LO-TRU scenarios and achieved smaller improvements in waste performance, with a maximum improvement factor of 1.39 for repository size and 4.46 for waste lifetime. For FULL-HI-AM scenarios, 1000 year radiotoxicity and radiotoxicity lifetimes were still improved over the direct disposal scenario, but repository size was similar to that of direct disposal. Committing to a closed fuel cycle for more than 300 years is unlikely as current UK nuclear policy does not extend past 2050, 25 years, however to make such a long fuel cycle worth while it can be assumed that a

significant reduction in repository size would be required, in addition to other measures of waste performance. To ensure a significant improvement in repository size in a short timescale, a short cooling time is necessary which makes the development of pyro-reprocessing techniques preferable.

Plutonium only reprocessing scenarios (PU) lead to repositories larger than the direct disposal scenario, with the lowest improvement factor of 0.56, and minimal improvements in 1000 year radiotoxicity, with an improvement factor of 1.08. The main benefits of PU scenarios are the reduced radiotoxicity lifetime and reduced stockpile inventory, however, these benefits are small compared to the need to develop a closed fuel cycle. Alternatively, PWR MOX has a higher TRL than PU scenarios and can effectively reduce the stockpile in a short timescale, with similar repository size to PU scenarios. There are no substantial benefits to a low-CR, PU scenario over PWR MOX. As such, only high-CR plutonium recycling scenarios should be considered, that aim to reduce dependence on natural uranium. High-CR plutonium recycling scenarios have been thoroughly covered in previous research on UK R&D pathways and were not considered as part of this study [127].

The greatest improvement in waste performance was achieved with all transuranics reprocessed and the most recycling of stockpiled material through low-CR SFRs. Fuel cycles that maximise recycling through an SFR do so by increasing the availability of fuel in the fuel cycle in two ways. Firstly, short cooling times reduce buffered material in cooling ponds and increase fuel availability. Secondly, fuel cycles which reprocess all transuranics increase the mass of fuel available for reactor operation by reducing the mass of transuranics sent to the waste stream at reprocessing. Therefore short cooling times and recycling all TRUs are the best way to increase reactor operation and maximise stockpile reduction. In addition, the maximum reactor operation in a FULL scenarios is limited by the total fuel needed to operate the final generation of reactors. By reducing the size and inventory of a reactor, the last reactors in a fuel cycle can be operated for longer, as a result of lower fuel requirements. As such, a smaller reactor size and inventory leads to more recycling and a better reduction in stockpile. Reactor inventory only becomes relevant when maximum stockpile reduction is considered, such as the FULL scenarios. Further reductions in the stockpile with smaller reactor inventories come at the expense of longer fuel cycle operation. As such,

fuel cycle timescale and acceptable waste performance levels are goals that should be outlined before choosing a fuel cycle scenario.

Overall, if significant improvements in waste performance are a priority, FULL-LO-TRU scenarios are the best option. The improvement factors as a results of FULL-LO-TRU scenarios, roughly an order of magnitude for waste performance factors, should have an impact on the public perception of nuclear waste, whilst minimising proliferation concerns. If reducing the burden of new build reactors on a repository is a priority, without a long-term commitment to a closed SFR fuel cycle then FEED-LO-AM scenarios are preferable. If the proliferation resistance of reprocessed fuel is a concern, then FEED-TRU scenarios could be preferable over FEED-AM scenarios to increase the intrinsic proliferation resistance of the fuel cycle, making reprocessed fuel more difficult to handle.

The key limitations of the results presented in this chapter include the approximation methods used for repository size and radiotoxicity lifetime, as well as the lack of quantitative assessment of proliferation, development needs and cost. These have been summarised previously, see Section 7.5. There are three limitation specific to this chapter. First the FULL-MOX and FULL-ZR scenarios are not directly comparable due to different TRU inventories leading to longer operation and more recycling of transuranics in FULL-ZR scenarios. Further work should investigate the use of similar transuranic inventory reactors for FULL scenarios so that the advantages of FULL-ZR and FULL-MOX scenarios are directly comparable. Second, further work should investigate fuel cycle facility and transport requirements, particularly shielding, to compare facility requirements for different recycling scenarios. Finally all improvement factors determined in this chapter will be smaller if all 16 GWe of new build reactor SNF is directly disposed of, see Figure 9.2 in the following chapter. Extending the fuel cycle scenarios in this chapter to include new build SNF as a fuel feed will be considered in Chapter 9.

Key findings:

- Improvements in closed fuel cycle waste performance require americium reprocessing.
- FEED closed SFR fuel cycle scenarios, operated to 2150, have a significant improvements over BURNER scenarios if low TRL FEED scenarios are used with short reprocessing cooling times and americium reprocessing.
- FEED scenario waste performance is similar for metallic or MOX fuelled reactors.
- Curium reprocessing in FEED scenarios has a minimal impact on waste performance results by 2150.
- FULL scenarios with long reprocessing cooling times would not be worth while due to long timescales and relatively small improvements in waste performance.
- The best case, FULL-LO-TRU scenarios, have improvement factors close to an order of magnitude for waste performance. Order of magnitude improvements in waste performance may help improve public perception of nuclear waste.
- Increasing the fuel available to operate reactors, with short cooling times and all transuranics recycling, is the best way to ensure significant waste performance improvements.

Chapter 9

Plutonium and new build SNF disposition with SFRs

9.1 Introduction

This chapter considers the use of SFRs and their ability to reduce the UK's plutonium stockpile and minimise the waste generated from 16.5 GWe of new build LWRs. Once-through SFR BURNERS and closed fuel cycles were considered using the UK's plutonium stockpile and reprocessed SNF from new build LWRs as SFR fuel feeds. Throughout this chapter, the general term 'stockpile' will be used to refer to the UK's plutonium stockpile with the addition of reprocessed SNF from new build LWRs, unless otherwise stated.

The UK plans to build 16 GWe of new build LWRs by 2050. The UK R&D strategy has considered pathways with 16 to 75 GWe of nuclear power in once-through fuel cycles and closed fuel cycles. Closed fuel cycle pathways included reprocessing of new build SNF and the potential re-use of TRUs in SFRs. As well as new build LWRs, the UK is considering different options for UK plutonium stockpile disposition, including a once-through SFR BURNER.

Chapter 8 considered the impact of closed SFR fuel cycles on the UK's plutonium stockpile. Any significant improvements in waste performance of SFR fuel cycles, over the direct disposal of the plutonium stockpile, were dependent on fuel cycle parameters that had large technology development requirements. Any waste performance improvements in the previous chapters would be relatively small if 16.5 GWe of new

build SNF is included in a repository, Figure 9.2.

To address these problems, SFR fuel cycles from Chapter 7 and 8 (using the UK's plutonium stockpile as the only fuel feed) were extended to include reprocessed SNF from 16.5 GWe of new build LWRs as a fuel feed. To ensure SFRs have the greatest impact when reducing reprocessed material, new build SNF was prioritised as a fuel feed over the plutonium stockpile to minimise the decay of fissile ^{241}Pu to ^{241}Am. Prioritising new build SNF will lead to the most effective reduction of reprocessed material, with an aim to get significant improvements in waste performance for low TRL and high TRL fuel cycle scenarios.

This chapter aims to answer similar questions on waste performance to the previous two chapters, but extended to include SNF from new build LWRs as a fuel feed. It is expected that some higher TRL fuel cycles, that were unsuitable for plutonium stockpile disposition, may have better waste performance if used in a fuel cycle scenario for plutonium stockpile and new build SNF disposition. This chapter aims to answer the following questions:

1. Will once-through SFR BURNERS have a significant impact on repository size (greater than a 20% reduction), 1000 year radiotoxicity (greater than a factor of 2 reduction), or radiotoxicity lifetime (greater than a factor of 10 reduction)?

2. How much of an improvement will running SFRs with reprocessing until 2200 (FEED scenarios) have over once-through BURNERS?

3. Does the reprocessing of curium have an impact on results up to 2200?

4. Given reprocessing until a maximum stockpile reduction is achieved (FULL scenarios), what is the maximum improvement over the direct disposal scenario, and over what timescale?

5. Do low-CR plutonium recycling scenarios (PU scenarios) have any impact on radiotoxicity or repository size compared to the direct disposal scenario?

6. With new build LWR SNF and the plutonium stockpile irradiated in SFR fuel cycles, are there any favourable scenarios that were not favourable when considering plutonium stockpile disposition alone in Chapter 7 and 8?

9.2 Methods

This methods section will discuss two things, the set up of the fuel cycle model in ORION and the fuel cycle variables that were tested. In addition, the assessment criteria used to measure the performance of each fuel cycle is summarised in Section 7.2.3. ORION was used to test a range of fuel cycle variables and reactor designs for once-through SFR fuel cycles (BURNER scenarios) and two closed SFR fuel cycles: the FEED equilibrium scenario and the FULL equilibrium scenario.

9.2.1 ORION model

The closed fuel cycle set up in ORION is illustrated in Figure 9.1. As described in Chapter 5, the fuel feed from AGR and Magnox reactor operating histories was used to produce a representative plutonium stockpile in the 'Pu Stock' buffer. The fuel feed from new build LWRs was used to produce a representative SNF inventory from 16.5 GWe of new build LWRs in the 'LWR Feed' buffer. The freshest fuel, 'LWR Feed' was used as the primary fuel feed, with 'Pu Stock' as the secondary fuel feed, and uranium enrichment tails 'U Tails' as the carrier feed. Fuel was fabricated for the inner and outer reactor regions and irradiated in 'SFR IF' and 'SFR OF' using reactor parameters, cross-section and fluxes from ERANOS, discussed below. Spent fuel was cooled and reprocessed in 'SFR Rep,' sending the recycled elements to 'Rep Feed' and waste to the 'HLW Buff' buffer. When there is enough fissile material available, the 'Rep Feed' buffer becomes the primary fuel feed, ahead of 'LWR Feed' and 'Pu Stock.' If there is a lack of material to fabricate fuel from 'Rep Feed' it is topped up with material from 'LWR Feed' and 'Pu Stock.' More detailed discussion of ORION fuel cycle models can be found in Section 4.1.2.

Three types of fuel cycle scenario were modelled, with sub-scenarios described in the following section:

- BURNER scenarios – Operating once-through SFRs to irradiate all stockpiled material by 2150;
- FEED scenarios – Operating enough SFRs to irradiate all stockpiled material by 2150, Figure 4.5;

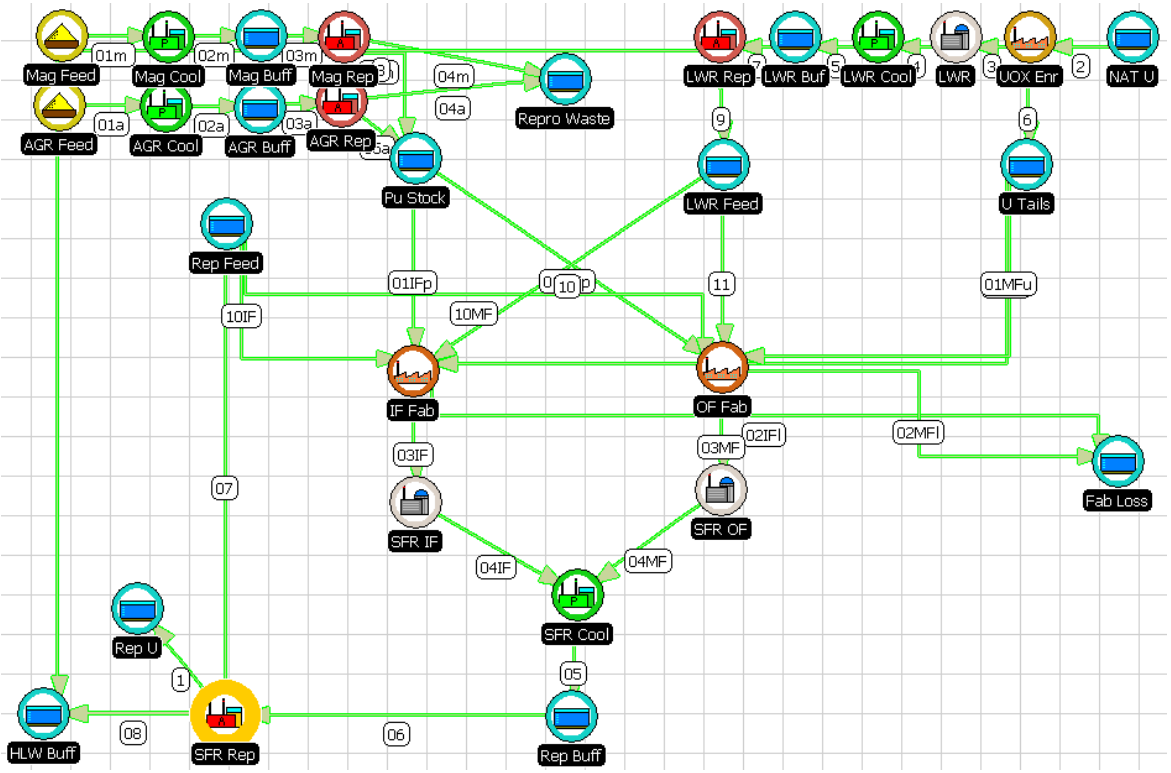


Figure 9.1: ORION model for a closed SFR fuel cycle using the UK's plutonium stockpile and reprocessed SNF from new build reactors as fuel feeds.

- FULL scenarios – Operating several generations of SFRs, reducing the number of reactors in each generation due to the reduction of fuel in the fuel cycle, Figure 4.5. The number of SFRs was reduced in each generation until the maximum stockpile reduction was achieved and no more reactors could be operated.

9.2.2 Parameters

Fuel cycle variables tested were based on different reactor designs, reprocessing options and timescales. Summarised as:

- Three timescales – BURNER scenarios modelled until 2150, FEED scenarios modelled until 2200, and FULL scenarios were modelled until material ran out (end dates in Table 9.2).
- Three reprocessed fuel feeds – Reprocessing of plutonium (PU scenarios), plutonium and americium reprocessing (AM scenario), or all transuranic reprocessing (TRU scenario).
- Two reprocessing variables – High cooling times and high losses (HI scenario),

Table 9.1: SFR parameters used for BURNER fuel cycle models with the UK's plutonium and new build SNF as fuel feeds. Assumes an 85% capacity factor for SFRs.

BURNER Scenario	**Zr PU**	**Zr AM**	**MOX PU**	**MOX AM**
TRL	6	4	9	5
CR	0.64	0.99	0.53	1.01
TRU/U (%)	30.0	30.0	38.4	38.4
GWe	0.38	0.38	0.38	0.38
GWt	1.0	1.0	1.0	1.0
GWy(e)	142.12	140.18	0.44	104.98
Peak #Reac	4	4	3	3
Cycle length (EFPD)	211	252	176	215

or low cooling times and low losses (LO scenario), Table 4.3.

- Two reactor designs – Metallic (Zr) and MOX fuelled reactor were tested for each scenario. Reactor parameters and self-shielded cross-sections were taken from the work completed in ERANOS, Chapter 6. The reactor parameters used in each ORION scenario are shown in Table 9.1 for the BURNER scenarios and in Table 9.2 for FEED and FULL scenarios.

Each varied parameter has its own fuel cycle scenario that was modelled in ORION, with reactor parameters and self-shielded cross-sections taken from the work completed in ERANOS, Chapter 6. An overview of assessment criteria used for fuel cycle scenarios can be found in Section 7.2.3.

A total of 28 SFR fuel cycle scenarios were modelled in this chapter. The once-through BURNER scenario has four sub-scenarios based on reactor design and fuel feed, Table 9.1. The FEED and FULL closed fuel cycle scenarios have twelve sub-scenarios, six based on reactor design and fuel feed, Table 9.2, with two variations based on reprocessing losses and cooling time. A stockpile direct disposal scenario was also modelled to compare the relative impact SFR scenarios had on the final inventory sent to a repository, repository size and radiotoxicity.

Table 9.2: SFR parameters used for FEED and FULL fuel cycle models with the UK's plutonium and new build SNF as fuel feeds. Assumes an 85% capacity factor for SFRs.

FEED Scenario	**Zr PU**	**Zr AM**	**Zr TRU**	**MOX PU**	**MOX AM**	**MOX TRU**
TRL	6	4	3	7-8	5	3
CR	0.73	0.94	0.94	0.68	0.99	1.00
TRU/HM (%)	30.0	30.0	30.0	38.4	38.4	38.4
MA/HM (%)	-	2.8	3.3	-	4.0	4.8
GWe	0.38	0.38	0.38	0.38	0.38	0.38
GWt	1.0	1.0	1.0	1.0	1.0	1.0
Cycle length (EFPD)	244	271	271	244	263	263
Lo GWy(e)	421.52	534.57	563.64	289.09	394.06	423.13
Hi GWy(e)	313.31	394.06	394.06	218.03	289.09	300.39
FULL Scenario	**PU**	**AM**	**TRU**	**PU**	**AM**	**TRU**
TRL	6	4	3	7-8	5	3
CR	0.81	0.96	0.95	0.81	1.05	1.04
TRU/HM (%)	30	30	30	38.4	38.4	38.4
MA/HM (%)	-	2.7	3.5	-	4.3	5.5
GWe	0.38	0.38	0.38	0.38	0.38	0.38
GWt	1.0	1.0	1.0	1.0	1.0	1.0
Cycle length (EFPD)	293	308	308	297	327	327
Lo GWy(e)	1027.14	1434.12	1531.02	852.72	1240.32	1240.32
Lo End Date	2400	2500	2600	2400	2500	2600
Hi GWy(e)	794.58	1124.04	1162.8	639.54	891.48	833.34
Hi End Date	2600	2700	2700	2500	2700	2700

9.3 Results

A summary of fuel cycle scenario results is tabulated in Table 9.8 and 9.9. Summarised results are given as the relative improvement factor over a reference case (e.g. direct disposal, PWR MOX) for each of assessment criteria so the relative merits of each scenario can be compared to their TRL.

Improvements in waste performance for SFR fuel cycles using the UK's plutonium stockpile, discussed in Chapter 8, can be large when compared to the stockpile direct disposal of the plutonium stockpile alone. If the direct disposal of 16.5 GWe of new build SNF is included, with SFRs only using the UK's plutonium stockpile as fuel, the waste performance of SFR fuel cycles is small compared to direct disposal. Figure 9.2 is an example of repository size and 1000 year radiotoxicity results for FULL scenarios in Chapter 8. The left hand figures include SNF from new build LWRs in a repository, the right hand figures do not. With the inclusion of new build SNF, the impact the FULL scenario on waste performance is small due to the large contribution of new build SNF. As such, fuel cycle scenarios in this chapter consider the use of the plutonium stockpile and new build SNF as fuel feeds for SFRs.

9.3.1 Masses

The final fuel cycle inventory of TRUs sent to a repository, after 50 years of cooling, is given in Table 9.3 and 9.4.

Table 9.3: Final masses (tHM) of transuranic elements to be sent to a repository after 50 years of cooling in BURNER scenarios that use the UK's plutonium and new build SNF as fuel feeds.

	Stock	**Zr Lo-PU**	**Zr Lo-AM**	**MOX Lo-PU**	**MOX Lo-AM**
TRU	349.76	304.41	303.81	271.80	285.02
Pu	295.94	246.68	257.81	215.93	239.28
Np	22.68	22.41	20.53	22.18	20.53
Am	30.99	35.08	25.04	33.48	24.83
Cm	0.15	0.23	0.44	0.20	0.39

BURNER-AM scenarios in this chapter lead to a net reduction in americium inventories. This was not the case in BURNER-AM scenarios that only considered UK

Table 9.4: Final masses (tHM) of transuranic elements to be sent to a repository after 50 years of cooling in FEED, FULL and WASTE scenarios that use the UK's plutonium and new build SNF as fuel feeds.

	Zr Lo-PU	**Zr Lo-AM**	**Zr Lo-TRU**	**MOX Hi-PU**	**MOX Hi-AM**	**MOX Hi-TRU**	**Zr Lo-PU**	**Zr Lo-AM**	**Zr Lo-TRU**	**MOX Hi-PU**	**MOX Hi-AM**	**MOX Hi-TRU**
FEED Scenario												
TRU	118.53	104.69	98.66	102.55	85.42	76.53	149.23	128.96	128.43	127.80	107.12	102.34
Pu	50.32	77.07	73.44	40.17	60.04	53.07	78.60	98.35	99.75	63.64	78.21	75.31
Np	25.05	18.86	17.22	24.12	18.45	17.09	25.79	19.28	17.79	24.71	18.97	17.55
Am	42.59	6.74	7.24	37.84	5.22	5.77	44.43	9.97	10.16	39.13	8.80	8.85
Cm	0.57	2.02	0.77	0.42	1.71	0.62	0.41	1.37	0.73	0.32	1.14	0.63
FULL Scenario												
TRU	148.88	77.91	36.54	161.22	86.90	39.28	183.50	77.90	47.89	197.25	86.35	60.99
Pu	25.39	50.13	17.41	28.40	58.60	19.38	38.24	45.23	25.02	44.83	50.86	34.60
Np	46.54	21.00	16.97	48.61	21.08	17.09	63.51	24.63	18.18	61.26	26.13	18.72
Am	75.97	2.08	1.63	83.38	2.88	2.18	81.04	4.66	3.76	90.58	6.26	6.65
Cm	0.97	4.69	0.54	0.83	4.34	0.64	0.71	3.39	0.93	0.58	3.10	1.02
WASTE Stream Only												
TRU	138.82	65.43	25.66	147.92	71.27	27.16	157.20	55.31	30.70	163.50	59.12	32.26
Pu	15.82	39.16	7.92	15.78	45.12	9.09	12.73	25.71	10.23	12.37	27.47	10.87
Np	46.53	20.96	16.91	48.58	20.98	17.01	63.48	24.45	18.00	61.20	26.08	18.30
Am	75.51	0.62	0.56	82.73	0.83	0.75	80.28	1.77	1.74	89.36	2.48	2.39
Cm	0.97	4.69	0.28	0.83	4.34	0.31	0.71	3.39	0.74	0.58	3.10	0.71

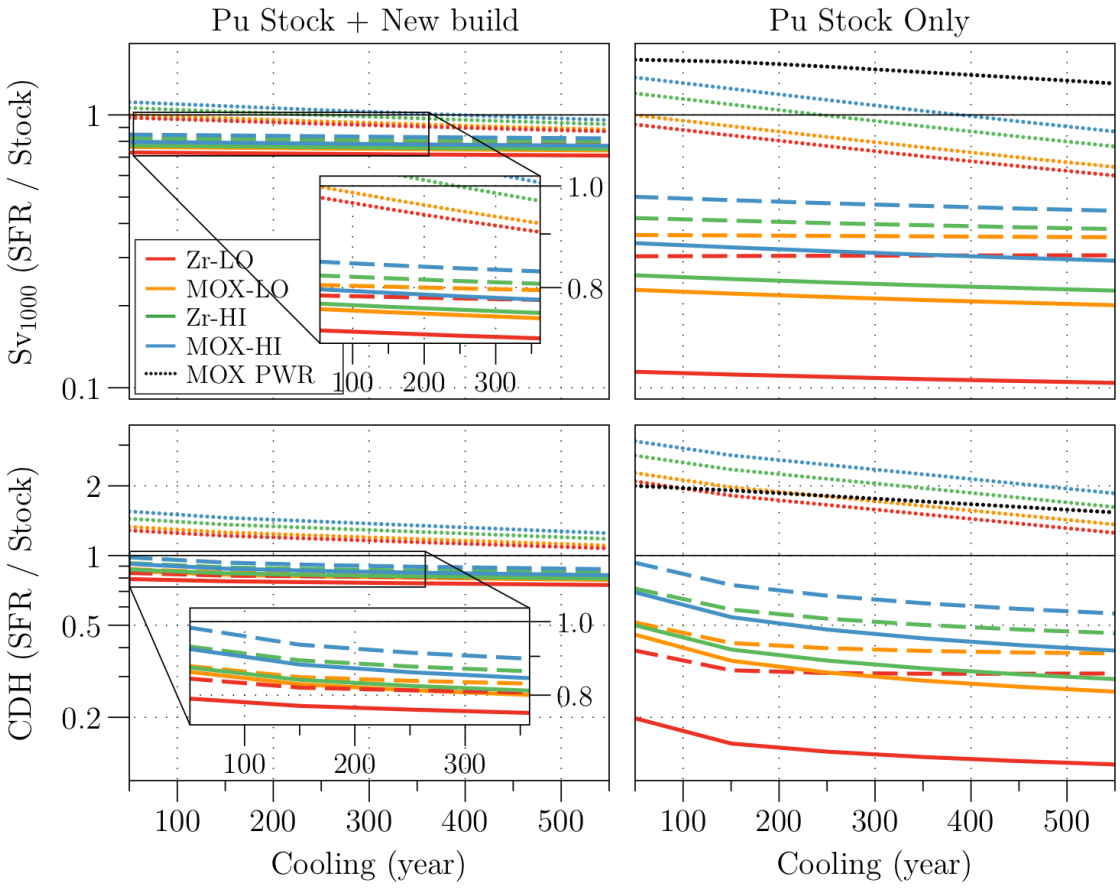


Figure 9.2: Relative 1000 year radiotoxicity and repository size for FULL scenarios in Chapter 8, where the SFR fuel cycles only use the plutonium stockpile as fuel. Results are given relative to the plutonium stockpile (right), and the plutonium stockpile with new build LWRs SNF (left). Fine dashed lines represent PU scenarios, thick dashed lines represent AM scenarios, and solid lines represent TRU scenarios.

plutonium disposition in Chapter 7.

The greatest overall TRU mass reductions were in scenarios where there was more recycling of TRUs through a low CR SFR. Each pass through an SFR leads to fission and transmutation, therefore more recycling through an SFR leads to a better overall reduction in stockpiled material. For FULL scenarios, plutonium reduction is more effective in the FULL-TRU scenarios than FULL-PU scenarios, as FULL-PU scenarios remove curium at reprocessing which decays to plutonium in the reprocessing waste stream. In FEED scenarios the FEED-PU scenario reduces plutonium more than the FEED-TRU scenario. There is less recycling in the FEED scenarios compared to the FULL scenarios, therefore less curium is sent to the waste stream. The more effective plutonium reduction in FEED-PU reactors, compared to FEED-TRU reactors, has more of an impact on final plutonium inventory than the removal and decay of curium

in the reprocessing waste stream. AM scenarios reduce the plutonium inventory less than the equivalent PU or TRU scenarios, due to americium in the fuel leading to the capture and the production of curium which decays to plutonium in the reprocessing waste stream. Most notably ^{243}Am transmutes to ^{244}Cm which is removed at reprocessing and decays to ^{240}Pu in the waste stream.

The greatest americium reduction was in FULL-TRU scenarios as there was a longer timescale for more recycling and all TRUs were reprocessed so there was greater fuel availability to operate more SFRs. For FEED scenarios, FEED-AM scenarios had a marginally better americium reduction than FEED-TRU scenarios. FEED-PU and FULL-PU scenarios lead to an overall increase in americium, as it is produced through plutonium capture in a reactor and removed at reprocessing, building up in the reprocessing waste stream.

The total amount of curium in the fuel cycle increases for all scenarios. AM scenarios have the largest build-up of curium, as curium is produced by capture in americium, then removed at reprocessing. PU scenarios are better at preventing curium build-up than AM scenarios because americium is removed at reprocessing, therefore americium in the fuel is minimised, preventing capture and the subsequent the build-up of curium. FULL-LO-TRU scenarios have the lowest build-up of curium as there is a lot of recycling and low losses, allowing for effective transmutation of curium and preventing a large build-up. FULL-HI-PU scenarios have lower curium build-up than FULL-HI-TRU scenarios, due to high (5%) curium losses at reprocessing for HI scenarios. Curium losses to the repossessing waste stream for FULL-HI-TRU scenarios are greater than the amount of curium generated in the FULL-HI-PU scenario. Similarly, FEED-PU scenarios generate less curium than the equivalent FEED-TRU scenario.

Comparing equivalent LOW and HIGH scenarios, there is less fuel available in HIGH scenarios, due to longer cooling times, which lead to more fuel being buffered in cooling ponds. With less fuel available, fewer reactors can be operated and fuel is recycled through reactors a fewer number of times in HIGH scenarios. Less recycling in HIGH scenarios leads to a smaller stockpile reduction than the equivalent LOW scenarios. HIGH scenarios also leads to considerably more americium than LOW scenarios, due to longer cooling times, resulting in more decay of fissile ^{241}Pu to ^{241}Am. There is less curium at the end of HIGH-PU and HIGH-AM scenarios compared to

the equivalent LOW scenarios as there is less recycling, leading to less of a build-up of curium. For FEED-TRU scenarios, curium inventories are similar for equivalent LOW and HIGH scenarios as there is a minimal build up of curium over the time-scale of FEED scenarios. However, for FULL-HI-TRU scenarios the higher curium losses at reprocessing leads to a greater final inventory of curium than the FULL-LO-TRU scenarios.

Comparing ZR and MOX fuelled scenarios, FEED-MOX and BURNER-MOX scenarios reduce inventories more than the equivalent ZR scenarios. However, FULL-ZR scenarios reduce inventories more than the equivalent FULL-MOX scenarios. MOX is better at reducing the stockpile for FEED and BURNER scenarios because MOX reactors have a higher enrichment and lower fuel volume than the equivalent ZR reactors, leading to high transmutation rates and low rates of capture in ^{238}U. When considering the FULL-ZR scenarios, reactors reduce stockpiles more than the equivalent FULL-MOX scenarios due to lower CRs and more recycling through the reactors, leading to a better reduction. Lower CRs for the FULL-ZR reactors compared to the equivalent FULL-MOX reactors was a result of the harder neutron spectrum leading to a lower proportion of MAs in the equilibrium fuel feed. More recycling in FULL-ZR scenarios is due to the lower TRU inventories in the core than for MOX reactors, leaving more fuel available to operate reactors.

One unexpected result is that FULL-HI-AM scenarios have a greater reduction in plutonium than FULL-LO-AM scenarios. It was assumed that the long fuel cycle time and long cooling prior to reprocessing in FULL-HI-AM scenarios resulted in more curium decaying to plutonium in the reprocessing buffer. As such, less curium was sent to the reprocessing waste stream in FULL-HI-AM scenarios, resulting in less plutonium in the reprocessing waste stream and an overall smaller plutonium inventory at the end of the fuel cycle. However, the lower final plutonium inventory in FULL-HI-AM scenarios requires a fuel cycle that is 200 years longer than the FULL-LO-AM scenario and generates less electricity.

9.3.2 Radiotoxicity

Elemental contributions to radiotoxicity over time are discussed in Section 8.3.2.1. They are not discussed in this chapter as the results are similar to those presented in

Table 9.5: Years taken for the radiotoxicity of final inventories to decay to the same radiotoxicity as natural uranium. Results are presented for BURNER, FEED, FULL and WASTE scenarios using the UK's plutonium stockpile and new build SNF as fuel feeds. UOX PWR SNF takes 290,000 years.

	Zr PU (years)	**Zr AM (years)**	**Zr TRU (years)**	**MOX PU (years)**	**MOX AM (years)**	**MOX TRU (years)**
BURNER						
	170,000	310,000	-	160,000	300,000	-
FEED						
LO	69,000	61,000	62,000	66,000	55,000	52,000
HI	84,000	81,000	84,000	80,000	74,000	73,000
FULL						
LO	63,000	30,000	14,000	71,000	33,000	17,000
HI	66,000	32,000	22,000	74,000	36,000	30,000
WASTE Only						
LO	59,000	26,000	4,900	67,000	29,000	8,200
HI	56,000	24,000	11,000	62,000	27,000	14,000

Chapter 8.

9.3.2.1 Radiotoxicity lifetime

Rather than describing all results, important results and trends will be described in this section. Figure 9.3 shows radiotoxicity per GWy(e) relative to natural uranium for each fuel cycle scenario. This shows the time it takes for final fuel cycle inventories to decay to natural uranium levels. The time it takes for the radiotoxicity to drop below the level of natural uranium is shown in Table 9.5, described here as the lifetime of radiotoxicity. In this thesis, a significant reduction in the lifetime of radiotoxicity was considered to be an order of magnitude less that UOX PWR, that is, less than 29,000 years. The fine dashed lines in figure 9.3 represent PU scenarios, long-dashed lines represent AM scenarios and the solid lines represent TRU scenarios.

Considering the once-through BURNER scenarios in Figure 9.3, BURNER-MOX and BURNER-ZR lifetime results are similar. The lifetime of waste is shorter for BURNER-PU scenarios than BURNER-AM scenarios, due to BURNER-PU scenarios reducing the final plutonium inventory more than BURNER-AM scenarios. The lifetime of waste from BURNER-PU scenarios is roughly half that of the same generating

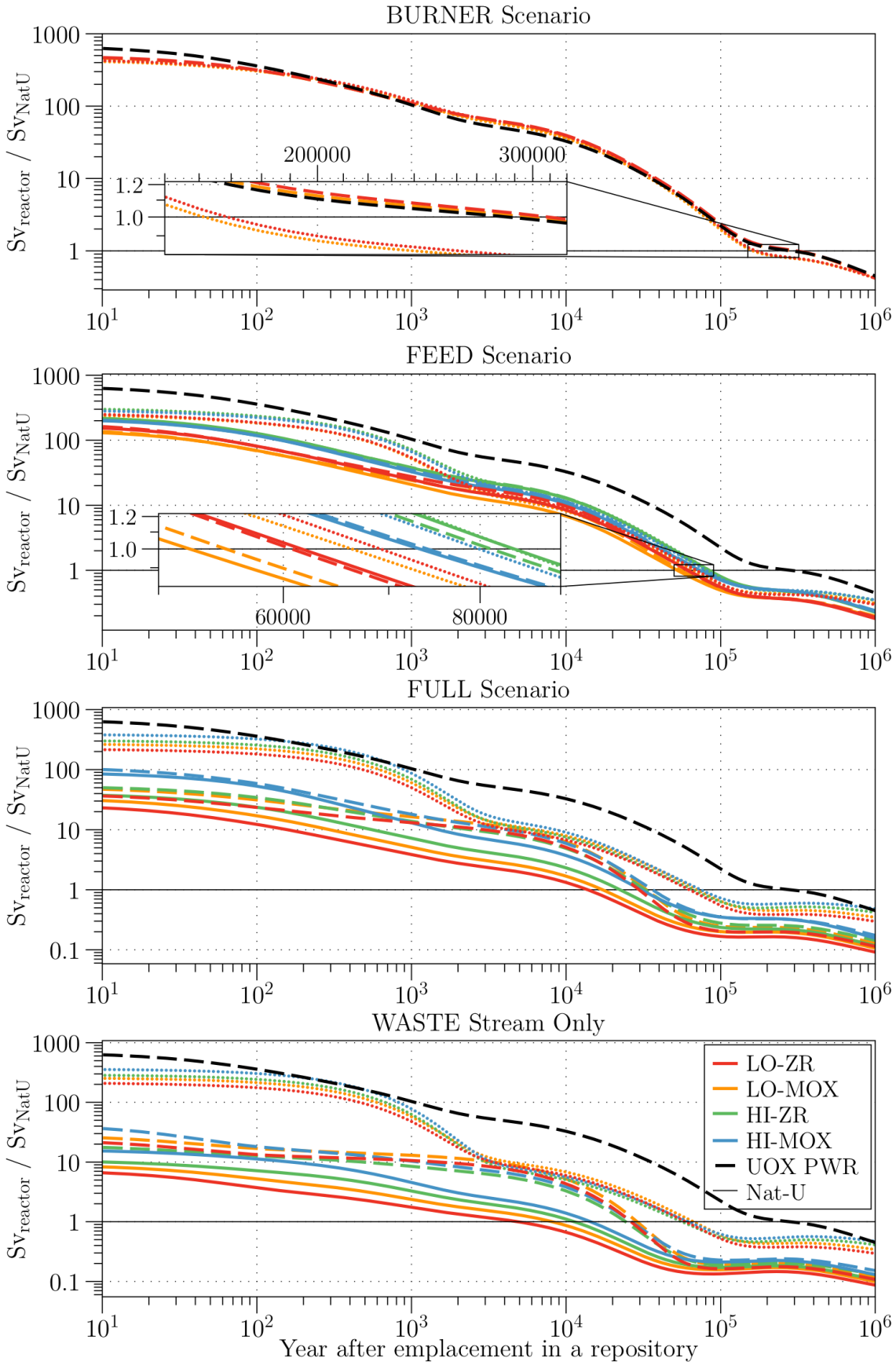


Figure 9.3: Radiotoxicity per GWy(e) for BURNER, FEED, FULL and WASTE scenarios after disposal in a repository. Radiotoxicities are given relative to the radiotoxicity of natural uranium required to fuel a UOX PWR. Fine dashed lines represent PU scenarios, thick dashed lines represent AM scenarios, solid line represents TRU scenarios.

capacity of UOX PWRs.

Considering the FEED scenarios in Figure 9.3, the lifetimes of FEED-MOX-AM and FEED-MOX-TRU scenarios are shorter than the equivalent FEED-ZR scenarios. Despite the FEED-MOX scenarios generating less power than the equivalent FEED-ZR scenarios, the reduction of plutonium and americium is greater per GWy(e), resulting in a shorter lifetime of waste. FEED-ZR-AM has a shorter lifetime than FEED-ZR-TRU as they have similar plutonium inventories, but FEED-ZR-AM has a lower americium inventory. Due to the large power generation and effective stockpile reduction in the FEED scenarios, the lifetime of all scenarios is shorter than for a UOX fuelled PWR, but they are not less than 29,000 years.

Considering the FULL scenarios in Figure 9.3, the radiotoxicity lifetimes are ordered such that all FULL-TRU scenarios have a shorter lifetime than all FULL-AM scenarios, and all FULL-PU scenarios have the longest lifetime. FULL-TRU scenarios have the shortest lifetime, some less than 29,000 years, due to the greatest stockpile reduction and greatest power generated. FULL-ZR scenarios always have shorter lifetimes than the equivalent FULL-MOX scenario, as a result their lower final inventory and greater power generation.

The WASTE stream only scenarios have the same order of results as in Chapter 8, but with shorter lifetimes. The analysis of these results can be found in Section 8.3.2.2, and does not warrant further discussion as the same factors are involved when considering WASTE streams only.

Radiotoxicity lifetime results, with LWR SNF included as a fuel feed, performs better than the equivalent scenarios using the plutonium stockpile only, discussed in Chapter 7 and 8. This is due to several factors, but most significantly the contribution of new build LWRs to power generation.

9.3.2.2 1000 year radiotoxicity

Figure 9.4 shows the total radiotoxicity relative to direct disposal scenarios at 1000 years (Sv_{1000}), which is assumed to be the earliest timescale for a disturbed repository. Scenario results are given relative to the stockpile direct disposal scenario, with and without the offset of once-through LWRs. The change in results over a period of 50 to 500 years shows how the radiotoxicity of different scenarios changes over time. In

this thesis a factor of two reduction in 1000 year radiotoxicity is considered to be a significant improvement.

For BURNER scenarios in Figure 9.4, the choice of BURNER-MOX or BURNER-ZR scenario makes a small difference to results and does not lead to a significant reduction in radiotoxicity. Including the offset of LWRs, the best case BURNER-MOX-AM scenario has a total radiotoxicity 20% less than the direct disposal scenario.

Considering the FEED scenarios in Figure 9.4, FEED-MOX scenarios have lower radiotoxicities than FEED-ZR scenarios. This is due to a greater reduction in plutonium in FEED-MOX scenarios compared to FEED-ZR scenarios. FEED-PU scenarios have greater radiotoxicities than FEED-AM and FEED-TRU scenarios due to the high americium inventory at the end of the fuel cycle, as shown in Figure 8.6. To get a significant reduction in radiotoxicity, americium must be recycled. Without americium reprocessing, plutonium capture produces americium which is removed at the reprocessing step and sent to the waste stream resulting in a large build-up of americium. As such, americium reprocessing is the most important factor in reducing radiotoxicity in FEED scenarios, independent of MOX, ZR, HIGH and LOW fuel cycle options. When the offset of LWRs are included, the choice of fuel cycle scenario has more of an impact, based on the total power generated in each scenario.

Considering FULL scenarios in Figure 9.4, the recycling of americium is still the most important factor, resulting in radiotoxicities 10 to 30% of the original stockpile. The FULL-ZR scenarios have lower radiotoxicities than the equivalent FULL-MOX scenarios due to lower final inventories. FULL-HI-AM scenarios have lower radiotoxicities than FULL-LO-AM scenarios as a result of lower final plutonium inventory. FULL-PU scenarios have very high radiotoxicities when compared to other FULL scenarios and FEED-PU scenarios, due to the production of large quantities of americium. However, the radiotoxicity of PU scenarios drops quickly over time, compared to the equivalent AM and TRU scenarios, due to the decay of americium.

Comparing the FULL scenarios to the WASTE stream only results in Figure 9.4, the WASTE radiotoxicities are lower than the equivalent FULL scenario. The FULL-TRU scenarios have lower radiotoxicities than the WASTE-AM scenarios. This is good considering the WASTE scenarios are not feasible and represent an upper bound to the performance of fuel cycle scenarios.

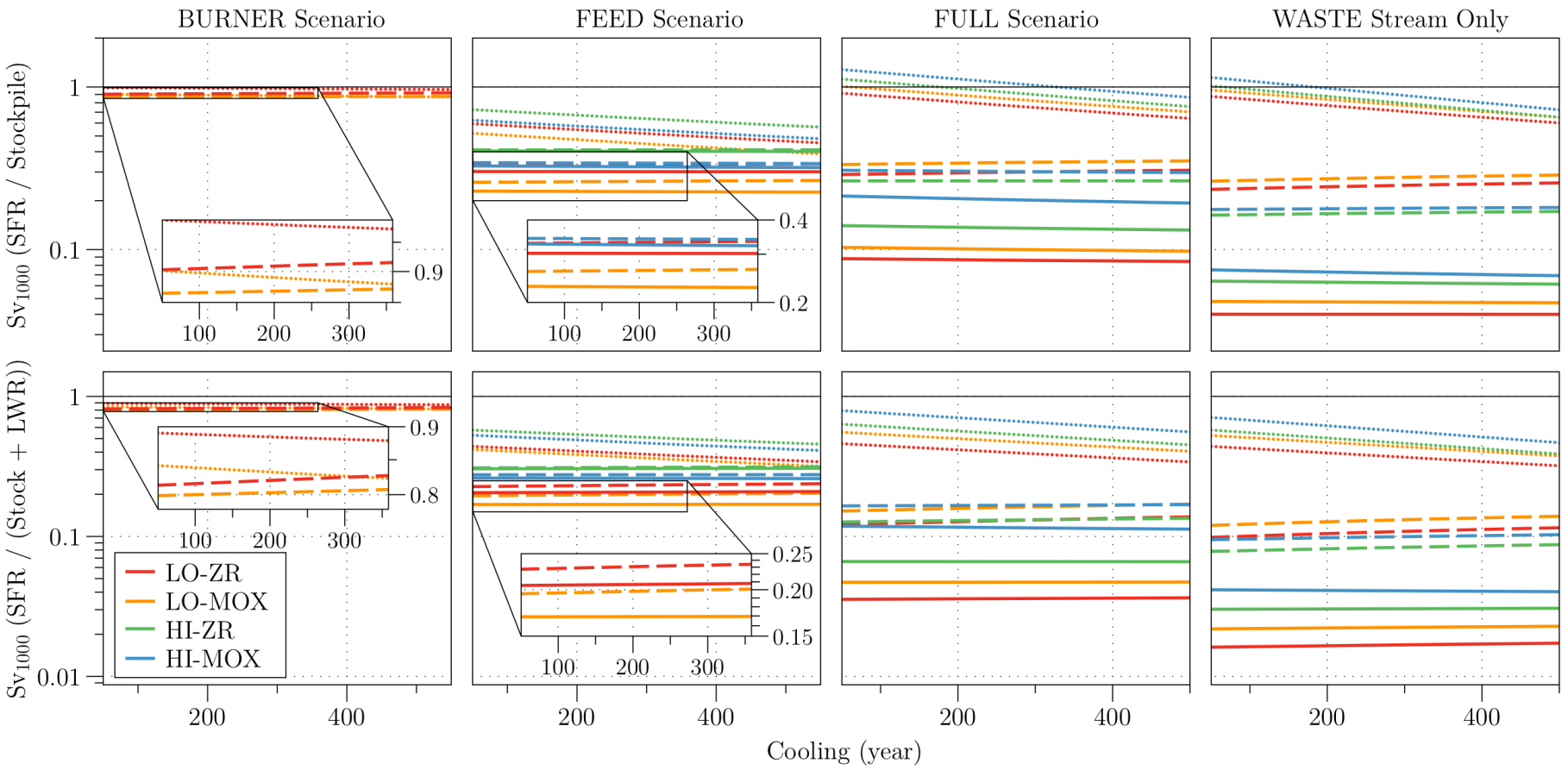


Figure 9.4: 1000 year radiotoxicity (Sv_{1000}) for BURNER, FEED, FULL and WASTE scenarios relative to direct disposal scenarios. Results in the upper figure are relative direct disposal of the UK's plutonium stockpile. Results in the lower figure are relative to a once-through PWR scenario (the UK's plutonium stockpile plus the same GWy(e) of PWR SNF). Fine dashed lines represent PU scenarios, thick dashed lines represent AM scenarios, solid line represents TRU scenarios.

9.3.3 Repository size

Figure 9.5 shows the cumulative decay heat (CDH) of each scenario, relative to the direct disposal reference cases. CDH is used to estimate relative repository size. The minimum cooling time of fuel cycle inventories before disposal was assumed to be 50 years and the longest 350 years. However, results were plotted up to 550 years to see how cooling time and the decay of short to intermediate lived components of SNF influence repository size. In this thesis a 20% reduction in repository size is considered significant, see Section 7.3.3.1.

Considering BURNER scenarios in Figure 9.5, BURNER-AM scenarios have the smallest repository size. However, to get a reduction in repository size BURNER-AM scenarios need at least 150 years of cooling. With 150 years of cooling BURNER-AM scenarios have a repository size approximately 10 to 20% smaller than the stockpile. When offset LWRs are included, BURNER-AM scenarios have a repository size approximately 20% smaller than the stockpile. BURNER-PU scenarios have large repositories close to the direct disposal scenario due to large americium inventories at the end of the fuel cycle. As such, BURNER-AM scenarios improve repository size but BURNER-PU scenarios do not.

Considering FEED scenarios in Figure 9.5, FEED-PU scenarios have 50 year cooled results similar to the BURNER scenarios. FEED-AM and FEED-TRU scenarios can reduce the a repository by more than a half when compared to the stockpile, and when offset LWRs are included the repository size can be less than a quarter of the direct disposal scenario. FEED-MOX scenarios have smaller repositories than the equivalent FEED-ZR scenarios due to the better reduction of TRUs. With the offset of LWRs included FEED-ZR and FEED-MOX scenarios are closer due to the greater power generated in FEED-ZR scenarios offsetting more LWR SNF. The equivalent FEED-TRU and FEED-AM scenarios have minimal differences due to similar americium inventories at the end of the fuel cycle. The most significant impact on FEED-AM and FEED-TRU scenarios is the choice of HIGH or LOW scenario, with FEED-HI having larger repositories. The difference between equivalent FEED-HI and FEED-LO scenarios is due to more fuel available to operate more reactors in FEED-LO scenarios and longer cooling times in FEED-HI scenarios generating more americium.

Considering FULL scenarios in Figure 9.5, FULL-TRU scenarios have the smallest

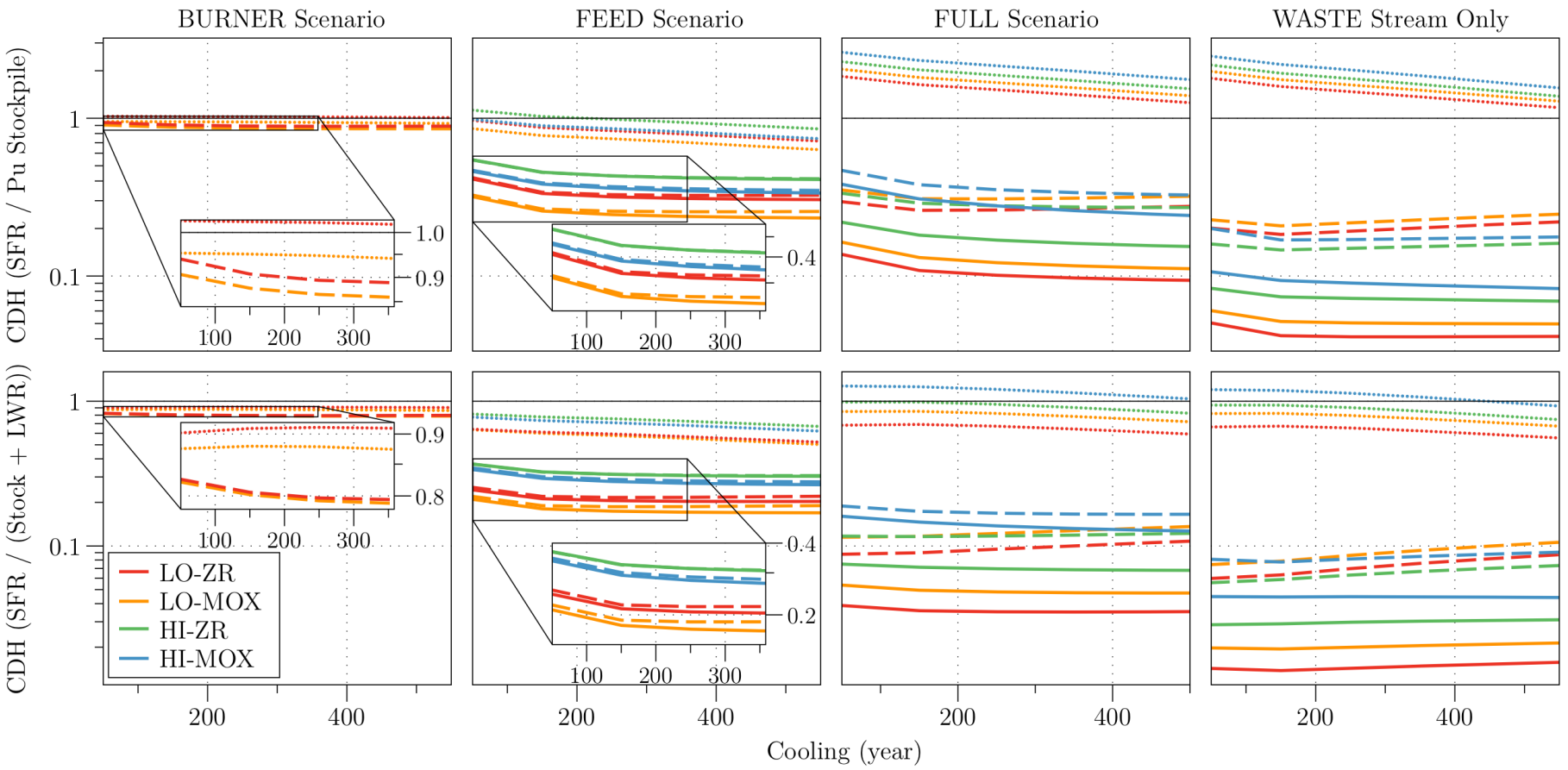


Figure 9.5: Relative repository size for BURNER, FEED, FULL and WASTE scenarios estimated by the CDH method and given relative to direct disposal scenarios. Results in the upper figure are relative direct disposal of the UK's plutonium stockpile. Results in the lower figure are relative to a once-through PWR scenario (the UK's plutonium stockpile plus the same GWy(e) of PWR SNF). Fine dashed lines represent PU scenarios, thick dashed lines represent AM scenarios, solid line represents TRU scenarios.

repositories, with an order of magnitude reduction in size compared to direct disposal scenarios. FULL-ZR scenarios have small repositories than the equivalent FULL-MOX scenario, due to smaller final inventories. Similarly, FULL-TRU scenarios have a smaller repositories size than the equivalent FULL-AM scenario as a result of lower plutonium inventories. Due to the large mount of americium generated in FULL-PU scenarios, repositories are large, even compared to FEED-PU scenarios.

Comparing FULL and WASTE results for repository size in Figure 9.5, it can be seen that they have the same trend as for Sv_{1000}, discussed in Section 9.3.2.2.

9.3.4 Bare sphere critical mass

The bare sphere critical mass for reprocessed fuel streams at the end of each fuel cycle scenario is presented in Table 9.6.

Table 9.6: Bare sphere critical mass (kg) for the waste streams of BURNER, FEED and FULL scenarios using the UK's plutonium stockpile and new build SNF as fuel feed. The initial stockpile of TRUs from the UK's plutonium and LWR SNF had a critical mass of 15 kg.

	Zr PU	**Zr AM**	**Zr TRU**	**MOX PU**	**MOX AM**	**MOX TRU**
BURNER						
	16.6	15.8	-	16.8	16.1	-
FEED						
Low	15.8	15.6	15.5	15.8	15.8	15.8
High	15.5	15.7	15.7	15.5	16.0	16.0
FULL						
Low	18.0	17.2	16.8	19.3	17.7	17.4
High	19.4	17.9	17.6	19.8	18.8	18.8

There is little to differentiate between the bare sphere critical masses of different scenarios. The bare sphere critical mass results were discussed in Section 8.3.4 and the same analysis applies to the results in this section due to identical trends.

9.4 Discussion

A summary of results for the key assessment criteria can be found in Table 9.9 for BURNER scenarios and Table 9.8 for FEED and FULL scenarios. Improvement factors over reference scenarios were tabulated for ease of comparing results. This chapter sought to answer several questions which were outlined in the introduction and discussed below.

Final TRU inventory mass results are not discussed in depth as repository size, radiotoxicity lifetime and 1000 year radiotoxicity are considered more important effects resulting from the final mass of transuranics. Bare sphere critical mass is also not discussed in depth as the scenarios have a relatively small effect on this criteria when compared to PWR MOX. Results for 1000 year radiotoxicity and repository size are only improved if americium is recycled (AM and TRU scenarios). As a result of the poor performance of PU scenarios, they are excluded from most analysis in this section, except when specifically referred to. In this section, waste performance is measured by the assessment criteria for repository size, radiotoxicity lifetime, 1000 year radiotoxicity and final transuranic inventory. Fuel cycles with long reprocessing timescales and no curium reprocessing are described as high TRL fuel cycles. Fuel cycles with short cooling times or curium reprocessing are described as low TRL fuel cycles.

BURNER scenarios BURNER scenarios with americium included in the fuel (BURNER-AM) have a net reduction of americium. As such, there is a small reduction in repository size 1.06 to 1.10 (1.21 to 1.22 with LWR offset), which is considered significant when the offset of LWRs is included. Due to a relatively small reduction in plutonium, once-through BURNER scenarios have a negligible impact on radiotoxicity lifetime and do not have a significant impact on 1000 year radiotoxicity, 1.11 to 1.16 (1.23 to 1.25 with LWR offset).

FEED scenarios FEED-AM and FEED-TRU scenarios have significant improvements in waste performance over BURNER scenarios, however, they have three main disadvantages: development of a closed fuel cycle; reactors to be operated to 2200 rather than 2150; and 2.75 to 5.37 more SFRs compared to previous FEED scenarios

not using LWR SNF. Improvements in FEED scenario waste performance is a result of a significant reduction in plutonium and americium in the final inventory. FEED-HI and FEED-LO scenarios both have waste performance improvements over BURNER scenarios. FEED scenario improvements over direct disposal are significant in terms of repository size, 1.83 to 3.14 (2.71 to 4.76 with LWR offset), and 1000 year radiotoxicity 2.44 to 4.37 (3.24 to 5.91 with LWR offset). Unlike BURNER scenarios, FEED scenarios have significant repository reductions over the direct disposal scenario without including the offset of LWRs. The inclusion of curium in FEED-TRU scenarios has a negligible improvement in waste performance over FEED-AM scenarios, as there is only a small build-up of curium over the relatively short timescale of the FEED scenarios. As such, FEED-TRU scenarios should not be selected over FEED-AM scenarios when considering waste performance and TRL alone.

FULL and WASTE scenarios FULL scenarios are operated until 2500–2700, which may be unrealistic, but have significant improvements in waste performance. There are a range of results between the best performing FULL scenario, FULL-Zr-LO-TRU, and worst performing FULL scenario, FULL-MOX-HI-AM. FULL scenarios have an improvement on final stockpile mass of 4.05 to 9.57, improvement on radiotoxicity lifetime of 8.06 to 20.71, improvement of 1000 year radiotoxicity of 3.25 to 11.36 (6.05 to 28.13 with LWR offset), improvement in repository size of 2.14 to 7.30 (6.23 to 25.75 with LWR offset), and a negligible improvement in material attractiveness.

WASTE-TRU improvements are approximately 2-3 times higher than the equivalent FULL-TRU improvement factors, and WASTE-AM improvements are approximately 1.5-2 times higher than the equivalent FULL-AM improvement factors.

PU scenarios PU scenarios have some improvements over direct disposal scenarios. When considering FEED-PU scenarios with the offset of LWRs, there is a significant improvement in repository size and 1000 year radiotoxicity. However, these are not significantly better than BURNER-AM scenarios. Any improvements a FEED-PU scenario has over the direct disposal scenario are lost when extended to the timescale of FULL-PU scenarios. With more reactors operating in FULL-PU scenarios, more americium is generated and sent to the waste stream at reprocessing, leading to large repository sizes and 1000 year radiotoxicities when compared to FEED-PU scenarios.

Scenarios with and without new build Comparing fuel cycle scenarios from this chapter (plutonium stockpile and new build SNF as the feed) and previous chapters (Ch. 7 & Ch. 8, plutonium stockpile as the feed), there are some scenarios that are favourable in this chapter that were not in previous chapters. This implies that preferred options for the UK's fuel cycle could vary depending on the future situation of the UK's nuclear industry. In the present chapter, the use of new build reactor SNF as the primary fuel feed leads to minimal americium in-growth. With minimal americium in-growth, improvements in waste performance are much higher when using the new build SNF fuel feed compared to the plutonium stockpile feed. As such, some fuel cycle scenarios are preferable when new build SNF is included, that were not when the plutonium stockpile was used as the only fuel feed. These are summarised in Table 9.7, showing that with new build SNF included, less restrictive fuel cycle parameters are needed to get significant improvements in waste performance.

9.4.1 Comparison to literature

Comparing results to the literature is difficult as all fuel cycles are unique. This chapter is particularly different to previous studies as there are two fuel feeds, one aged plutonium feed and one fresh LWR feed. Previous closed fuel cycle studies of relevance more closely resemble fuel cycle scenarios in Chapter 8, as such they are not discussed here.

Comparing results to the literature of previous UK scenarios is difficult as the goals of the fuel cycles are different. In the UK R&D roadmap [21] 75 GWe of reactors are built, transitioning to SFRs by 2100. Results presented by NNL [127] had repository size improvements of 1.8 over a once-through LWR fuel cycles, when plutonium was reprocessed, and 2.5 when americium was reprocessed as well. Results in this chapter are not directly comparable to the UK R&D roadmap, due to the different scale of reactor deployment and method of calculating repository size. However, the most applicable results have been included for comparison. In this chapter, with plutonium recycling only, the reduction in repository size is 1.23 to 1.57 by 2200, with a peak generating capacity of 20 to 23 GWe. In this chapter, with all TRU reprocessed the reduction in repository size is 2.72 to 4.54, with a peak generating capacity of 22 to 26 GWe. The plutonium reprocessing results for repository size at 2200 are similar to

Table 9.7: Key factors that influence waste performance of fuel cycles. Comparing plutonium stockpile disposition scenarios and to plutonium and new build SNF disposition scenarios.

	Stockpile only Ch. 7 & Ch. 8	**Stockpile and new build SNF**
BURN	2100	2150
	Negligible improvements.	Significant improvements when the offset of LWRs are considered.
FEED	2150	2200
	Improvements require short cooling times and americium recycling.	Improvements require americium recycling without the need for short cooling times.
	Recycling of curium has minimal impact.	Recycling of curium has minimal impact.
	Only recycling plutonium has a negative impact.	Only recycling plutonium has a positive impact when LWR offset is considered.
FULL	2350–2460	2500–2700
	Americium must be recycled.	Americium must be recycled.
	Only recycling plutonium leads to negative impact.	Only recycling plutonium leads to negative impact.
	Short cooling and/or curium recycling is required to gain an improvement in repository size	Long cooling and no curium recycling (just plutonium and americium) can result in positive improvements in repository size.

those presented in the UK R&D roadmap.

From literature based on Eurobarometer polls, the three main factors that influence public perception of nuclear power are: terrorism concerns, waste disposal and proliferation, in that order [4]. The most important factor, terrorism, cannot be prevented by fuel cycle scenarios, but waste disposal and proliferation can. Separated plutonium, such as the UK's plutonium stockpile and separated material from the reprocessing of SNF, is a large proliferation concern. Tackling proliferation concerns requires the same approach as discussed in Section 8.4.1. In terms of public concern about waste, the most effective way to reduce waste is in FULL scenarios where the lifetime of radiotoxicity can be reduced by a factor of 10.71, repository size by a factor of 7.30 and 1000 year radiotoxicity by a factor of 11.36. These improvements are

Table 9.8: For scenarios using the UK's plutonium stockpile and new build SNF as a fuel feed, BURNER fuel cycle scenario improvement factors are presented relative to a [reference case] for each assessment criteria. Improvement factors greater than one show a positive improvement. Improvement factors less than one show a dis-improvement.

		Zr Lo-PU	Zr Lo-AM	MOX Lo-PU	MOX Lo-AM
TRL		6	4	9	5
GWy(e) [PWR MOX]		0.81	0.80	0.52	0.60
Mass [Stock]	TRU	1.15	1.15	1.29	1.23
CDH [Stock]	50y	0.97	1.06	1.05	1.10
CDH [Stock+LWR]	50y	1.11	1.21	1.14	1.22
Sv_{1000} [Stock]	50y	1.01	1.11	1.11	1.16
Sv_{1000} [Stock+LWR]	50y	1.12	1.23	1.19	1.25
Sv Lifetime [PWR UOX]		1.71	0.93	1.81	0.97
Critical Mass [Stock]		1.11	1.05	1.12	1.08

very significant and may improve public perception of nuclear waste. However, FULL scenarios require a closed fuel cycle operated until 2600–2700 which may have a negative impact in terms of public perception. In a reasonable timescale, such as 2200 in the FEED scenarios, radiotoxicity lifetime can be reduced by a factor of 5.58, repository size by a factor of 3.14 and 1000 radiotoxicity at 1000 years by 4.37. Whilst these are large improvements they may not be large enough to address public concerns. In addition, these are waste performance improvements for best case scenarios using low TRL fuel cycles. The cost of developing technology to get these improvements in waste performance may have a negative impact on public perception. Alternatively, higher TRL fuel cycles could be developed higher TRL fuel cycle scenarios have poorer waste performance, which may not address public concerns as discussed in Section 8.4.1.

9.4.2 Recommendations and further work

Depending on the priorities of decision makers, different fuel cycle scenarios would be preferable in the UK. In the present authors' opinion, the use of FULL scenarios is unlikely due to the long commitment to a closed fast reactor fuel cycles. It is unlikely that any UK drivers would be large enough to commit to a closed fuel cycle for 450 years as current UK nuclear policy is only determined to 2050, 25 years. If fast

Table 9.9: For scenarios using the UK's plutonium stockpile and new build SNF as a fuel feed, FEED, FULL and WASTE fuel cycle scenario improvement factors are presented relative to a [reference case] for each assessment criteria. Improvement factors greater than one show a positive improvement. Improvement factors less than one show a dis-improvement.

		Zr Lo-PU	Zr Lo-AM	Zr Lo-TRU	MOX Lo-PU	MOX Lo-AM	MOX Lo-TRU	Zr Hi-PU	Zr Hi-AM	Zr Hi-TRU	MOX Hi-PU	MOX Hi-AM	MOX Hi-TRU
TRL		6	4	3	7	5	3	6	4	3	8	5	3
FEED Scenario													
GWy(e) [PWR MOX]		2.40	3.05	3.22	1.65	2.25	2.41	1.79	2.25	2.25	1.24	1.65	1.71
Mass [Stock]	TRU	2.95	3.34	3.54	3.41	4.09	4.57	2.34	2.71	2.72	2.74	3.27	3.42
CDH [Stock]	50y	1.03	2.38	2.43	1.16	3.07	3.14	0.88	1.84	1.83	1.02	2.14	2.17
CDH [Stock+LWR]	50y	1.56	3.93	4.09	1.57	4.54	4.76	1.23	2.72	2.71	1.29	2.89	2.97
Sv_{1000} [Stock]	50y	1.68	3.04	3.31	1.92	3.85	4.37	1.38	2.44	2.47	1.60	2.92	3.06
Sv_{1000} [Stock+LWR]	50y	2.27	4.40	4.87	2.39	5.13	5.91	1.74	3.24	3.29	1.89	3.62	3.82
Sv Lifetime [PWR UOX]		4.20	4.75	4.68	4.39	5.27	5.58	3.45	3.58	3.45	3.63	3.92	3.97
Critical Mass [Stock]		1.05	1.04	1.03	1.05	1.05	1.05	1.03	1.05	1.05	1.03	1.08	1.08
FULL Scenario													
GWy(e) [PWR MOX]		5.86	8.18	8.73	4.86	7.07	7.07	4.53	6.41	6.63	3.65	5.09	4.75
Mass [Stock]	TRU	2.35	4.49	9.57	2.17	4.02	8.90	1.91	4.49	7.30	1.77	4.05	5.74
CDH [Stock]	50y	0.54	3.38	7.30	0.49	2.85	6.11	0.44	2.99	4.56	0.38	2.14	2.62
CDH [Stock+LWR]	50y	1.47	11.40	25.75	1.18	8.70	18.62	1.01	8.54	13.33	0.79	5.29	6.23
Sv_{1000} [Stock]	50y	1.09	3.46	11.36	0.99	3.00	9.68	0.89	3.78	7.12	0.78	3.25	4.68
Sv_{1000} [Stock+LWR]	50y	2.17	8.23	28.13	1.80	6.58	21.26	1.58	7.88	15.10	1.26	6.05	8.44
Sv Lifetime [PWR UOX]		4.60	9.67	20.71	4.08	8.79	17.06	4.39	9.06	13.18	3.92	8.06	9.67
Critical Mass [Stock]		1.20	1.15	1.12	1.29	1.18	1.16	1.29	1.19	1.17	1.32	1.25	1.25
WASTE Stream Only													
GWy(e) [PWR MOX]		2.52	5.35	13.63	2.36	4.91	12.88	2.22	6.32	11.39	2.14	5.92	10.84
Mass [Stock]	TRU	0.56	4.97	19.84	0.51	4.40	16.58	0.46	6.27	11.97	0.40	5.00	9.40
CDH [Stock]	50y	1.51	16.75	70.04	1.22	13.43	50.54	1.06	17.91	34.96	0.83	12.36	22.34
CDH [Stock+LWR]	50y	1.14	4.26	24.95	1.04	3.80	20.83	0.98	6.14	15.62	0.88	5.67	13.32
Sv_{1000} [Stock]	50y	2.27	10.15	61.79	1.90	8.33	45.74	1.74	12.79	33.12	1.41	10.54	24.02
Sv_{1000} [Stock+LWR]	50y	4.92	11.15	59.18	4.33	10.00	35.37	5.18	12.08	26.36	4.68	10.74	20.71

reactor development is a priority for the UK whilst minimising development costs, then starting a once-through BURNER-AM scenario would be preferable. When the offset of LWRs is considered, repository size is reduced for BURNER-AM scenarios, having a positive affect on waste performance without committing to the development of SFR fuel reprocessing. BURNER-AM scenarios would work well as the start of an SFR R&D programme, with the potential to extended the scenario to include reprocessing and more fast reactors if desired (FEED-AM or FEED-TRU scenarios). Extending BURNER scenarios to FEED scenarios would have a more significant impact on waste performance. If natural uranium usage is not a concern, but repository size and radiotoxicity is a priority, the FEED-MOX-AM scenarios would be preferable to reduce the burden LWRs have on a repository. The use of FEED-MOX-AM is preferable over FEED-MOX-TRU due to higher TRL of the fuel cycle and the minimal impact of curium recycling on FEED waste performance. FEED-MOX-AM is preferable over FEED-ZR-AM due to better improvements in waste performance, as well as there being more experience using MOX fuel. If proliferation is a priority, as well as other factors, then FEED-MOX-TRU scenarios would be preferable. If reducing dependence on natural uranium is a priority then transitioning to SFR scenarios in the UK R&D roadmap [21] would be preferable as discussed in Section 8.4.1.

Further work should look at UK fuel cycle studies, related to the UK R&D roadmap [21], comparing the 75 GWe SFR closed fuel cycle scenarioin the road map to 75 GWe of combined LWRs and low-CR SFRs. Low-CR SFRs would be used to reduce the waste generated from LWRs with an aim to reducing the burden on a repository without fully transitioning to SFRs.

9.5 Conclusions

This chapter modelled once-through and closed SFR fuel cycles using two fuel feeds, the UK's plutonium stockpile and reprocessed SNF from 16.5 GWe of new build LWRs. Three scenarios were considered which aimed to improve waste performance: once-through BURNERS operated to 2150; closed fuel cycles irradiating all material by 2200 (FEED scenarios), and a closed fuel cycle running for as long as possible to maximise reduction of the stockpile (FULL scenarios). Waste performance is measured by the

assessment criteria for repository size, radiotoxicity lifetime, 1000 year radiotoxicity and final transuranic inventory. Fuel cycles with long reprocessing timescales and no curium reprocessing are described as high TRL fuel cycles. Fuel cycles with short cooling times or curium reprocessing are described as low TRL fuel cycles.

Once-through BURNER scenarios with americium included in the fuel (BURNER-AM) reduce repository size significantly, by a factor of 1.21 – 1.22, when the offset of once-through LWRs is considered. FEED scenarios that include americium recycling (FEED-AM and FEED-TRU) perform significantly better than BURNER scenarios, with improvement factors greater than 2. FULL scenarios can achieve an order of magnitude improvement in waste performance. FEED-PU scenarios have small waste performance improvements, a factor of 1.29 – 2.39, when the offset of once-through LWRs is considered, whereas FULL-PU scenarios have negligible improvements in waste performance considering that 350 years of reactor operation is required. When the plutonium stockpile and new build SNF are used as SFR fuel feeds, more fuel cycle scenarios have significant improvements in waste performance when compared to the equivalent scenarios in Chapter 7 and 8 that only use the UK's plutonium stockpile as a fuel feed.

Once-through BURNER-AM scenarios have a significant impact on repository size, by a factor of 1.21 – 1.22, when the offset of LWRs is considered. This result is significant because it shows that improvements in repository size are possible without the need for SFR reprocessing. Therefore, BURNER-AM scenarios would work well as the start of an SFR R&D programme, to develop SFRs whilst having a positive impact on the fuel cycle. Development of reprocessing and FEED scenarios would be preferable over BURNER scenarios if a reduction in repository size is considered to be important, with improvement factors of 1.84 – 3.14 without considering LWR offset and 2.72 – 4.76 with LWR offset. FEED-TRU scenarios should not be developed over FEED-AM scenarios as FEED-TRU scenarios have similar improvements in waste performance and a lower TRL. However, if the proliferation resistance of reprocessed fuel is a concern, then FEED-TRU scenarios may be preferable over FEED-AM scenarios to increase the intrinsic proliferation resistance of the fuel cycle.

The largest improvement factors, of an order of magnitude, are due to FULL scenarios, however these involve the operation of closed SFR fuel cycles for more than

450 years, which may be unrealistic.

FEED-PU scenario waste performance is not substantially better than BURNER-AM scenarios which do not require a closed fuel cycle. As a result, if waste performance is a primary concern, there is no real benefit to operating a FEED-PU scenarios over BURNER-AM scenarios. If reprocessing is developed to improve waste performance then americium must be reprocessed.

Fuel cycle scenarios that considered the plutonium stockpile with new build SNF as a fuel feed were compared to scenarios that just consider the plutonium stockpile, Table 9.7. This shows how preferable fuel cycle options vary depending on how a UK fuel cycle develops. More fuel cycle options have significant improvements in waste performance when new build SNF is included. The inclusion of new build SNF as a fuel feed has three main advantages when developing a fuel cycle: BURNER scenarios can reduce repository size when LWR offset is considered; FEED scenarios can improve waste performance with long reprocessing times (FEED-HI), and FULL scenarios can reduce repository size with long reprocessing times and americium recycling (FULL-HI-AM). This is not the case for scenarios in Chapter 7 and 8, where only the plutonium stockpile is used as the only fuel feed. Extending FEED scenarios that use the UK's plutonium stockpile to include new build SNF as a fuel feed, allows higher TRL fuel cycles to be used while still improving waste performance. However, extending FEED scenarios to include new build SNF requires two to three times more SFRs, and the associated fabrication and reprocessing throughput at facilities. Benefits of extending BURNER scenarios to include new build SNF may not be worth the large scale reprocessing of LWR SNF.

The key limitations of the results presented in this chapter are the approximation methods used for repository size and radiotoxicity lifetime, as well as the lack of quantitative assessment of proliferation, development needs and cost. These have been summarised previously, see Section 7.5. Specific limitations of this chapter are the fixed lifetime of the LWR reactors and focusing on the impact of SFRs on waste from a single generation of LWRs. Further work should investigate the steady state operation of LWRs in equilibrium with SFRs.

Overall, if reducing the impact LWRs have on a repository is a priority, then FEED-AM scenarios are preferable. However, if reducing dependence on natural uranium is

a priority, then transitioning to an SFR only fuel cycle is preferable, as discussed in the UK's R&D roadmap [21]. The main recommendation for the continuation of this study is to model 16 to 75 GWe scenarios, similar to the UK R&D closed fuel cycle pathway, but operating a combination of LWRs and low-CR SFRs in equilibrium, to reduce the waste generated from LWRs. This would be a useful to compare to the UK R&D closed fuel cycle pathway, to compare the impact on uranium demand and repository size.

Key findings:

- BURNER-AM scenarios can reduce the size of the stockpile when LWR offset is considered, without the need for SFR reprocessing.
- FEED scenarios waste performance is improved over BURNER scenarios if americium is reprocessed.
- FULL scenarios have large improvements in waste performance but require the operation of a closed fuel cycle until at least 2500, which may be unrealistic.
- Extending FEED scenarios from previous chapters to include new build SNF allows significant improvement in waste performance to be achieved whilst using higher TRL fuel cycles.

Chapter 10

Decision analysis

10.1 Introduction

Improvement factors tabulated for the assessment criteria of different fuel cycle scenarios have been calculated in Tables 7.4, 8.5, 9.8 and 9.9 at the end of Chapters 7, 8 and 9. Two potential situations for the UK nuclear industry have been described in this thesis: disposition of the UK's plutonium stockpile, and disposition of both the plutonium stockpile and SNF from new build LWRs.

Recommendations for preferred fuel cycle scenarios have been made in previous chapters based on hypothesised fuel cycle goals and factors which affect public perception of nuclear power. However, different people will envisage different goals for the UK's nuclear programme and attribute a different level of importance to the improvement factors that were determined in this study.

To resolve this problem an example of decision analysis methods were used to show how the improvement factors calculated in this study could be used to determine preferred fuel cycle scenarios. In the example, improvement factors were normalised and weighted based on the present author's interpretation of fuel cycle priorities: TRL, stockpile mass reduction, electricity generation and repository size. Results were presented along with the methodology so that the reader can perform their own assessment. The spreadsheet for decision analysis is available from the present author upon request.

10.2 Methods

All assessment criteria results from fuel cycle analysis in Chapter 7, 8 and 9 were condensed in the discussion sections into individual improvement factors for each assessment criteria in Table 7.4, 8.5, 9.8 and 9.9 . The improvement factors represent specific, measurable improvements when compared to a reference case. Improvement factors can be normalised across fuel cycle scenarios using,

$$x' = \frac{x - x_{min}}{x_{max} - x_{min}}. \tag{10.1}$$

Where x' is the normalised improvement factor for a scenario, x is the original improvement factor for a scenario, x_{min} is the minimum improvement factor across all considered fuel cycle scenarios, and x_{max} is the maximum improvement factor across all considered fuel cycle scenarios.

Results for four key situations of the UK nuclear industry can be used as a basis for comparison.

- Scenarios considering the disposition of the UK's plutonium stockpile:
  - Where SFRs are used to reduce stockpiled material – This does not consider that SFRs are being used as an alternative to LWRs. Instead the SFRs are used to reduce the plutonium stockpile with the electricity generated considered as a by-product.
  - Where SFRs are used to reduce stockpiled material and contribute to the total nuclear generating capacity, offsetting LWRs – This assumes that any SFRs operated are an alternative to once-through LWRs, offsetting SNF generated from LWRs.
- Scenarios considering the disposition of the UK's plutonium stockpile and SNF from 16.5 GWe of new build LWRs:
  - Where SFRs are used solely to reduce stockpiled material
  - Where SFRs are used to reduce stockpiled material and contribute to the total nuclear generating capacity, offsetting additional LWRs

Each of these situations have different drivers and represent a different policy for the future of the UK's nuclear fuel cycle. As such, they should be treated separately, not comparatively.

The improvement factors for a scenario were normalised between 0 and 1, so that all assessment criteria were equally weighted. For each assessment criteria a WEIGHTING factor was applied, based on the importance given to that criteria.

Additional SCALING factors were also used to capture the development needs or personal preferences for a fuel cycles scenario. SCALING should be based on three factors. First the user's interpretation of the TRL and development needs as TRL is a very broad definition. Second, the importance of a specific scenario to the goals of the fuel cycle. Finally, the user's interpretation of other qualitative factors such as proliferation resistance and cost estimations. Ten SCALING factors were added for SFR fuel cycle scenarios, these were applied to BURNER, FEED, FULL, HIGH, LOW, ZR, MOX, PU, AM, and TRU scenarios. In the case of no SCALING, these factors would be set to 1.

Previous chapters discuss the relative merits of each fuel cycle scenario which should be used as a guide to setting the WEIGHTING and SCALING factors. The following section gives examples of WEIGHTING and SCALING factors to show how the decision analysis methodology can be applied.

10.3 Example results

Examples of the decision analysis method are presented in this section. Three examples were used to compare three UK plutonium disposition methods outlined by the NDA: once-through SFRs, PWR MOX and direct disposal. These three examples are presented in Section 10.3.1. A more complex fourth example case was used to show how the four SFR fuel cycle situations described in the methods have different preferred fuel cycle scenarios given the same WEIGHTING and SCALING factors. The fourth example is presented in Section 10.3.2.

SCALING and WEIGHTINGS used for all examples are shown in Table 10.1 and results of the decision analysis are shown in Figure 10.1 and 10.2.

10.3.1 Example 1 – 3

Three examples were used to compare three UK plutonium disposition methods outlined by the NDA: once-through SFRs, PWR MOX and direct disposal. SCALING and WEIGHTINGS used are shown in Table 10.1 and results of the decision analysis are shown in Figure 10.1.

SCALING factors were used to express that the UK has more experience fabricating MOX fuel, and that direct disposal is the least industrially intensive process. WEIGHTINGS for each of the assessment criteria were applied based on the following key fuel cycle priorities:

- Example 1: Prioritise plutonium stockpile inventory reduction and repository size equally – Resulting in SFR BURNERS being preferable, Figure 10.1. SFR BURNERS reduce the plutonium stockpile with the smallest impact on repository size.

- Example 2: Prioritise plutonium stockpile inventory reduction, repository size and TRL equally – Resulting in direct disposal of the plutonium stockpile being preferable. Although the plutonium stockpile is not reduced in direct disposal scenario, it has smallest development barriers and the smallest repository.

- Example 3: Prioritise plutonium reduction, repository size, electricity generation and TRL equally – Resulting in PWR MOX being preferable. Despite having the largest repository, PWR MOX has a high TRL and reduces the plutonium stockpile more significantly than the other scenarios.

These three examples show how the decision analysis method works for a simple case, relevant to the current options for UK plutonium disposition. It is worth noting that cost is not covered as part of this assessment, despite being a significant priority. To some extent TRL will represent cost, with more ready technology costing less, but TRL and cost will not be a linear relation.

10.3.2 Example 4

A more complex fourth example case was used to show how the four SFR fuel cycle situations described in the methods have different preferred fuel cycle scenarios given

Table 10.1: Weightings and scaling factors applied to four decision analysis examples.

	Examples			
	1	**2**	**3**	**4**
WEIGHTING				
FC Length	-	-	-	1.0
GWYe	-	-	1.0	1.0
TRU	1.0	1.0	1.0	0.1
Critical Mass	-	-	-	0.1
Sv lifetime	-	-	-	0.5
Sv_{1000}	-	-	-	0.25
Repository size	1.0	1.0	1.0	1.0
TRL	-	1.0	1.0	0.25
SCALING				
FULL	-	-	-	0.8
FEED	-	-	-	1.1
BURNER	1.0	1.0	1.0	1.2
PWR MOX	1.0	1.0	1.0	-
Disposal	1.1	1.1	1.1	-
ZR	1.0	1.0	1.0	1.0
MOX	1.1	1.1	1.1	1.1
PU	1.0	1.0	1.0	1.0
AM	1.0	1.0	1.0	1.2
TRU	-	-	-	0.9
LOW	-	-	-	1.0
HIGH	-	-	-	1.1

the same WEIGHTING and SCALING factors.

In the fourth example, all assessment criteria are important but repository size, electricity generation and fuel cycle length are prioritised in the weightings. Americium fuels and near-term BURNER scenarios were scaled to prioritise high TRL transmutation scenarios. The aim of the scaling and weighting, shown in Table 10.1 was to compare short-term, more feasible SFR scenarios to repository reduction scenarios.

The results were plotted for all four UK situations in Figure 10.2. Figure 10.2 shows that the preferable scenario for a set of scaling and weighting factors varies based on the UK's situation.

Situation 1 in Figure 10.2, considering disposition of the plutonium stockpile only, the BURNER-MOX-AM scenario is preferable. Situation 2, considering disposition of the plutonium stockpile and offset of once-through LWRs, the FULL-LO-ZR-AM

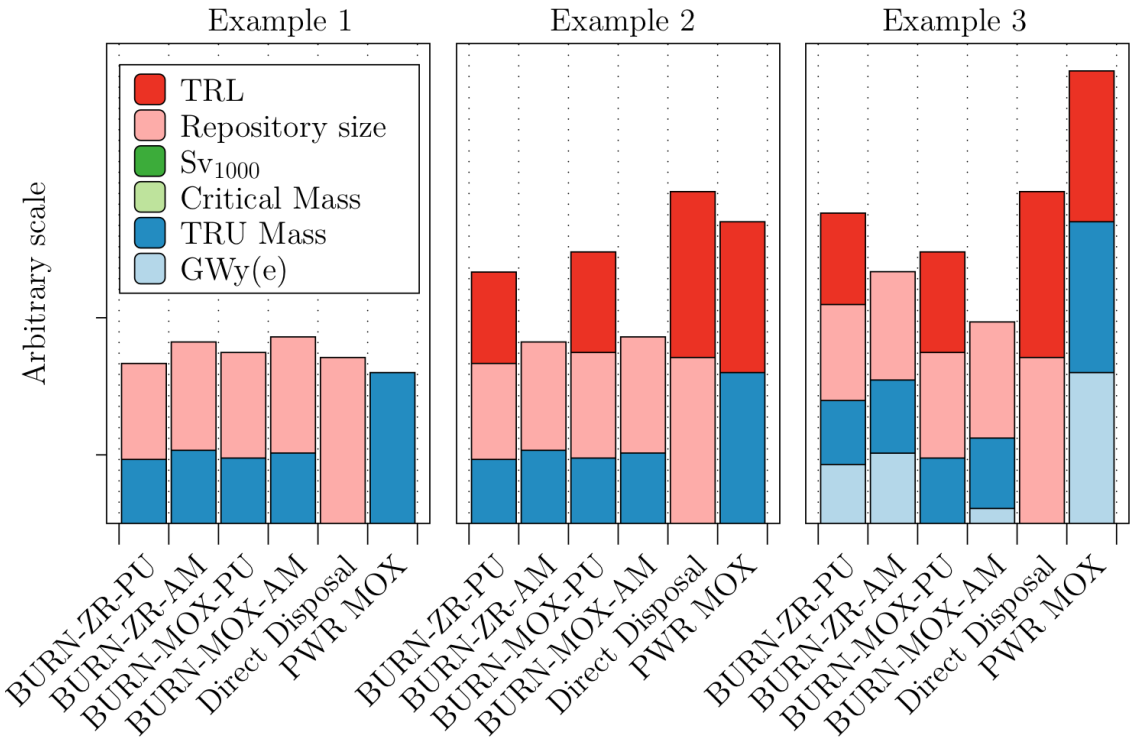


Figure 10.1: Example 1–3 of decision analysis, outlined in Table 10.1, used to compare once-through UK plutonium disposition options: BURNER SFRs (similar to PRISM), PWR MOX and direct disposal.

scenario is preferable. Situation 3 and 4, considering disposition of the plutonium stockpile and new build SNF (with and without once-through LWR offset) the FEED-LO-MOX-AM scenario is preferable. As well as the difference in most favourable scenario, the order of results clearly changes for each situation.

10.4 Discussion

This chapter aims to provide a method of using improvement factors for decision making. Methods for normalisation, application of scaling and weightings are dependent on policy or personal priorities. The methods and results provided in this study can be used as a decision making tool to determine preferred reactors and fuel cycles for further study.

Improvement factors are tabulated in Table 7.4, 8.5, 9.8 and 9.9 at the end of Chapters 7, 8 and 9 and available for the reader to use in decision analysis. Methods used for assessment criteria have been provided in this chapter so that the reader can assess the performance of other fuel cycle scenarios by the same metrics and

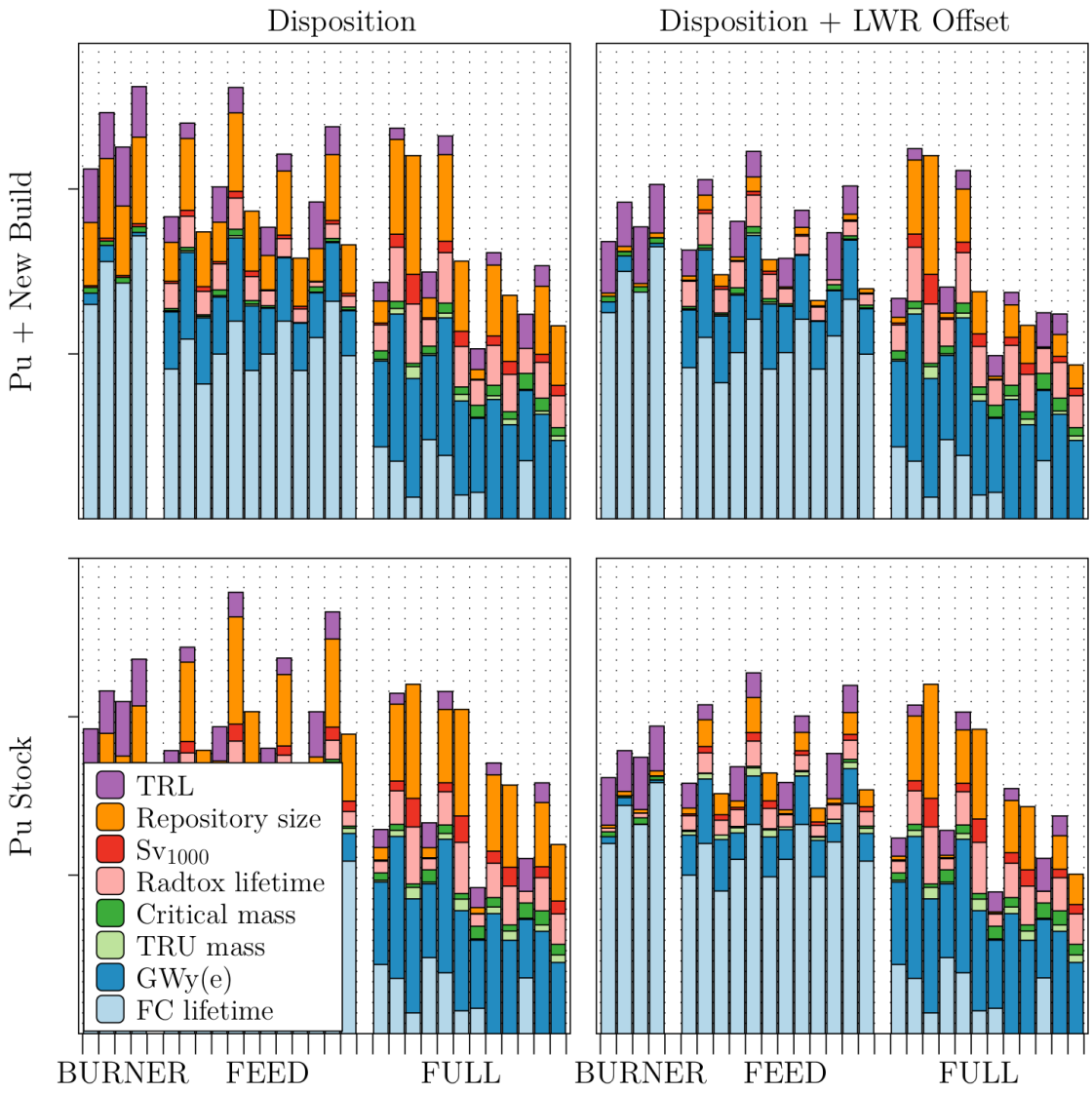


Figure 10.2: Example 4 of decision analysis results, using the same SCALING and WEIGHTING factors for four UK nuclear situations: disposition of the plutonium stockpile (upper left); disposition of the plutonium stockpile with the offset of once-though LWRs (lower left); disposition of the plutonium stockpile and new build SNF (upper right); disposition of the plutonium stockpile and new build with offset of once-through LWRs (lower right). BURNER, FEED and FULL scenarios are grouped but not labelled specifically.

compare them directly to the results in this thesis. Detail of fuel cycle models have been provided so that models can be repeated and assessed with alternative criteria to include in decision analysis.

The best case result of decision analysis are not necessary the most preferable. User scaling and weightings are used to give an idea of what is important. User scaling and weightings should not be seen as an absolute, quantitative value for the importance of different factors.

Example results presented in this chapter show that preferred fuel cycle options are sensitive to weightings. Examples also showed that the preferred fuel cycle options are sensitive to the situation of the nuclear industry. If new build does not go ahead, the same weighting and scaling factors result in very different preferred fuel cycle options. Equally, if SFRs are used to offset the number once-through LWRs constructed, preferred options will be also be different.

10.4.1 Limitations and further work

The main limitations of this work is that the weightings and scaling are entirely dependent on the user. Factors can be adjusted to get the results a user wants. However, the same weightings and priorities lead to different favourable scenarios when different UK nuclear situations are considered.

Other limitations apply to the normalisation of results; in particular the use of TRL as a linear scale. Further work should apply more detailed analysis to the development needs of each scenario, replacing the TRL scale with a more finely binned scale.

There are no high-CR fuel cycle results for comparison. As such, there is the potential for scenarios to look preferable, compared to other low-CR fuel cycle scenarios, but may not be when compared to high-CR fuel cycle scenarios. For example, if electricity generation and TRL are the highest priorities, a high-CR plutonium recycling scenario could be preferable over low-CR scenarios. Further work to include high-CR scenarios, similar to those in the UK's nuclear R&D pathways [3], would be useful if electricity generation or natural uranium demand are priorities.

Other limitations include additional assessment criteria which were not evaluated in this thesis. For example, cost estimates or a way to quantitatively assess the proliferation resistance of a fuel cycle scenario.

10.5 Conclusions

This chapter provided a methodology for using the results of this thesis in decision analysis. Examples of decision analysis were used to compare three UK plutonium disposition methods outlined by the NDA, once-through SFRs, PWR MOX and direct disposal. Decision analysis was used to show how the best disposition option is dependent on the UK's fuel cycle priorities. Decision analysis was also used to show that preferred closed fuel cycle options are sensitive to the situation of the nuclear industry.

Results and methods presented in this thesis are useful tools for evaluating fuel cycles with decision analysis methods. Further work could add additional assessment criteria or fuel cycle scenarios to the results presented in this thesis for further comparison of fuel cycle options.

Key findings:

- Preferred fuel cycle scenarios are dependant on priorities and the weightings given to different assessment criteria.
- As well as assessment criteria priorities, the UK fuel cycle situation influences preferred fuel cycle scenarios.
- Decision analysis is a useful tool for scoping fuel cycle scenarios. However, it is subjective and should only be used as a guide to show the potential benefits of different fuel cycle scenarios.

Chapter 11

Summary and conclusions

This work considered fast reactor fuel cycle scenarios and their potential to reduce the UK's nuclear waste inventory. Fuel cycle scenarios for waste disposition were selected, based on UK plutonium disposition options currently being considered by the NDA and the construction of 16 GWe of new build LWRs. Fuel cycle scenarios were modelled and their performance compared based on assessment criteria defined in this work. Waste performance was described using the waste inventory, repository size and radiotoxicity assessment criteria. Fuel cycles with long reprocessing timescales and no curium recycling were described as high TRL fuel cycles. Fuel cycles with short cooling times were described as low TRL fuel cycles, as were fuel cycles with curium recycling. The main contributions to knowledge from this work were the following.

- This work showed preferred options for once-through UK plutonium disposition, given different potential priorities for a UK fuel cycle.
- In this work closed SFR fuel cycle scenarios for UK plutonium disposition were modelled. Best case scenarios were determined based on defined assessment criteria and different priorities for a UK fuel cycle.
- It was shown that the stockpile reduction achieved by high TRL, closed SFR fuel cycle concepts performed poorly in terms of repository size. A significant reduction of the UK plutonium stockpile requires low TRL fuel cycles to perform well in terms of waste performance.
- It was shown that extending fuel cycle scenarios that used the UK's plutonium stockpile to include new build LWR SNF resulted in better waste performance,

with higher TRL closed SFR fuel cycles still showing good waste performance.

- It was shown that once-through SFR BURNERS of the UK's plutonium stockpile and SNF from new build LWRs can reduce repository size without the need for a closed SFR fuel cycle.

- It was shown that over timescales of 100 to 150 years, closed SFR fuel cycles, in a UK context, were not influenced significantly by the reprocessing of curium.

- It was shown that improvement factors of greater than an order of magnitude were reached when assessing the waste stream only (waste and losses from reprocessing) but these high improvement factors were not achievable when realistic fuel cycles were operated for more than 300 years.

11.1 Overview of research method

Initial TRL assessment of fast reactor and fuel cycle technology was used to give an overview of potential fuel cycle options. Gaps in previous research were assessed, both in terms of general fuel cycle studies and UK specific fuel cycle studies. UK waste disposition fuel cycle scenarios were down selected and outlined for modelling. The UK's plutonium stockpile and SNF from the UK new build programme were estimated and used as fuel feeds. SFRs were designed using ERANOS to meet the requirements of fuel cycle scenarios. Fuel cycle scenarios were modelled with ORION using the UK's plutonium stockpile as a fuel feed and reactors designed in ERANOS.

ORION fuel cycle scenarios modelled once-through fuel cycles for UK plutonium disposition and closed SFR fuel cycles, testing different fuel cycle and reactor parameters. Fuel cycle scenarios were extended to include new build LWR SNF as an additional fuel feed to see how waste performance would change. Fuel cycles were assessed using a set of assessment criteria, these were: power generated; fuel cycle lifetime; final inventory; radiotoxicity lifetime; radiotoxicity at 1000 years; repository size and bare sphere critical mass of final material. Decision analysis methods were presented to show how best case scenarios are dependent on the different priorities for a UK fuel cycle.

11.2 Main findings

The UK's plutonium stockpile was estimated and compared to published information on the total mass of the UK's plutonium stockpile, approximately 112.6 tonnes at the end of reprocessing. The plutonium stockpile results were representative (within 10% of published data) and sufficient for use as a reference case to compare fuel cycle models. SNF from UK new build was estimated based on the UK's current 16 GWe goal by 2050, resulting in a stockpile of 207.3 or 223.6 tonnes depending on whether americium was reprocessed. New build was approximated as a single type of PWR for simplification, as there are large uncertainties in the UK's new build plan based on: reactor type, burnup, lifetime, capacity factor and final generating capacity.

The fast reactor neutronics code ERANOS was compared to an IAEA benchmark study to compare burnup inventory calculation results with other deterministic neutronics codes. Agreement between fast reactor neutronics code was good, with plutonium isotopic mass changes within 6% of the average. ERANOS was also compared to the SERPENT Monte-Carlo code, using the same JEF-2.2 cross-sections, for a reference SFR design. Agreement was good between ERANOS and SERPENT, with most isotopic plutonium mass changes within 10%, but there were discrepancies in ^{240}Pu burnup inventory. The reason for ^{240}Pu discrepancies was not determined, but the impact on results is expected to be small. The use of more recent ENDF/B7 or JEFF-3.1.1 cross-section libraries was compared to the JEF-2.2 library in SERPENT and had a significant impact on k_{eff} results, up to 1500 pcm, but a relatively small impact on fuel depletion results. As such, the use of ERANOS for reactor design with JEF-2.2 cross-sections was deemed valid.

Once-through SFR BURNER, PWR MOX and direct disposal scenarios were modelled for UK plutonium disposition and assessed at 2100. Direct disposal leads to a smaller repository size and lower 1000 year radiotoxicity than any once-through scenario. PWR MOX reduces plutonium stockpile mass by a factor of 1.30, more than SFR BURNER scenarios which reduce the stockpile mass by a maximum factor of 1.16. However, the PWR MOX scenario results in a large repository, twice the size of direct disposal, compared to SFR BURNER scenarios which have a maximum improvement factor of 0.83. The best case SFR BURNER scenario has a similar repository size

to that of the direct disposal scenario when the offset of once-through LWRs is considered. The best case SFR BURNER scenario requires a low-CR reactor design and the inclusion of americium in the fuel (BURNER-AM). There is little difference between results for equivalent SFR BURNERS using metallic fuel or MOX fuel in terms of the assessment criteria, therefore higher TRL MOX fuels are preferable. The choice between PWR MOX and a once-through BURNER-AM scenarios depends on the relative importance of three assessment criteria: TRL, the reduction in stockpile mass and repository size.

Closed SFR fuel cycles for plutonium disposition were tested with the aim of improving waste performance. Two types of closed SFR fuel cycles were considered to test reactor and fuel cycle parameters. FEED equilibrium scenarios modelled a small deployment of SFRs up to 2150 to determine any advantages in waste performance over once-through BURNERS. FULL equilibrium scenarios modelled several generations of SFRs, reducing the generating capacity as the transuranic fuel cycle inventory reduced. FULL equilibrium scenarios aimed to maximise stockpile reduction to determine the waste performance and timescale of the best case scenario.

FEED scenarios with short reprocessing times, and where americium was reprocessed, had better waste performance than BURNER scenarios. FEED scenarios have improvement factors as high as 2.02 and 2.54 for repository size and 1000 year radiotoxicity when LWR offset is considered. However, americium recycling and short reprocessing times are low TRL scenarios. Better developed, higher TRL FEED scenarios with longer reprocessing times, or just plutonium recycling, did not result in significantly better waste performance than BURNER scenarios. Recycling of curium in FEED scenarios did not have a significant impact on waste performance by 2150. Metallic and MOX fuelled reactors in FEED scenarios had similar waste performance, therefore MOX fuel is preferable as a result of greater fabricating and operating experience with MOX fuel.

FULL equilibrium scenarios had a large improvement on waste performance over direct disposal, close to an order of magnitude, but required 300 to 400 years of reactor operation, short reprocessing times and all transuranics to be recycled (FULL-LO-TRU) which have a low TRL. It is worth noting that 300 to 400 years of reactor operation may be unrealistic, but gives a best case scenario for waste performance.

FULL-LO-TRU scenarios reduced repository size, radiotoxicity lifetime and 1000 year radiotoxicity by about an order of magnitude, similar to results from the literature. It was assumed that order of magnitude improvements in waste performance factors would improve public perception of nuclear waste. However, FULL-LO-TRU scenarios have low TRLs. Higher TRL FULL scenarios, which have not been discussed in the literature, with longer reprocessing times or the recycling of americium without curium. Higher TRL FULL scenarios did not improve waste performance as significantly as lower TRL FULL scenarios, making 300 to 400 years of reactor operation unacceptable.

BURNER, FEED and FULL scenarios were extended to include reprocessed SNF from new build LWRs as a fuel feed as well as the plutonium stockpile. By extending scenarios to include new build SNF, several fuel cycle scenarios had good waste performance in this situation, which had poor waste performance when using the plutonium stockpile alone. Once-through BURNER-AM scenarios reduced the size of a repository, which was not possible when the plutonium stockpile was the only fuel feed. Extending FEED scenarios to include new build SNF resulted in good waste performance for higher TRL scenarios which have long reprocessing times. FULL scenarios that used new build LWR SNF required the operation of SFRs past 2500 which may not be realistic.

Decision analysis methods were presented so that the reader could use improvement factors, determined in this work, for decision analysis. Examples of UK once-though plutonium disposition options were provided to show how the relative importance of TRL, repository size and stockpile reduction leads to different disposition scenarios being preferable. As the weighting of different assessment criteria is user dependent, the methods were presented so the reader can use the results from this thesis to determine which SFR fuel cycles are preferable depending on the relative importance of different factors. A spreadsheet containing improvement factors and the decision analysis methods is available from the author on request.

11.3 Limitations

There were several limitations to the work presented in this thesis. In particular the modelling of the UK plutonium stockpile used an approximate average burnup without

accounting for the increase in average reactor burnup over time. As a result, AGR fuel burnup was underestimated and Magnox fuel burnup was overestimated. Another key limitation was the accuracy of cross-sections which were the limiting factor in the accuracy of neutronics modelling. Cross-sections determine the accuracy of depletion calculations used in the fuel cycle models. There are many approximations used in fuel cycle modelling such as the use of one set of cross-section to model in-core depletion over the lifetime of the fuel cycle. As such, the accuracy of cross-sections, whilst a limitation, is expected to be small relative to the inherent approximations used in fuel cycle modelling.

There are key limitations in the method used to estimate repository size. Design and modelling of a repository was outside of the scope of this PhD. A simplified estimation method was used based on the accumulated decay heat over the first 1000 years of a repository. The sensitivity of cumulative decay heat was tested, but it was not compared to any real models of thermal repository loading.

The same limitations outlined here will apply to all fuel cycle scenarios modelled. Comparing the relative merits of fuel cycle scenarios to one another is valid, as the same set up and analysis methods were used consistently for all fuel cycle scenarios, independent of the fuel cycle and reactor parameters used. As a result, the limitations outlined in this thesis were assumed to not be significant. The aim of this thesis was not to simulate realistic fuel cycle scenarios but to model a range of potential options for comparison. As such, the parameters selected in this thesis aim to give an idea of the performance of potential fuel cycle options and do not aim to accurately represent what a real life fast reactor fuel cycle would look like.

11.4 Further work

The purpose of the decision analysis chapter was to provide a framework for the user to determine which fuel cycle scenarios warrant further study. In addition, further work should assess more fuel cycle scenarios to add to the decision analysis framework as well as adding more assessment criteria for current fuel cycle scenarios. A primary focus of further work should be to develop a repository model to more accurately calculate repository size. Further work should also investigate fuel cycle facility and

transport requirements, particularly shielding, to compare facility requirements as an assessment criteria for scenarios.

Future fuel cycle studies of interest would focus on continuous fuel cycle scenarios, not just stockpile reduction methods. In line with the UK R&D pathway studies, it would be of interest to look at a similar nuclear power generating capacity, 16 to 75 GWe, operating a combination of LWRs and low-CR SFRs which would use reprocessed spent fuel from LWRs. Results from this scenario would be of interest to compare to the UK R&D closed fuel cycle scenarios which transitions completely to SFRs. These represent very different scenarios to the stockpile reduction scenarios modelled in this work.

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Appendix A

SFR components

This section discusses out-of-core material and components for current and future SFR development. This section was originally part of Section 2.3.3, as part of a technical report written by the present author for DECC [25], but has been moved to an appendix as SFR components do not feature in SFR design and fuel cycle modelling aspects of this study.

Near-term SFRs would be a three loop design, using a sodium intermediate loop, mechanical pumps and a steam cycle for power production [166]. Intermediate heat exchangers have typical designs and use austenetic stainless steels such as 316. For steam production, tube and shell steam generators (straight or helical coil) would use low-Cr steel, 2.25Cr-1Mo, for evaporators and austenetic steels or nickel alloys used for super heaters, such as 304, 316 or 800H [30, 41, 66, 167, 168].

In the medium- to long-term, a three loop system remains a promising configuration, but with more efficient, higher pressure steam generators using advanced designs for enhanced safety (e.g. double walled, inverted [36, 166], modular designs [33], tube-to-tube Hot Isostatic Pressed (HIPped)). High-Cr FM steels (9 wt%Cr [68]) are considered promising due to their higher resistance to carbon transfer [41], their ability to reduce the amount of material used and to increase operating temperature and pressure [169]. Double walled steam generators have been demonstrated on a small scale with EBR-II and more recently with small scale testing at JAEA [30, 167, 170]. Similarly, inverted steam generators have been successfully operated at BOR-60 and BN-350. The relatively large size of the BN-350 steam generator modules and the length of operation suggests that they have significant potential [51, 171]. Materials

for advanced steam generator design include 9Cr-1Mo variants, which are equivalent to ASME Grade 91. However, to the authors' knowledge, no prototypes have been constructed [42].

There is also the potential to move away from a steam cycle and towards a gas Brayton cycle (nitrogen, S-CO_2 or multiple reheat helium) [48,172,173,174,175]. This would utilise more compact PCHEs (with tube and shell as a fall back [36]). In the long-term, with more robust design and inert gas, there may be the potential to reduce a reactor to a two-loop system [176]. At present there is a significant body of research into Brayton cycles. Helium Brayton cycles have been developed and demonstrated with HTRs, but these differ in design to the multiple re-heat Brayton cycle intended for SFR use [174, 176]. For S-CO_2 Brayton cycles, research in the USA and France has focused on turbine development, materials tests and sodium-CO_2 interactions [48, 177, 178]. Also, France is considering an inert nitrogen Brayton cycle for use with ASTRID [166]. PCHEs for this purpose are already in industrial service and manufactured by Heatric in the UK [36, 179]. However, the fabrication methods used for PCHEs (HIP-diffusion bonding) have not been used previously in the nuclear industry and therefore require qualification [166]. There is limited evidence of PCHE testing with sodium, although research is planned at Argonne National Laboratory in the USA. The alternative tube and shell heat exchanger design is currently used for HTRs (gas to gas) and SFRs (sodium to water) but have not been used as a sodium-to-gas heat exchanger [176]. However, they have the benefit of using qualified materials and fabrication use, and significant experience in the nuclear industry [36, 174].

Also worth considering is the development of electromagnetic (EM) pumps. Despite lower efficiency compared to mechanical pumps, they can pump impure sodium and have been shown to have better reliability, simply due to the lack of moving parts [35, 39, 180]. General Electric (GE) and Toshiba have successfully built and demonstrated a large EM pump, which was tested under expected conditions and performed well [181]. Smaller EM pumps have been used for EBR-II and in other reactors for backup and purification circuits very successfully. CEA and Toshiba currently have an R&D project examining an EM pump for the ASTRID reactor. Computational and experimental investigations are currently underway [35].

Hot piping and reactor vessels are typically made of austenitic stainless steels such

as 316 [41]. While reactor vessel and piping materials are more than adequate, there is the potential for advanced alloys to increase safety characteristics during accidents and to reduce component size, making materials savings [169]. Some of these materials are qualified, or close to being qualified (e.g. Cr-1Mo-V ASME qualified up to 649°C [182]), but have limitations such as forging size. Without further development and qualification this prevents their use for larger components. Other advanced materials, such as NF616 and HT-UPS, have the potential to minimise component size and save on material weight but are not yet qualified for this purpose [169].

A.1 TRL and justification

Advanced steam generator designs and material are under consideration as they have the potential to increase efficiency, reliability and safety. The only problem with these concepts is the use of relatively inexperienced designs and the qualification of new materials and fabrication techniques. Steam generators using advanced materials or alternative designs have been assigned a TRL between 3 and 6 (depending on specific technology). Inverted steam generators have been successfully operated on a relatively large scale so they have been assigned a TRL of 6. Similarly, double walled tube steam generators have been assigned a TRL of 5 as they have been demonstrated on a smaller scale. Materials considered for higher efficiency steam generators, including 9Cr-1Mo variants, are assigned a TRL of 6 due to materials being qualified or close to being qualified. More advanced materials, joining and fabrication techniques such as HIPping have not been qualified and are therefore assigned a TRL of 3. The technology and understanding exists to manufacture such steam generators and to couple them to an SFR, but this has not been demonstrated.

There is considerable ongoing research on Brayton cycles using supercritical CO_2 and inert gases (nitrogen and multiple re-heat helium) that are much more efficient than Rankine steam cycles and deemed to have improved safety characteristics. PCHEs also have the added advantage of being very compact, robust and reliable, thereby minimising the probability of leaks. Leaks which do occur have less severe repercussions compared to traditional steam cycles. The working fluid is more chemically compatible with sodium and reaction rates are relatively slow due to the way leaks can occur in

PCHEs. Brayton cycles for power conversion have been assigned a TRL of 3 or 4. The TRL limiting factor of these designs is the use of PCHEs. PCHEs are in industrial service but the HIPing fabrication methods require qualification in the nuclear industry. Due to the PCHE being the main limiting factor, all Brayton cycles with PCHEs are assigned a TRL of 3. Backup tube and shell designs have been used for SFRs before but not as sodium-to-gas heat exchangers. Due to their use of conventional material and fabrication techniques they are assigned a higher TRL of 4 [36, 174].

In the past, large electromagnetic (EM) pumps have been overlooked due to their low efficiency and significant heat generation. Being restricted to use in secondary systems such as decay heat removal or coolant purification loops. However, there is significant interest in them due to improved reliability over mechanical pumps and the lack of moving parts which can fail or corrode. Large EM pumps for the primary coolant circuit are assigned a TRL of 5 due to successful demonstrations of the technology but not being demonstrated in an SFR.

SFR	**TRL**	**Justification**
SFR with advanced piping and reactor vessel materials.	4	Such materials have been qualified but not necessarily for use as piping or reactor vessel. Further testing and qualification is required but considering their present uses is SFR systems they are assigned a TRL of 4.
SFR with advanced steam generator designs.	3 – 6	Inverted steam generators have been tested with a large demonstration reactor, so are assigned a TRL of 6. The use of 9Cr-1Mo in steam generators is assigned a TRL of 6 due to it being ASME code qualified. Double walled straight-tube steam generators have been used on small prototype systems and are therefore assigned a TRL of 5. The use of advanced manufacturing techniques for steam generators is assigned a TRL of 3 due to limited experience and lack of qualification for use in the nuclear industry.

SFR with Brayton cycle.	3 – 4	Brayton cycle using a PCHE is assigned a TRL of 3 due to testing of coolant-gas interactions and the large use of PCHEs, yet lacking qualified manufacturing techniques for nuclear components. Using a tube and shell heat exchanger increases the TRL to 4 due to their widespread use but lack of experience as a sodium-to-gas heat exchanger.
SFR with EM pump for primary coolant loop.	4	EM pumps have been used in SFRs for small systems and for primary loops on smaller prototype reactors. Their use for large coolant loops has been demonstrated successfully but not in a reactor system.

Table A.1: SFR TRL summary table.

Appendix B

Fuel cycle facilities

An overview of fuel cycle facility TRLs can be found in Section 2.5. This chapter provides a supplementary review of the literature to support the TRL assessment in in Section 2.5.

B.1 Reprocessing

B.1.1 Aqueous

Aqueous reprocessing is based on the PUREX (Plutonium and Uranium Recovery by Extraction) solvent extraction process. The typical 'head end' of the PUREX process mechanically disassembles fuel to be dissolved in nitric acid, leaching all fuel and removing all undissolved particles and cladding to leave a nitrate feed liquor. The nitate is mixed with tri-butyl phosphate in orderless kerosene (TBP-OK) for the solvent extraction process. The organic (TBP-OK) and aqueous (nitric acid) liquids separate with the plutonium and uranium in the organic layer, leaving other actinides and fission product in the nitric acid. The nitric acid is treated as waste. Chemical conditions in TBP-OK are altered to extract the plutonium which is converted back to PuO_2. Flow sheets for the process vary in different countries but the basic principle of all aqueous solvent extraction processes are the same.

More advanced concepts look at co-extraction of plutonium with uranium or other actinides, or consider further separation steps to extract higher actinides from fission products. Examples of these processes that have been studied and demonstrated on

a lab scale are outlined in Table B.3. Advanced aqueous methods are currently being developed and have not left the demonstration or lab scale

PUREX Experience PUREX reprocessing techniques have been commercially deployed in several countries to reprocess thermal reactor, UOX fuel, as shown in Table B.1. Current burnup limit of UOX in modern reprocessing plants is 40 to 55 GWd/t [183]. Pilot and demonstration facilities to reprocess thermal MOX and fast reactor fuels have also been operated, and some commercial plants have been used to demonstrate reprocessing of these materials. An overview of some key commercial, demonstration and pilot PUREX reprocessing facilities can be found in Table B.1, which is not an exhaustive list of all reprocessing, just key information that is available from the literature.

Table B.2 gives an overview of key fast reactor fuel reprocessing programmes, using the PUREX process. This outlines the key achievements in terms of peak plutonium content, fuel burnup and shortest cooling time, as well as details of the programme.

Advance aqueous reprocessing experience Many advanced aqueous reprocessing techniques have been developed to extract MAs as well as plutonium for transmutation scenarios. Some techniques have been demonstrated on a lab scale with representative HLW feed. However, nothing has been demonstrated in a basic integrated system or a pilot plant. A summary of some key techniques can be found in Table B.3, these focus on two areas:

- Post-PUREX extraction of actinides – Actinide separation in one step after PUREX, (PALADIN, DIDPA, SETFICS). Or two steps: actinides and lanthanides together (TRUEX, DIAMEX), followed by separating actinides from lanthanides (SANEX, TALSPEAK, ALINA/CYNEX). This allows for further steps to separate americium from curium (SESANE) or caesium and strontium (CSEX, SREX, CCD-PEG) for storage in partitioning schemes.

- Group separation of plutonium with other actinides – Group extraction (GANEX, COEX, UREX, NUEX) are alternatives to the PUREX process to reduce the proliferation risk of a fuel cycle, with no isolated plutonium stream.

Table B.1: Summary of key pilot demonstration and commercial aqueous PUREX reprocessing plants adapted from Ref. [86] (as of 2003 unless stated otherwise), with additional information from: [85, 86, 87, 88, 89]

				Total Fuel Rep (tHM)		
Country	**Facility**	**Year**	**Capacity (tHM/y)**	**Thermal UOX**	**Thermal MOX**	**FR**
Pilot						
Belgium	Eurochemic	1966	60	105		
France	SAP	1963				8[1]
France	AT1	1969	0.135[2]			1
France	APM	1973	6			10.5
Germany	WAK	1970	35	180		
India	CORAL	2003				
UK [86]	Dounreay	1980	10[3]			14
USA [86]	NFS	1966	300	194		
Demonstration						
India [88]	PREFRE-1	1982	100	250		
India [88]	KARP	1996	100	230		
India	PREFRE-2	1998	100			
Japan	TRP	1997	210/90[4]	1000	18	
Commercial						
France	UP1	1958	600	18000		
France	UP2/3	1967	1700[5]	22450	150	100[6]
Japan	RRP	2012[7]	800			
Russia	RT-1	1977	400	3550		450
UK	B205	1967	1500	46000[8]		
UK	THORP	1994	<900	7120[8]		

[1] Mostly fast reactor fuel [90].
[2] One Rhapsodie core per year.
[3] Based on 0.03kg per day [89].
[4] [85]vs [86].
[5] 1000 for UP2 and UP3, 1700 total for site.
[6] FR fuel blended with UOX and reprocessed in presence of poisons [90].
[7] Aim (throughput not available) [110].
[8] 2012.

Table B.2: Fast reactor PUREX experience. Examples of minimum cooling times and peak burnup to give an idea of representative conditions for future reprocessing. Aadapted from Ref. [91] with additional information from: [87, 89, 90].

Country	Plant	Peak Enrich (Pu %)	Peak BU (GWd/t)	Min Cool Time	Additional info
France	AT1, SAP, UP2	30	120	1.5 y	Recycling up to 3 times through a FR. Cooling times ranged from 5–50 months, 5 months for 50-120 GWd/t was achieved. UP2 used poisons (Gd) and blended FR MOX with LWR UOX, 99.6-99.8% recovery for 33–41 GWd/t and 3.5–5 year cool. [87]
UK	Dounreay	<25	83	136 d	Blankets and fuel reprocessed. Peak decay heat of 3kW/assem. Pu recovery >99.8%. 3.7 tonnes reused in MOX in UK, most stored as PuO_2 [89]
Japan		18	40	1.2 y	
Germany	MILLI	30	100	10 m	Blankets and fuel reprocessed together.
Russia	RT-1	-	100	-	-
India	CORAL	0	155	2-6 y	Carbide fuels. Recovery of 99.8% Pu and 99.9% U
USA					Dissolution tests on small scale of very high burnup FFTF MOX 220 GWd/t [87]

Table B.3: Sample of advanced aqueous reprocessing techniques being studied, adapted from Ref. [92]. Note that technique name and flow sheet vary in different reports.

Method	Country	Details
Grouped extractions, aternative to PUREX		
GANEX	France Europe	Uranium extraction then group extraction of actinides [85, 92, 93]
UREX	USA	Uranium extraction then several variants with TRU, TRU+Ln in second stream and the potential for more streams with further separation steps. Lab scale experiments < 4 kg batch [85, 86, 87, 92, 95]
NEXT	Japan	U pre-recovery and U-Pu-Np (Possibly Am/Cm [101]) co-recovery. Lab experiments [86, 87]
COEX	France	Extract U+Pu or U+Pu+Np. A U+Pu (50:50) stream for MOX has been demonstrated at the lab scale [85, 87, 90, 101]
One step separation of An and Ln from FP (after PUREX)		
SETFICS	Japan	Modification of TRUEX to successively remove TRUs. Not efficient Ln removal. Tested on HLW from FR [87, 94, 95]
DIDPA	Japan	Successive stripping of each TRU from solvent similar to TALSPEAK. Tested in a hot cell [87, 94, 95]
PALADIN	France	Successfully tested at ATALANTE facility [87, 94, 95]
Multiple steps, extract An and Ln, then individual An's		
Step1	**Co-extract An and Ln after PUREX**	
DIAMEX, TRUEX, TRPO	China France Italy Germany Europe Japan USA Russia India	Varous tests on real waste [87, 90, 92, 94, 95]
Step 2	**Serpeate An from Ln**	
TALSPEAK, SANEX, ALINA, ARTIST	Russia USA Sweden France Germany China India EURATOM Japan	A lot of data worldwide on TALSPEAK. Various tests with real spent fuel and HLW [87, 90, 92, 94, 95]
Step 3	**Removal of Am from Cm**	
SESAME Am(V)	France, Japan, USA	SESAME CEA lab scale demonstrations. Am(V) high losses [92, 95]
Step 4	**Removal of others**	
CSEX, SREX, CCD-PEG	USA France EURATOM Czech	Extract Cs and/or Sr. Tested on effluents [92, 95]

B.1.1.1 Technology specific issues

Fast reactor fuel reprocessing differs from thermal reactor reprocessing as the fuel is irradiated to higher burnup and has a greater proportion of plutonium and fission products. As a result, the spent fuel has greater criticality concerns and produces more heat and specific activity. Originally, commercial reprocessing plants such as THORP and UP2 were reprocessing fuel upto 3% enriched fuel, irradiated to 30 GWd/t and cooled for 3 years. Now fuels are more in the range of 3.7% enriched and irradiated to 45 Gwd/t, needing a minimum of 4 years cooling to reduce the heat and specific activity. MOX fuel at the same burnup will be more active and hotter, causing the same issues as higher burnup fuel. Burnup for fast reactor fuels is higher that thermal fuels and uses MOX fuel [87].

Burnup limit of current thermal reprocessing plants is approximately 40–55 GWd/t. Higher specific activity leads to more alpha radiolysis causing more degradation of the solvent which causes the formation of cruds and reduce the efficiency of reprocessing. This can be solved with longer cooling times before reprocessing and blending spent fuel with other material such as UOX to reduce the specific activity. In UP2, MOX fuel dissolved in nitric acid was diluted with uranium fuel to get the ratio of plutonium to uranium to 2% [89]. In other demonstrations, advanced mixing methods (centrifugal contactors) have been developed to reduce the solvent-fuel contact time needed and therefore reduce the amount of alpha radiolysis [87, 183]. Despite these concerns, aqueous reprocessing of fuel with burnups up to 150 GWd/t have been successful with cooling times as short as 6 months, Table B.2.

MOX fuel has issues compared to UOX due to lower solubility of plutonium. As such the fuel needs to have a homogenous distribution of plutonium grains[1] and that there is enough porosity to ensure that all plutonium is reached and dissolved. This limits the proportion of plutonium in the fuel whilst still ensuring complete dissolution [183]. The limit for complete solubility of MOX is thought to be around 40–45% for conventional PUREX methods [89, 118]. However, the CORAL facility in India has reprocessed fuel with enrichments up to 90%. Also, due to the greater fissile content, absorbers may be needed and different plant geometry to prevent criticality

[1] It is easier to dissolve irradiated MOX as the plutonium distribution becomes more homogeneous. In Russia better dissolution was achieved by adding HF, however this can cause component dissolution leading to failures [89]

[87]. Thermal and fast reactor MOX have been reprocessed successfully, homogeneity issues in irradiated MOX are not considered a stopping point due to advances in MOX fabrication and changes in heterogeneity during irradiation [89].

Explosions can occur when dissolving uranium-zirconium compounds in nitric acid, such as aqueous reprocessing of U-Pu-10Zr metallic fuels. This can be overcome with large enough quantities of fluorine present [96, 97, 98]. However, there is limited experience reprocessing zirconium based metallic fuels, therefore, less commercially developed reprocessing techniques could be favourable, such as pyro-reprocessing, given that both techniques could have similar development time and costs. However, this would need further investigation and is outside the scope of this study.

Issues with most advanced aqueous reprocessing techniques are based around the need to reduce acidity to low levels for extraction, which may be difficult on a large industrial scale. Most advanced methods have no defined method to clean solvents or deal with effluents and waste streams. Some methods also have issues with solvent degradation and loading which have not been solved [87]. Many studies believe that advanced aqueous reprocessing can lead to extraction of all U-Am isotopes with losses $< 0.1\%$, with Cm levels ranging from $> 1 - 0.1$ %, and a total contamination of MAs with $< 5\%$ of lanthanides [101].

B.1.2 Dry

Molten salt electro-refining uses a basket filled with chopped fuel as an anode. Fuel is dissolved into the salt and uranium is deposited onto on a solid cathode (eg. solid steel). TRUs are then deposited onto a liquid cathode (liquid due to improved kinetics, typically cadmium or bismuth), typically with some proportion of lanthanides. Choice of salt and cathode materials varies but the process is typically the same [87]. Similarly, oxide electro-winning involves the dissolution of oxide fuel with the deposition of products onto cathodes as oxides. Adaption of molten salt electro-refining and oxide electro-wining processes can lead to the co-deposition of uranium and plutonium. Each element in the salt has a different potential which is dependent on the salt and cathode materials. In some cases TRUs are close together, in others they are further apart allowing for a choice of co-deposition or selective deposition of actinides. In most systems there is a significant amount of lanthanides extracted with TRUs. A

Fluoride volatility methods powder the fuel which is then fluorinated with pure fluorine gas [184]. This forms volatile and non-volatile fluorides which is used as the basis to remove FPs from TRUs. Further separation of Pu and U can be done based on their thermodynamic stability [101,185]. Further separation steps are used to clean up the product streams from lanthanides and fission products, and potentially isolate pure actinide streams.

Experience Pilot to demonstration scale plants using pyro-reprocessing techniques have been successfully operated. Most of these programmes have stopped but research is still ongoing at the experimental level, Table B.4 and B.5.

Key pilot programmes to note are the Russian Dimitrovgrad Dry Route (DDR) which has been successfully used to produce material for fast reactor fuel and reprocess very high burnup MOX fuel. In the USA similar pyro-reprocessing techniques are being used to extract and purify uranium from EBR-II spent fuel, this has been successfully piloted and a larger refiner will be built to undertake the reprocessing contract to extract 23 tonnes of U-Zr fuel and down blend it before storage.

Fluoride volatility methods have been piloted for fast reactor fuel in several counties but development has slowed since the end of Molten Salt Reactor (MSR) development. Most notable work has been in France using Rhapsodie fuel and in Russia using BOR-60 fuel. In the USA and Russia this method was adapted for uranium purification post-PUREX reprocessing, although this did not progress past the design and flow sheet stage.

B.1.2.1 Technology specific issues

Deployment of pyro-reprocessing methods will need considerable R&D to get it to a commercial scale. In addition, development is needed for a suitable waste form. Unlike aqueous reprocessing there is no experience from operating large plants which are comparable in design [186] therefore it is expected to take longer to deploy than advanced aqueous methods. However, pyro-reprocessing is a batch processes, therefore scaling to a commercial scale is less significant, requiring smaller facilities but more of them rather than larger batches. Cost of scaling up will depend on the number of batch processes and economies of mass production rather than economies of scale [99].

Table B.4: Samples of key pyro-reprocessing experiments and pilot plants.

Country	**Scale**	**details**
USA	Exp	LANL tested FP removal from molten plutonium LAMPRE fuel [87]
USA	Exp	ORNL extract U and Pa from MSRE fuel [87]
USA	Pilot	EBR-II fuel melt refining. 2.3 tonnes of fuel with a 2 month turn around, 7% HM losses [87, 99, 105]
USA	Pilot-Demo	Extracting U from EBR-II fuel (developed for IFR TRU rep). 1 tonne demo (MK-IV refiner), 23 tonnes to be treated (MK-V refiner, 100 kg batch, 1 tonnes/year), uranium metal for storage and reuse, FP and TRU waste. 99.7% U recovery demonstrated as well as co-deposition of Zr and U [87, 94, 95, 99, 100]
USA	Lab	GNEP TRU extraction. 50 kg batch efficiency tests of TRU element based fuel [87]
USA	-	PYROX process developed for LWR. Deposition demonstrated for vipac, 3.5kg batch > 99.95% pure Pu [99]
Russia	Demo-Pilot	DDR – Pyrochemical production of fuel for fabrication and Kg scale batch reprocessing of MOX upto 240 GWd/t. Several scales: glove box, hot cell, military Pu facility and semi-industrial complex producing 100's Kg Pu per year. Semi-industrial complex achieved 99.6% U and 99.95% Pu recovery for unirradiated fuel, 95.6% for irradiated fuel tests (expected to get to 99% for industrial deployment). 7.2 tonnes of fresh fuel and 40 kg of irradiated processed. [95, 99, 101, 102, 103]
Russia	Exp	DOVIA programme actinide burner. Co-deposition of U+Np, U+Pu+Am (Am only partially). Cm and RE are left over, Am left in salt [101, 102, 104]
UK	Exp	Electrowinning of UOX in reduction step and U-Pu-Ma-Ln separation. Pu separation on 50 gram scale done in collaboration with Korea and Japan and EURATOM [99]
Japan	Exp	Metal fuel reprocessing and reduction of oxides. Gram quantities, US and EUROTOM collaborations [87, 92, 99]
Korea	Exp	Reducing oxides to metal and uranium extraction from UOX. 20kg batch reduction, 1kg batch U exaction (mixture made, not real waste) [87, 99]
EURATOM	Exp	France, extract MA from aqueous waste [95, 99, 103]
EURATOM	Exp	Metallic fuel extract An and decontaminate FP from irradiated and unirradiated METAFIX fuel with 5% MA content [105]. Getting 97% actinide recovery and 3% Zr included [95, 99, 103]

Table B.5: Samples of key fluoride volatility experiments and pilot plants.

Country	**Scale**	**details**
France	Pilot	1962-1972 ATTLIA reprocessing Rapsodie fuel focused on pureificaton of uranium stream. 10 kg batch, MOX 25% enrich, 50 GWd/t and 6 months cooling. U recovery 97.45%, Pu 95-97.5% [87, 95]
Belgium	Pilot	1960-1968 SCK-CEN MOX FR fuel. 10 kg batch [87, 95, 99]
Japan	Pilot	5 kg batch Urnaium fuel [185]
Japan	Design	FLUOREX FR and LWR fuel. Designed to produce U-Pu MOX to be directly fabricate [87]
USA	Pilot	Military reactor, research reactor and MSRE fuel. Up to 40 kg batch of fuels. [87, 99, 185]
USA	Design	The Morris, reprocessing plant with fluoride volatility for uranium decontamination after PUREX. Never operated with hot fuels [99]
Russia/czech	Pilot	FREGAT, U fuel and MOX up to 100 GWd/t, 3-6 months cooling. 99.4-99.6% uranium. 85% of FPs were concentrated into residues waste stream stream. Only 89-91% of Pu was recovered. Got to scale of 3 kg/h [87, 99]
Russia	Design	RT-2 flow sheet design to incorporate uranium purification [87]
Czech	Exp	FERDA continuation of USSR work. Experiments and flow sheet design [184]

Smaller batch-wise facilities can be used on-site rather than large centralised facilities (as is the case with aqueous methods). On-site facilities requires no off-site transport, minimising proliferation risk and transport costs [92].

Pyroreprocesing methods also tend to be insensitive to burnup, fissile content and fuel type. The methods are chemically stable compared to aqueous methods, as such they can handle greater heat and radiation doses. This allows for shorter cooling times, which results in a shorter turn around time and less fuel storage required [92]. The methods of reprocessing do not contain moderators and are smaller so reduce criticality concerns [99].

Some early test have low separation efficiencies, with losses generally higher than aqueous methods and more impurities [95]. Lower losses and greater efficiencies have been demonstrated but there is a lot of contamination of materials such as La in most systems, and mixed MA streams [100]. In the USA, contaminating TRU feed with $<$ 5% LA is considered good. This can be negative if pure streams are wanted (extra separation steps would be required) and in terms of handling with respect to doses and facility shielding. However, higher doses can be advantageous in terms of proliferation

resistance as the dirty fuel streams have a high dose and heat and do not contain pure fissile material. It also allows TRUs to be recovered in one or very few steps, simplifying the extraction process for transmutation scenarios [92].

Electrochemical Specific issues related to electrochemical methods includes cleaning salts, impurities and waste forms. Zirconium is present in metallic fuels over 10 wt.%. Zirconium can get carried over during reprocessing and would be left dissolved in the system in large quantities. As such, extraction steps are need to remove Zr, or methods designed for the co-deposition of Zr and U [100]. The salt collects FPs from fuel as well as other fuel materials, such as sodium from sodium bonded fuels. This leads to reduced efficiency, and several ion exchange steps are needed to clean the salt. Even with salt cleaning the salt will needs to be disposed of eventually. There are generally issues when it comes to purification of salts and waste form design that require more R&D [187]. Another issue is with the conditions and materials. Material choices are difficult due to high temperatures, doses and aggressive condition with changing chemistry. Also, there must be a low oxygen environment which is hard to maintain [99].

Fluoride volatility Fluoride volatility has some specific advantages and disadvantages. Plutonium changes between being between a volatile and non-volatile fluoride reducing separation efficiencies (98-99.5%) [99] and neptunium is difficult to isolate (60-70%). Americium and curium are not separable in the non-volatile stream, needing additional separation steps. Overall it is difficult to form a concentrated FP stream and and a pure plutonium stream [87]. The biggest technical issues is with heat removal as volatilisation is a very exothermic reaction in a confined area and heat needs to be removed. This is even more of an issue when considering the need for cooling and a small geometry for criticality safety. Remote operation is needed due to high temperatures, radiation and the use of fluorine [99]. Some advantages over electrochemical processes is that UF_6 can be used directly for re-enrichment.

B.2 Fabrication and fuel experience

B.2.1 MOX

There are many processes used to fabricate MOX pellets, these will not be discussed in favour of a general overview of the process. Mechanical blending (atritors or ball mills typically) of feed powders UO_2 and PuO_2 produce a uniform distribution of powders. Frequently lubricants are used and additives to lower the density of FR MOX and help pore formation and granulation. Pellets are then pressed into green pellets and sintered in a furnace to form fuel ready for use in a reactor.

For VIPAC MOX, in the case of material produced from pyro-reprocessing routes, MOX deposited on an electrode is crushed to the size needed, mixed and loaded into a rod which is axially vibrated. This is advantageous as it can use reprocessing products directly loaded into a pin. The VIPAC process has fewer steps than the pellet process and does not require the movement of powders which is difficult and a dose hazard.

Experience Over fifty thermal reactors have used MOX fuel and eleven fast reactors. The main pilot and commercial plants used to fabricate these fuels are outlined in Table B.6. Thermal MOX has been fabricated on a commercial scale in several countries, most notably in France at the Melox plant. Fast reactor MOX has been fabricated up to a demonstration scale in France at CFCa, making fuel for Rhapsodie, Phenix and SuperPhenix. RIAR in Russia is the only facility to properly pilot VIPAC fuel which was tested in BOR-60, BN-350 and BN-600 reactors [106]. A full list of fast reactors, the fuels that have been used and operating conditions can be found in Table 2.4.

Thermal MOX fabrication experience does not directly translate into fast reactor MOX experience. They both have different goals, whilst some facilities (BN/Dessel, Hanau, CFCa) have manufactured both, it would be expected that different facilities in a fuel cycle would be used to fabricate each type of fuel. Fast reactor fuels need higher enrichments so criticality and doses are a bigger concern in terms of shielding and geometry of a plant. The fuel fabrication itself is slightly different, the fuel must be lower density or annular to allow space for fuel restructuring, and there needs to reduced the oxygen to HM ratio to minimise fuel-cladding chemical interaction.[2]

[2] $(PuU)O_2$ extect O/HM ratio to be 2. For a fast reactor 1.98 or less is preferable to reduce oxidation and corrosion.

Table B.6: MOX fabrication experience as of 2000, adapted from Ref. [89] with peak Am data from Ref. [106].

Country	**Facility**	**Year**	**Fuel**	**tHM/y**	**Method**	**Total tHM**	**Max Am (%)**
Belgium	BN/Dessel	1973	TR	35[1]		467	1
Belgium	BN/Dessel	1973	FR	35[1]		4.2	1
France	CFCa	1962[2]	FR	10[3]	COCA	110	
France	CFCa	1990[2]	TR	40[4]	MIMAS	248	1
France	MELOX	1995	TR	100	MIMAS	455	3
Germany	Hanau	1972	TR	20-25[1]	OCOM	158	
Germany	Hanau	1972	FR	20-25[1]	OCOM	5.9	
Germany		[5]	TR	120			
India	BARC	1994	TR	18	Conv	3	
Japan	PFFF[6]	1973	FR	10[7]	Conv	4	1
Japan	PFFF[6]	1972	TR	10[7]	Conv	120	1
Japan	PFPF	1988	FR	5	Conv	10	3
Russia [103]	RIAR	1981	FR	1	Vipac	7.2	
Russia	Paket	1986	TR	0.3	Conv	1.4	
UK	DMP	1970	FR	0.7[8]	[9]	13	
UK	MDF	1994	TR	8	SBR	14	3.6
UK	SMP	2002	TR	40[10]	SBR	5.5[11]	3
USA	[12]		TR	50-70			

[1] When LWR only
[2] Two pilot scale FR then converted one LWR only in 1990.
[3] 0.5 Rapsodie line to 4 for phenix to 20 of Superphenix
[4] If no FBR fuel made on FBR line
[5] Consructed but never operated
[6] ATR and Fast lines
[7] Based on ATR and FR fabrication fabrication as well
[8] 13 tonnes over 1970-1988 or PFR
[9] VIPAC was tested but other routes used [89]
[10] Temporarily suspended due to FBR leak
[11] 2007
[12] Total over 5 sites
[13] OCOM/MIMAS

MA bearing MOX fuel Advanced MOX fuels containing MAs have only been fabricated on a lab scale. Table B.7 lists key experiments where MA containing MOX fuels have been irradiated in reactors. These come in the form of low proportions of MAs for Minor Actinide Bearing Driver Fuel (MADF) for homogeneous transmutation strategies and high proportions of MA for Minor Actinide Bearing Blankets (MABB) for heterogeneous transmutation strategies. In addition to these in-core tests there have been additional tests on the fabrication of MA bearing MOX fuels, but the most significant tests have also been used for irradiation programmes. Low fertile and inert matrix fuels have not been included here as they are outside the scope of this study. Here the focus is on oxide driver fuels in the form of ($< 45\%$TRU,U)O_2, this helps limit options for SFR down selection and increases the TRL of design options. It is reasonable to assume that TRU fuels up to the same enrichments as those used in plutonium only fuel will have a higher TRL than higher enrichments or inert matrix fuels which have not been qualified for use in reactors. Low fertile and inert matrix fuel studies include: EBORA, EFFTRA, ECRIX, CAMIC, COCHIX, and HELIOS.

B.2.1.1 Technology specific issues

Fabrication Powders used for pellet fabrication are hard to move and handle. They can can be problematic in terms of the dose hazard. Powder-less routes would be preferable and have been demonstrated, but all commercial technologies currently use powder methods. For MA bearing fuels, the high volatility of americium is an issue and must be managed to make sure it is retained during the sintering process [58]. Due to higher plutonium content and potential MAs in FR MOX fuel, doses will be higher. To reduce worker doses at the Melox plant, a lot of automation was used. A high level of automation and shielding will be needed for advanced FR fuel [89]. If curium is to be included, more shielding will be needed due to curium being a strong neutron emitter. This will pose difficulties and, to the author's knowledge, no curium bearing fuels have been fabricated on any scale.

VIPAC fuel, although not represented very well in terms of demonstration, is being developed in many countries to simplify the fabrication process. Its use can be integrated with advanced on-site reprocessing techniques minimising the need for powders

Table B.7: Table summarising MOX MA bearing fuel irradiation experiments. Examples show uranium bearing fuel with <45% TRU content [57, 58, 61, 188, 189, 190].

Experiment	Irradiated	Fuel	Burnup	kW/m
SUPERFACT[1]	**Phenix**	**Pellet - Dust free - Sol-gel Fab**		
		2%Am 24%Pu	6.4-6.8 at%	38
		2%Np 24%Pu	6.4-6.8 at%	38
		45%Np	4.5-4.6 at%	28
		20%Np 20%Am	4.3-4.5 at%	27
FUJI[2]	**HFR**	**VIPAC**		
		5%Np 20%Pu	6 days	53
DOVITA[3]	**BOR-60**	**VIPAC**		
		3-6% Np 20% Pu	12.5-19.5 at%	
		5% Np	12.5-19.5 at%	
AM-1[4]	**JOYO**	**Pellet - Powder**		
		3-5%Am	10 mins to	43
		2%Am 2%Np	24 hours	43
AFC-2[5]	**ATR**	**Pellet - Powder**		
C		2%Np 3%Am 20%Pu	5.8-8.4 at%	22-32
D		2%Np 3%Am 20%Pu	13.3-19.1 at%	22-32
SPHERE[6]	**HFR**	**(P)ellet & (V)IPAC**		
		(P) 3%Am 20%Pu	18 months	30
		(V) 34%Am 22%Pu	18 months	30
Amboine[7]	**BOR-60**	**VIPAC**		
		$(UAm)O_2$		
GACID[8]	**MONJU**	**Pellet - Co-precip**		
		3%Am 20%Pu		

[1] France/EURATOM. Irradiated with 28% Pu MOX, fuel restructuring about the same. No migration of Pu/Am, cladding corrosion about the same and full helium release [58].
[2] Japan/EURATOM focused on thermal effects and restructuring. 48 hour ramp up to linear power, 96 hours at linear power [57].
[3] Russia Np containing fuels fabrication is no different to Pu fuel [57].
[4] Japan looking at migration of Am. More Am in centre, near void [57].
[5] USA Tested using Cd filter to emulate fast reactor [61].
[6] ERUATOM compare sphere pac and pellet fuel. Not finished at time of producing this table [58, 190].
[7] France/Russia mostly an Am recycling study to separate Am from RE [57].
[8] France/Japan/USA planned for future 2017 [57] [58].

and the processes required between reprocessing and fabrication. The minimal number of steps and direct fabrication of reprocessing products reduces the complexity of a plant, which in turn reduces the operator dose, recycling time and makes remote operation much easier. VIPAC was tested in the UK with PFR, however the distribution of fuel in the cladding lead to a high peaking factors in some areas of the fuel and corrosion [89]. Despite the UK's issues it has been demonstrated in Russia with good results, irradiating 3.5 kg of fuel MOX fuel [99, 103] upto 210–240 GWd/t in BOR-60 [87]. However, development time for VIPAX compared to pellet methods is likely to be slow due to less experience.

In-core issues Lenticular pores and the central void in the fuel starts to form in the first few minutes. Porosity migrates towards the fuel centre, along the thermal gradient due to vapourisation-condensation [191]. Fuel also sinters in-core causing grain refinement and restructuring. Fission product migration occurs due to irradiation and high temperatures and fuel restructuring.

The addition of TRUs reduces the melting point and conductivity of fuel, increasing the temperature gradient and centre line temperature, enhancing the migration of more mobile species [58].

Fission gas release is approximately 80% for 10 at% burnup in oxide fuels [192]. The release of fission gasses reduces swelling, but a plenum region above or below the fuel is needed to accommodate the release of gas with out a large build-up of pressure in the fuel pin. More fission gasses are produced from MOX than UOX, which is even higher with TRU fuels. Americium transmutation in reactor leads to a lot more helium production which can cause more fuel swelling during irradiation [192]. As such an even larger plenum region would be needed.

Fuel cladding interactions causes more than 40% of failures in fast reactors. One process is Fuel Cladding Chemical Interaction (FCCI), the other is Fuel Cladding Mechanical Interaction (FCMI). A reduced oxygen potential is required to minimise oxidation of inner cladding. Volatile fission products can migrate to the cladding edge and cause dissolution of cladding components [193]. FCCI increases with temperature and burnup. FCMI is less of an issue as the smear density of fuel is chosen to minimise the mechanical interaction between fuel and cladding.

VIPAC fuel tends to be in contact with cladding from very beginning of irradiation, increasing thermal conductivity across the fuel pin when compared to pellets, thus reducing the centre line temperature [194].

Oxide fuels are not very compatible with the coolant. In the case of a cladding breach, the fuel coolant interaction leads to the cladding breach to growing and a large amount of fuel lost to the coolant.

B.2.2 U-TRU-Zr

China, India, South Korea and the USA consider metallic fuels as a long-term design goal for SFRs [45, 46, 47, 48, 49]. The main fabrication process is injection casting where a crucible is heated by an induction coil, the molten material is given time to evenly distribute itself in the melt and a vacuum is used to force molten fuel into a mould. This is cooled, the moulds removed and fuel cut to size [150]. The amount of fuel fabricated at once is restricted by criticality safety [57]. This method was used to make americium containing fuel for the X501 experiment and a lot of americium was lost due to volatilisation with impurities in the feed stock. Americium volatility is not expected in typical americium feeds, but americium volatility and impurities cause the most significant issue in fabricating americium bearing fuels [57,149]. Other fabrication methods have been tested, including arc casting which aims to speed up fabrication and prevent the loss of americium [57, 195, 196].

Experience Many uranium based fuels have been fabricated for thermal military reactors, early gas-cooled commercial reactors and some fast reactors: DFR (U-Cr, U-Mo), EBR-I (U-Zr, Pu-Al) and EBR-II (U-5Fs, U-Zr). The performance of these fuels was varied. Most early fuels experienced large swelling, FCMI and FCCI. The only fuels viable from these demonstrations for a fast reactor are those developed for EBR-II. These were mostly based on zirconium as an alloying element to improve dimensional stability.

U-10Zr was demonstrated and qualified using EBR-II and FFTF for whole core use in EBR-II, up to approximately 10 at% [49], but achieving peak burnups of 20 at% (burnup was cladding dependent). These fuels were manufactured at the ANL-W coldline [107]. Following this, fabrication and demonstration of the of Mark-IV

plutonium bearing fuel (U-Pu-10Zr) began for the IFR programme with burnup in the range 10-20 at%. There were also tests of higher enrichment fuel for PRISM in EBR-II and FFTF. Qualification of U-Pu-10Zr was cut short due to the end of the programme and closure of EBR-II [108], with only 244 fuel pins manufactured [107]. Studies have shown that U-Pu-10Zr fuel is limited to an enrichment of 30%, with some fuel being irradiated that was 31.1% Pu/HM (X489 was ongoing when EBR-II was closed) [108]. Plutonium containing fuels were mostly tested with plutonium contents of around U-19Pu-10Zr, with fuels tested with plutonium contents ranging from 17-28 at.% overall [49]. U-19Pu-10Zr fuel reaching a peak burnup of 19.3 at.% in the X425 experiment [49].

MA bearing metallic fuels MA containing fuels have been fabricated by injection casting, in the case of the X501 tests, using the same equipment as EBR-II plutonium fuel fabrication. Arc casting methods have also been used on a lab scale. 10Zr metallic fuels, like those discussed above, containing MAs have been irradiated in tests along with fuels with higher zirconium contents to maintain fuel stability at higher enrichments. These experiments have been summarised in Table B.8. Experiments cover the fertile containing MADF and MABB fuel enrichments that were discussed at the beginning of this section. Higher enrichment fuels require a higher zirconium content due to changes in the fuel melting temperature and conductivity, with a higher zirconium content offsetting the issues associated with higher enrichment.

Metallic fuel selected for USA the GNEP 1000 MWth SFR is U-28TRU-10Zr, with average and peak burnup of 13 and 17 at% respectively, which is cladding dependent. USA GNEP fuel is similar to the fuels selected by KAERI for their small burner design which suggests enrichments up to U-30TRU-10Zr [57, 150].

B.2.2.1 Technology specific issues

Fabrication There is less experience fabricating metallic fast reactor fuels than MOX fast reactor fuels. For varying TRU, Pu and Zr quantities the phase diagram of the fuel changes and there are a lot of unknowns in terms of fuel density and phases. During the fabrication of TRU bearing fuels the retention of americium is difficult in injection casting methods [57].

Table B.8: Summary of MA bearing metallic fuel irradiation experiments, excluding fertile free materials.

Experiment	Reactor	Fuel	BU (at%)	Peak lin kW/m
X501[1]	**EBR-II**	**Injection casting**		
		U-20.2Pu-10Zr-1.3Np-1.2Am	7.6	44.9
METAPHIX[2]	**Phenix**	**Ark casting**		
		U-19Pu-10Zr-2MA-2RE	2.5/7.1/11.2	31/28/25
		U-19Pu-10Zr-5MA	2.4/7.0/11.2	33/29/27
		U-19Pu-10Zr-5MA-5RE	2.6/7.5/11.9	31/28/26
AFC-1F/H[3]	**ATR**	**Ark casting**		
1		U-29Pu-4Am-2Np-30Zr	4.4/26.7	26/25
2		U-34Pu-4Am-2Np-20Z	2.8/17.8	22/22
3		U-27Pu-3Am-2Np-40Zr	5/30.2	25/23
4		U-29Pu-4Am-2Np-30Zr	3.1/18	19/17
5		U-28Pu-7Am-30Zr	4.6/26	27/25
6		U-27Pu-3Am-2Np-40Zr	3.9/22.5	19/18
FUTURIX[4]	**Phenix**	**Ark casting**		
		35U-29Pu-4Am-2Np-30Zr	7	27

[1] For USA IFR programme. Aim to look at homogeneous recycling fuels. A lot of americium lost in fabrication. Restructuring during irradiation similar to U-20Pu-10Zr [49, 57].
[2] CEA/EURATOM/Japan homogeneous transmutation of MAs [57].
[3] USA: F to 94.3 EFPD and H to 653.3 EFPD [60].
[4] USA/Japan/Eutraom/France [57].

In-core The stability of different U-TRU-Zr alloys under irradiation requires development considering TRU and FP migration and new phases forming.

Metallic fuels have a lot of swelling due to fission gas build-up. Radially fuels swell up to the cladding edge, and axially U-Pu-Zr fuels swell by 3 to 4% [150]. A smear density of 75% is required to prevent fuel cladding mechanical interaction [150, 197].

Uranium and zirconium migrate to form zirconium rich regions in centre and edge, leaving a uranium rich intermediate zone, with plutonium staying evenly distributed. This is beneficial as there is a zirconium rich layer at the cladding, minimising FCCI, particularly the inter-diffusion of fuel with cladding [198]. Zirconium in the centre of the fuel also improves thermal conductivity across the centre of the fuel pin, reducing the peak centre line temperature.

Zirconium fuel and sodium are chemically compatible. A sodium bond is typically used in the fuel pin to improve thermal conductivity across the fuel-cladding gap. In addition, the liquid sodium bond can fill any pores that are made by fission gas release, ensuring that metallic fuel has a high thermal conductivity throughout burnup. Compatibility with sodium is so good that fuel can be operated safely after a cladding beach with no fuel-coolant interaction and only the loss of volatile fission products [199]. This is very advantageous over oxide fuel, where fuel-coolant interaction propagates fuel failure, potentially leading to failures in adjacent rods.

Appendix C

ERANOS supplementary work

C.1 IAEA benchmark problem

Extended version of Section 6.2.1.2 containing more detail.

C.1.1 Results compared to CEA ERANOS2.2

Results are compared directly to CEA's ERANOS2.2 results, assuming that the same geometry and solution method were used. CEA used JEFF-3.1 and JEF-2.0 cross-section libraries with ERANOSS2.2, it is unclear which one was used for the inventory calculation, but the k_{eff} values used JEF-2.2. The University of Manchester only has access to the JEF-2.2 cross-sections and ERANOS2.0, which could be a source of some discrepancies. Other small discrepancies could come about due to the difference in mesh sizes, user defined branching ratio and the cell lattice calculation procedure.[1]

Figure C.1 shows the relative change in fuel inventory compared to CEA for the in inner fuel region and inner fertile zone. The important neptunium and plutonium isotopes are within 4% of CEA's with ^{238}Pu and ^{242}Pu being furthest out. Americium results are up to 8% different. Curium is further out, greater than 10% for some isotopes. In the inner fuel region ^{240}Pu changes by the greatest mass, but this is small in proportion to the amount in the fuel region. ^{241}Am and ^{242}Cm vary compared to CEA's results by a few hundred grams which is equivalent to approximately 1% of the total americium and curium inventory of the region. The large percentage difference

[1]Many of these were manually adjusted to see how they influenced results. The only significant factor was mesh size (Appendix C.2).

of curium in the breeder region is very small in terms of real mass.

C.2 Sensitivity of model variables in ERANOS

There are three key parameter variables in ERANOS that were tested to determine how sensitive k_{eff} and burnup inventory results were to these variables. These were mesh size, the number of axial burnable zones and the time between flux re-calculation steps in burnup. Some guidance was taken from other publications [163], but these previous studies in the literature tended to look specifically at safety aspects, such as reactivity coefficients, which require more in depth analysis than k_{eff} and inventory calculations. As such, sensitivity calculations on these aspects were made to optimise these values for fast runing calculations whilst maintaining accuracy.

Figure C.2 shows that results are influenced by varying mesh size, axial burnable zones and burnup time step. The first y-axis represents the percentage change from the most rigorous calculation (smallest mesh, most axial burnable zones or smallest time steps between flux re-calculation). The second y-axis represents the approximate computation time. There is some noise in the computation time due to other processes taking place on the machine. The white points represent burnup reactivity loss, which was more sensitive to changes than the initial k_{eff}, and the black dots represent the change in ^{241}Pu consumptions.

It is clear that reactivity changes are more sensitive to changes in the model, where as the plutonium inventory is relatively insensitive. The number of burnable zones and flux calculation time steps have a more significant influence on results than mesh size.

C.3 ^{241}Am and ^{242}Am Branching Ratios

^{241}Am and ^{242}Am branching ratios are function of neutron energy. The choice of branching ratio has an influence on k_{eff} as ^{242m}Am has a significant positive contribution to reactivity.

Metallic fuel branching ratios for reactor design scoping work will be different due to the harder neutron spectrum in metallic fuelled SFRs. These branching ratios were taken from Ref. [200] as 83%$\rightarrow^{242}$Cm.

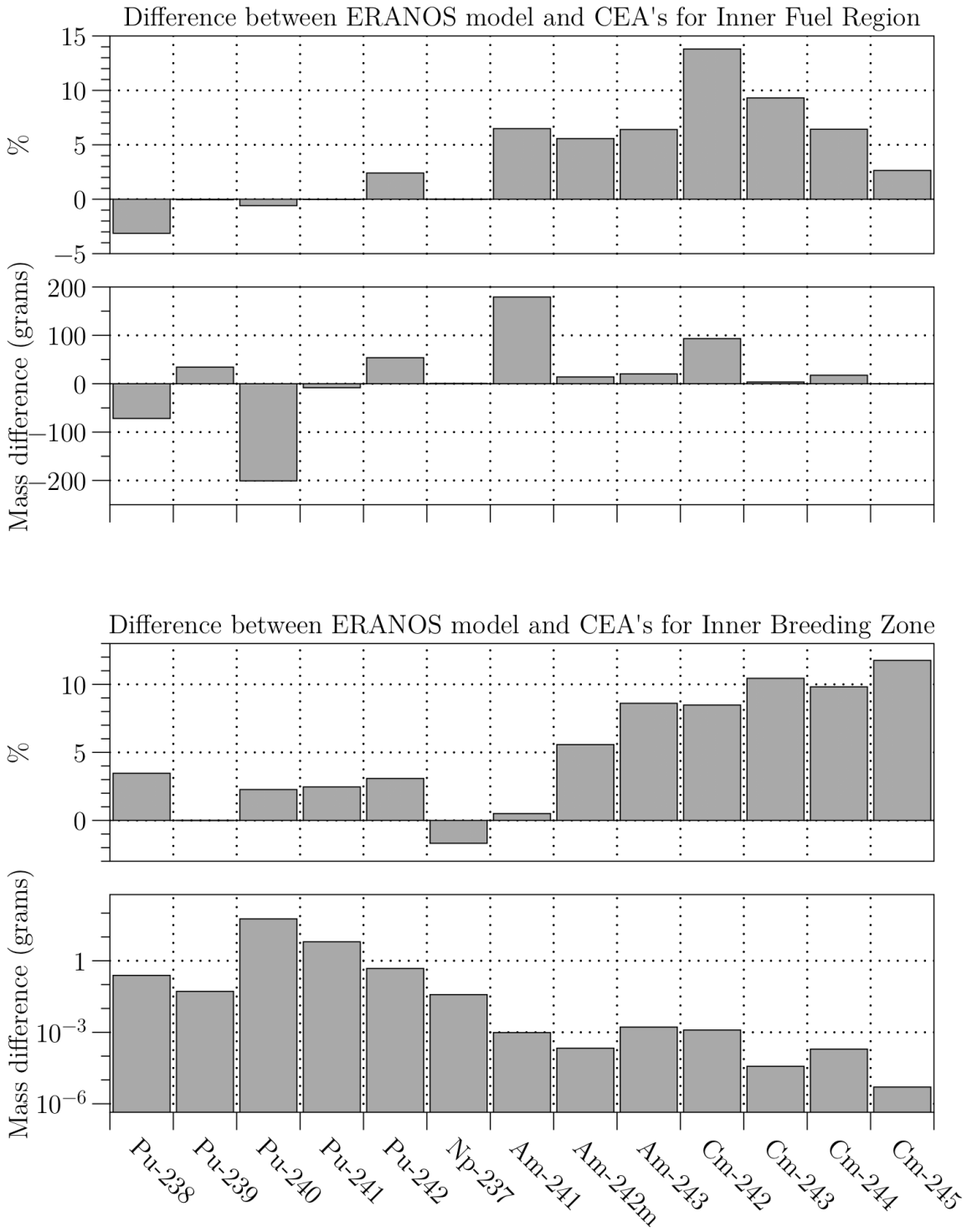

Figure C.1: Present author's BN-600 benchmark results compared to CEA's ERANOS2.2 results, taken from Ref. [146]. Results show mass and percentage difference to CEA results.

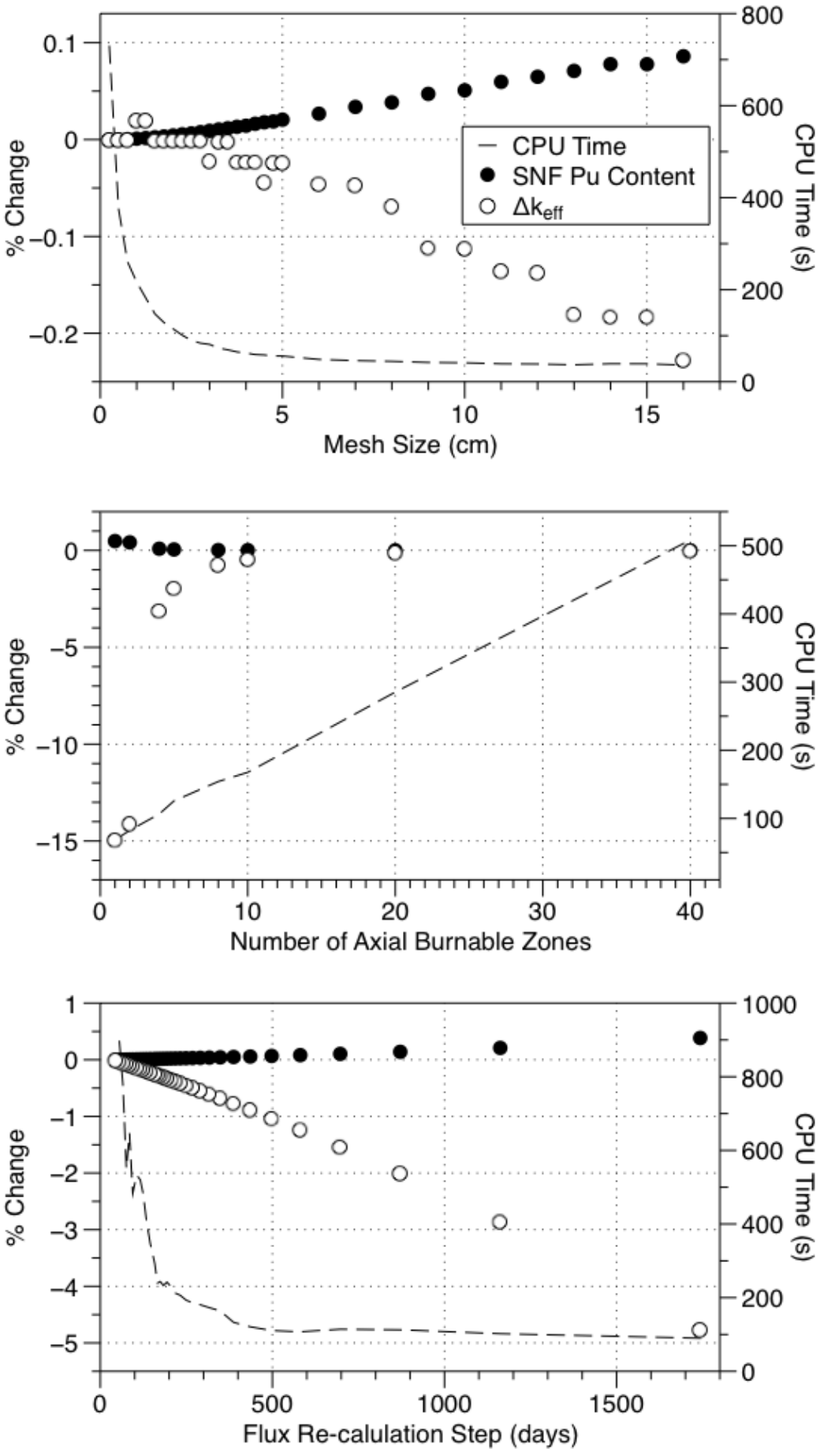

Figure C.2: Differences in run time, burnup reactivity loss and ^{241}Pu content with varying mesh size, axial burnable zones and burnup steps between flux recalculations.

Table C.1: Influence of branching ratios on Δk in IAEA benchmark case Ref. [146].

	Am241→Am242f	**Am242f→Cm242**	**Δk**
ERANOS Tutorial	85	84	443
ERANOS Adjusted	85	82.7	447
Proprietary	89	84	515
From Ref. [201]	90	82.7	539
From JEFF-3.1 [202]	92	84	570
NON	-	-	-1533

Six branching ratios for the IAEA BN-600 Phase 6 neutronic benchmark problem were tested [146]. Δk results using diferent branching ratios are shown in Table C.1. This shows that Δk is sensitive to branching ratio.

C.4 Comparison of Equilibrium routes

ERANOS was used to change the enrichment at each recycling step to keep k_{eff} the same at the beginning and end of each cycle until an equilibrium was reached. Enrichment was changed based on ^{239}Pu equivalence calculations. These were compared to the same simulation but using the final enrichment for every recycling step, allowing k_{eff} to be different for each recycling step. The change in enrichment and k_{eff} for an example case can be seen in Figure C.3. Both methods appear to reach the same equilibrium. Final equilibrium spent fuel masses were also compared in Table C.2 for a whole core irradiated to 100 GWd/t, with results varying by less than 1%.

The final one group cross-sections were also compared in Table 6.12. Showing that both methods were appropriate to get equilibrium reactor cross-sections that would be used n ORION.

C.5 Equilibrium fuel feed vectors

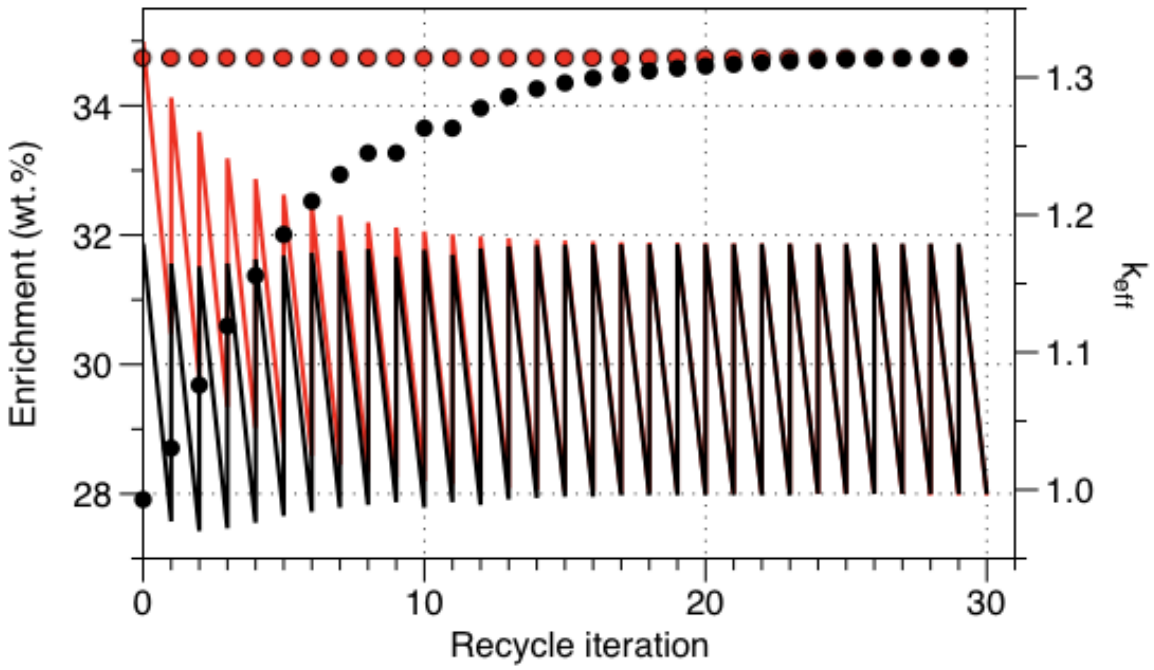

Figure C.3: Constant enrichment method compared to the varied enrichment method of reaching equilibrium. Red shows the constant enrichment method. Black shows the varied enrichment method. Points represent enrichment, lines represent k_{eff}.

Table C.2: Equilibrium cycle k_{eff} and SNF masses for changing enrichment and final enrichment methods.

	Changing Enrichment	**Final Enrichment**	**Difference (%)**
$k_{eff(BOEC)}$	1.179	1.179	0.03
Δk_{eff} (pcm)	15520	15517	-0.02
SNF Mass (kg)			
Pu238	8.598E+00	8.594E+00	0.04
Pu239	1.096E+03	1.097E+03	-0.10
Pu240	1.098E+03	1.098E+03	0.01
Pu241	1.754E+02	1.753E+02	0.03
Pu242	1.749E+02	1.739E+02	0.56
Am241	1.819E+01	1.818E+01	0.03

Table C.3: Equilibrium feed vector for Low-CR SFRs using the UK's plutonium stockpile run to FEED and FULL equilibrium.

FEED	**Zr-low** Pu	**Zr-low** Pu+Am	**Zr-low** TRU	**MOX-low** Pu	**MOX-low** Pu+Am	**MOX-low** TRU
'NP237'	-	-	0.33	-	-	0.26
'PU238'	0.31	2.38	2.51	0.34	2.98	3.08
'PU239'	50.19	46.36	45.44	45.96	41.43	40.25
'PU240'	40.07	35.50	35.19	42.89	37.27	36.98
'PU241'	3.76	3.29	3.25	4.41	3.87	3.84
'PU242'	5.67	6.04	5.89	6.39	6.86	6.69
'AM241'	-	4.62	4.52	-	5.36	5.24
'AM242M'	-	0.33	0.33	-	0.42	0.41
'AM243'	-	1.48	1.44	-	1.81	1.77
'CM242'	-	-	0.00	-	-	0.00
'CM243'	-	-	0.02	-	-	0.04
'CM244'	-	-	0.89	-	-	1.15
'CM245'	-	-	0.20	-	-	0.28
FULL	Pu	Pu+Am	TRU	Pu	Pu+Am	TRU
'NP237'	-	-	0.47	-	-	0.43
'PU238'	0.47	3.15	3.26	0.60	4.28	4.30
'PU239'	38.12	36.64	35.62	30.13	29.04	27.97
'PU240'	43.03	36.07	35.89	46.12	37.12	37.40
'PU241'	4.47	3.74	3.66	5.28	3.97	3.91
'PU242'	13.91	11.74	10.17	17.87	14.47	12.21
'AM241'	-	4.52	4.34	-	5.65	5.44
'AM242M'	-	0.40	0.37	-	0.54	0.51
'AM243'	-	3.75	3.19	-	4.94	4.08
'CM242'	-	-	0.00	-	-	0.00
'CM243'	-	-	0.03	-	-	0.04
'CM244'	-	-	2.40	-	-	2.93
'CM245'	-	-	0.60	-	-	0.78

Table C.4: Equilibrium feed vector for Low-CR SFRs using LWR SNF and run to run to FEED and FULL equilibrium.

	Zr-low	**Zr-low**	**Zr-low**	**MOX-low**	**MOX-low**	**MOX-low**
FEED	Pu	Pu+Am	TRU	Pu	Pu+Am	TRU
'NP237'	-	-	0.37	-	-	0.31
'PU238'	1.12	3.61	3.64	1.22	4.33	4.32
'PU239'	46.58	42.21	41.11	41.91	37.23	35.91
'PU240'	39.88	33.53	33.32	42.72	35.16	35.01
'PU241'	3.67	2.99	2.98	4.36	3.44	3.42
'PU242'	8.76	8.45	8.06	9.79	9.35	8.90
'AM241'	-	6.18	5.92	-	6.98	6.69
'AM242M'	-	0.43	0.41	-	0.53	0.50
'AM243'	-	2.59	2.46	-	2.98	2.82
'CM242'	-	-	0.00	-	-	0.00
'CM243'	-	-	0.03	-	-	0.04
'CM244'	-	-	1.40	-	-	1.67
'CM245'	-	-	0.31	-	-	0.40
FULL	Pu	Pu+Am	TRU	Pu	Pu+Am	TRU
'NP237'	-	-	0.51	-	-	0.43
'PU238'	0.47	3.14	3.29	0.59	4.21	4.24
'PU239'	38.05	36.38	35.36	30.08	28.99	27.93
'PU240'	42.56	35.79	35.80	45.33	36.90	37.31
'PU241'	4.40	3.53	3.47	5.17	3.96	3.92
'PU242'	14.52	12.27	10.51	18.84	14.77	12.30
'AM241'	-	4.56	4.41	-	5.57	5.40
'AM242M'	-	0.40	0.38	-	0.53	0.50
'AM243'	-	3.94	3.30	-	5.07	4.13
'CM242'	-	-	0.00	-	-	0.00
'CM243'	-	-	0.03	-	-	0.04
'CM244'	-	-	2.36	-	-	3.00
'CM245'	-	-	0.58	-	-	0.80

Appendix D

Proliferation Resistance of Fast Reactor Fuel Cycles

Presented to the 2012 UK Project on Nuclear Issues (PONI) annual conference, "Nuclear Stability: From the Cuban Crisis to the Energy Crisis" [203].

D.1 Introduction

The purpose of this paper is to review previous studies looking at the proliferation resistance (PR) of fast reactor (FR) fuel cycles, and how this pertains to the future use of the UK's plutonium stockpile.

A lot of attention has been given to the future of the UK's plutonium and special nuclear material. Ideas have focused on; disposal, use of plutonium and uranium in mixed oxide (MOX) fuel for new build reactors, storage for future use in FRs, and FR burning [16]. FRs have the potential to utilise the UK's material in different ways: irradiating to high burnup (large number of fuel atoms undergoing fission), which is currently being considered with GE-Hitachi's PRISM reactor; and the sustainable use of resources by breeding as much plutonium as is burnt in a reactor, as was originally planned for the UK's reprocessing program [2, 11]. There has been little discussion about the different proliferation risks between FRs for plutonium disposition and FRs for sustainable resource use, for a country with large civil plutonium stocks. FR systems are especially interesting in this area as they have inherent issues regarding PR, due to fuel requiring a higher proportion of fissile material than current reactors,

and plutonium from the breeding process being ideal for weapons-use [11].

Whether methods of increasing barriers to proliferation in the fuel cycle are adequate tends to depend upon the state using the system and the weighting given to these barriers by people assessing it [204, 205]. Generally, a diversion-resistant fuel cycle in the UK is of little concern. The UK is already a nuclear weapons state and a signatory of the non-proliferation treaty so unlikely to need to divert civilian material for military purposes. Non-state threats have not caused concern in the past so one might argue that the fuel cycle only needs to be as PR, or better, than present. However, it is hard to justify a fuel cycle system that is deemed acceptable for a nuclear weapons state but not for 'insecure,' or 'prestige-seeking,' non-weapons states, when all nations have the right to the peaceful use of nuclear energy [206]. This can cause international tension and removes the ability of the UK to export technology and expertise in fuel cycle systems that are not suitable for 'insecure,' or 'prestige-seeking' states. Therefore, anything used in the UK must have high PR and be suitable for other states to use. In short, our nuclear program should set an example to other states developing their own. Otherwise, denying another state the chance to develope a fuel cycle similar to ours on grounds of proliferation would be both difficult and hypocritical. An historical example is that of India, where India got details of the PUREX process and used it to separate weapons material from spent fuel [207].

This paper will focus on sodium-cooled FRs and the intrinsic proliferation barriers of their associated fuel cycles. General proliferation issues and barriers to proliferation will be outlined before explaining how these may or may not be relevant to different states. A qualitative assessment and discussion of different fuel cycle systems will be done, considering how worthwhile they might be. Finally, the influence of intrinsic barriers on other fuel cycle aspects will be looked at, showing areas where future research will take place to assess the impact on sustainability, waste toxicity, cost, socio- economic aspects and technology readiness.

D.2 Proliferation aspects of FR systems

D.2.1 Types of proliferator

Increasing PR can be very effective in preventing diversion, but it depends on the capabilities of the threat to PR [208]. The Generation IV International Forum (GIF) groups proliferators into three sub-groups: [204, 208, 209]

- Sophisticated state – A state with a well-developed nuclear infrastructure.
- Unsophisticated state – A state without a fully-developed nuclear program.
- Sub-national/Non-state threat – Mainly concerns physical protection measures to impede theft or sabotage. Intrinsic barriers also factor in, for example, having material that is in a form unsuitable for dispersion devices or requires processing to produce weapons usable material.

The issue of physical protection will not be addressed here as it is not generally considered to be inherent to fuel cycle technology options.

D.2.2 Routes to proliferation

There are different ways that sensitive materials can be acquired. The GIF and previous studies have shown that barriers to proliferation have different weightings depending on the proliferation route a state takes [204, 208]:

- Dedicated facilities – Building a specific facility to produce weapons material, removing the need to use civil facilities. It is largely ignored here as this paper focuses on civilian fuel cycle facilities.
- Overt – Misuse of a civil facility or diversion of declared materials with no attempt to hide it. In this case it is assumed that only a sophisticated state has the infrastructure and technical ability to overtly acquire material [208, 209].
- Covert – Concealed diversion of material or use of a civil facility to produce weapons material. It is generally described as a route that both sophisticated and unsophisticated states can take. However, the time taken to produce weapons

material will depend on whether the proliferator is a sophisticated of unsophisticated state.

It is assumed that there are four different threats that barriers must be effective against; sophisticated-overt, sophisticated-covert, unsophisticated-covert and non-state [208, 210]. These threats are described below in terms of the effectiveness of different barriers to proliferation.

D.2.3 Types of barriers

There are no barriers that can be put in place to make a fuel cycle proliferation-proof. However, they can make proliferation harder and minimise the incentive to use pre-built civil facilities. Barriers can be grouped into intrinsic and extrinsic barriers as described by the GIF in [204] and summarised below. These subgroups have different weightings depending on the threat [208, 209, 210]:

- Intrinsic barriers are the inherent design features of a fuel cycle and its facilities. They are generally grouped into:
  - Technical difficulty – Inherent difficulty, such as radiological hazard, requiring equipment or information that is not openly available. Sophisticated states have the infrastructure and resources to deal with this barrier.
  - Cost – Associated cost of staffing and running a facility to overcome barriers. Ideally costs to develop a weapon would be higher than a military budget or a dedicated facility.
  - Time – Minimum time to overcome barriers and recover a significant quantity of material. This can be limited by the time it takes to detect diversion (described below) and how easily equipment can be modified.
  - Material – How useable a material is for weapons and how much processing is required. This includes the properties of a material which effect critical mass, heat generation and gamma radiation.
- Extrinsic barriers are institutional barriers such as physical barriers, inspections, treaties and policies, grouped into:

  - Detection probability – Probability that measures such as surveillance and inspections identify a proliferation route being taken through operational anomalies. Influenced by the time it takes to divert a significant quantity of material, described above.
  - Detection efficiency – How efficiently staff, money and equipment are used to apply inspections and surveillance to check for operating anomalies.
  - International politics – Agreements, treaties and policies to make proliferating unfavourable and costly for a state.

Extrinsic barriers are very important but, for the most part, ignored in this work as it focuses on fuel cycle systems and intrinsic barriers which are inherent to the fuel cycle. However, it is important to bear extrinsic barriers in mind when considering intrinsic factors and how they are influenced.

To summarise proliferation aspects relevant to this work - depending on the threat, intrinsic barriers are weighted differently. The weighting depends on the type of state and proliferation route: [208]

- Sophisticated-Overt – When a state wishes to overtly misuse a facility the most significant intrinsic barriers influence the quantity of material required, its quality, and the time it takes to produce it. An example of which would be the ease in which a facility can be modified to make suitable material. Detection is less important as there is no attempt to hide proliferation and technical barriers have little impact as expertise and resources are available due to a states developed nuclear infrastructure.

- Sophisticated-Covert – This route has similar dependence on the barriers mentioned above, but with detection given significantly more weight as diversion is being hidden.

- Unsophisticated-Covert – When a state has an undeveloped fuel cycle and lacks technical expertise only covert routes can be taken and intrinsic barriers are very important with technical barriers being most significant.

Numerous studies have attempted to quantify PR and evaluate different fuel cycle systems such as the International Project on Innovative Nuclear Reactors and Fuel

Cycles (INPRO) and GIF methods, the 'Technology Opportunities for Increasing the Proliferation Resistance of Global Civilian Nuclear Power Systems' (TOPS) study and many more [204, 208, 208]. However PR is heavily dependent on the weighting a person or group gives to different aspects. Also, it can be assumed that the above barriers become less significant when a state has the intention and capability to building dedicated facilities [209]. At this stage the only real barrier is the ability of the international community to detect and find dedicated facilities being built or operated.

D.3 Discussion

When considering FR fuel cycles in the UK, there are several options which can be categorised into:

- Once through burn – Currently considered in the UK with the PRISM reactor, involving no reprocessing of FR fuel and the disposal of current plutonium though high burnup in a reactor [2]. This can all be done on site and is the least efficient use of resources but the most proliferation resistant method described.

- Supporting the back end of new build fuel cycle – Extend reprocessing to new build reactors and utilise plutonium in FRs. The same as the PRISM concept but continued for all new build reactors by having a reprocessing capability. If the PRISM route was to be used in another state it would need a reprocessing facility for thermal reactor fuel. This is advantageous as it is a more efficient use of resources than above and with no FR reprocessing, the plutonium is always unfavourable for weapons and it is unlikely that reprocessing facilities will be able to handle FR fuel. However, a state will still retain the capability of producing weapons grade plutonium via FR breeding and the reprocessing technology to extract it.

- Closed FR fuel cycle – Reprocessing of FR fuel with reactors operating to produce as much fissile material as they use [11]. This would start from one of the above systems and eventually equilibrate in a closed FR system. It is the least proliferative resistant method described because FRs produce plutonium ideal for weapons and the FC requires the separation of plutonium [27].

It is worth considering that, in the UK, plutonium could be made more intrinsically safe by irradiating it as MOX in new build reactors and storing for future use [121]. This is a less economical way of using resources but eliminates the concern of storing separated plutonium in the medium-term. However, for the purpose of this study it is irrelevant where the initial driver fuel comes from, as it is the PR of FR fuel cycles that is being considered.

Each of the above fuel cycle options can take advantage of different intrinsic barriers, depending on the technology used. These take into account reactor design, fuel cycle facilities and transport as these issues influence intrinsic PR. However, improving these barriers could result in negative aspects, such as: increased costs and requirement for extensive research and development. A few important examples of issues which influence intrinsic barriers are given below:

- Fuel material – Plutonium in metallic form is less proliferation resistant as fissile material needs to be in a metallic form for weapons. The alternative is ceramic fuel which has more commercial experience, but minor downsides in terms of fuel efficiency [204]. In the UK plutonium is stored as an oxide, so any metallic fuel will involve a step to reduce plutonium into a metal, something which is not preferable.

- Reactor size – Small modular reactors can improve intrinsic barriers as they involve less material on a site [211]. However, there are cost implications due to economies of scale.

- Transport – Transporting fuel that is not self-shielding has inherent risks as it does not require sophisticated facilities to handle it. To make it self-shielding fuel can be homogeneously reprocessed (see below) with some highly radioactive, short-lived fission products included to make the dose more than 1 mSv at one meter away (feature of self-shielding) [212]. Alternatively, fuel can be pre-irradiated before transport.

- Reprocessing techniques – Currently the aqueous PUREX process is the most common reprocessing technology but involves an isolated stream of plutonium which is a proliferation concern. Modified versions of PUREX incorporate minor actinides (elements with higher mass than plutonium that make up the majority

of long-lived waste radiotoxicity) and do not require separate plutonium stream. However these modifications require significantly technical development. Aqueous methods also require long cooling times so a large stock of used fuel is built up. Alternatively, pyroprocessing methods could be used to homogeneously recycle plutonium with minor actinides. Pyroprocessing requires much shorter cooling times and has been considered for on-site reprocessing in schemes such as those for the BOR-60 and the Integral Fast Reactor (IFR) [11]. Generally, homogeneously reprocessing plutonium with minor actinides causes problems; it requires more shielding for facilities and generates more hazards during maintenance, increasing costs.

- On-site or centralised FC facilities – Centralised fuel cycle facilities will contain significant quantities of material (so the time needed to divert a high mass of material is small) and requires the transport of sensitive materials to centralised facilities [208]. Fuel cycle facilities have smaller stockpiles of material and require minimal transport off site. This has a high intrinsic PR but does not take advantage of economies of scale, so can be expensive.

A few of the technologies mentioned above, such as pyroprocessing, have only been tested in laboratory or demonstration scale plants [27]. Developing this technology for commercial use would be very costly, rendering the cost-benefit balance of improving these intrinsic barriers unclear. However, subjective considerations will also be important based on national perceptions. There are other advantages such as waste minimisation through the burning of minor actinides (which make up the majority of long-lived waste) and the possibility of the same fuel cycle technology being exported to non-nuclear weapons states.

In the long-run it is likely that, if global Uranium stocks diminish significantly, fuel cycles will move towards being closed, requiring reprocessing technology. The existence of this and FRs, even if designed for burning, has the potential to be used overtly to produce weapons-grade material. So even the most proliferation resistant FR fuel cycle (once-through burn (PRISM)) has proliferation concerns despite the lack of FR fuel reprocessing. In general, the reprocessing stage, which closes the fuel cycle, generates the greatest proliferation concern so intrinsic barriers influencing

reprocessing are very significant. Two fuel cycle designs that differ in this respect were considered for Superphenix/EFR type reactors and that of the IFR [11]. The former has an isolated plutonium stream from PUREX reprocessing and large centralised facilities involving transport and stockpiles of material. The IFR route does not involve separated plutonium due to pyro-reprocessing methods and uses small, on-site facilities to minimise transport and stockpiles of material. Fuel cycle designs, like that of the IFR, are thought to be one of the most intrinsically proliferation resistant, ticking a lot of the boxes mentioned above [205]. Despite these very effective intrinsic barriers, studies have come to the conclusion that the IFR is not proliferation resistant enough for an 'insecure' state [205]. No fuel cycle can be completely proliferation resistant and, if determined, a sophisticated state with a FR fuel cycle could use facilities overtly to acquire material. So, are increased intrinsic barriers like those of the IFR fuel cycle worth the extra operation and development costs? Especially when considering that intrinsic barriers only reduce the incentive for a state to use civil facilities. If a state is determined to acquire a weapon this will push them towards developing dedicated facilities and, over time, most states can develop technical skills and resources to build dedicated facilities.

D.3.1 In the UK

In the UK state-state proliferation is unlikely and internal proliferation is not a concern as the UK is already a weapons state and has tons of separated plutonium stockpiled. THORP, and previous reprocessing facilities, produce isolated plutonium streams and large masses of sensitive material are stored within facilities - all things that should be avoided [16]. However, the UK has never had a proliferation issue with fissile material since signing the NPT.

Should future fuel cycles in the UK aim to be as proliferation resistant as possible, or just more proliferation resistant than at present?

The easiest way to ensure high intrinsic barriers to proliferation would be to develop FRs and associated fuel cycle facilities through government funded projects. Alternatively, if private industry is to provide the technology, high development costs will more than likely lead to the development of more commercially experienced technology with lower intrinsic barriers (e.g. PUREX reprocessing and centralised facilities).

To get around this there would have to be a large market for exporting the technology and expertise to other nations. Alternatively, government incentives could be used or legislation about the PR of facilities in the UK.

Even if a future fuel cycle with high intrinsic PR is provided through government funded projects, the cost-benefit balance could be unfavourable as initial development and high operating costs could render nuclear power very uneconomical. This is unless the technology has other benefits that are important to the UK such as the repository size or long-term radiotoxicity of waste. In terms of proliferation alone, the benefit to the UK is minimal and the potential for the technology to prevent other users from proliferating is low if, over time, states develop a sophisticated nuclear infrastructure.

D.4 Summary

It is not possible to make a fuel cycle proliferation proof, particularly when considering intrinsic barriers alone. Unsophisticated states, over time, will become more sophisticated and have the potential to overtly use facilities and develop the technical ability to build dedicated facilities. An implication of this is the possibility that the route taken by the UK is of little concern.

In general, it seems that developing a fuel cycle with greater intrinsic barriers is likely to cost more, have more technical issues, and cannot be provided by private industry alone. It may not be suitable to export this technology, even with high intrinsic and extrinsic barriers, to states seen as insecure or a threat, as there is the potential for states to develop the capability to overtly misuse facilities. Alternatively, more commercially developed technology could be used which is less intrinsically resistant. As states develop more advanced nuclear infrastructures, the relative PR between less- developed, more-intrinsically resistant technology and more-developed, less-intrinsically resistant technology will be small. Therefore, in the long-term, the cost-benefit balance may come down in favour of less proliferation resistant technology due to the risk of making nuclear power uneconomical.

D.4.1 Further Work

Facilities with high intrinsic barriers might have other potential advantages such as reducing a repository size or the lifetime of radioactive waste through burning minor actinides in homogeneously reprocessed fuel. So further work will involve looking more specifically at other fuel cycle aspects. Depending on the weighting given to these aspects, there could be advantages to developing less commercially ready technology.