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Abstract

The essential processes in the out-of-pile nuclear fuel cycle are described, i.e. mining and milling of uranium ores, enrichment, fuel fabrication, storage, transportation, reprocessing of irradiated fuel, waste treatment and waste disposal. The aspects of radiation (mainly gammas and neutrons) and of heat production, as well as special safety considerations are outlined with respect to their potential operational impacts and long-term hazards. In this context the importance of nuclear data for the out-of-pile fuel cycle is discussed. Special weight is given to the LWR fuel cycle including recycling; the differences of LMFBR high burn-up fuel with large PuO_2 content are described. The HTR fuel cycle is discussed briefly as well as some alternative fuel cycle concepts.

Handhabung von Kernbrennstoffen und deren Wiederaufarbeitung einschließlich Abfallbehandlung mit Diskussion zugehöriger Aspekte nuklearer Daten

Zusammenfassung

Es werden die wesentlichen Prozesse im nuklearen Brennstoffkreislauf, nämlich Uranerzgewinnung und Verarbeitung, Anreicherung, Brennstoffherstellung, Zwischenlagerung, Transport und Wiederaufarbeitung bestrahlten Brennstoffs, Abfallbehandlung und -lagerung beschrieben. Radioaktive Strahlung (hauptsächlich Gamma- und Neutronenstrahlung), Wärmefreisetzung und spezielle Sicherheitsaspekte werden im Hinblick auf ihre mögliche Einwirkung auf Prozeßführungen und hinsichtlich ihrer Langzeitwirkung umrissen. In diesem Zusammenhang wird die Bedeutung nuklearer Daten für den Brennstoffkreislauf diskutiert. Besonderes Gewicht liegt dabei auf dem Brennstoffkreislauf von Leichtwasserreaktoren einschließlich Plutoniumrückführung. Die Unterschiede hoch abgebrannten Brennstoffs aus natriumgekühlten schnellen Reaktoren mit hohem PuO_2-Gehalt werden beschrieben. Der Brennstoffkreislauf des Hochtemperaturreaktors sowie einige alternative Brennstoffkreislaufkonzepte werden angesprochen.

1. Introduction

The nuclear fuel cycle constitutes the entire range of processes to which the fuel is subjected from ore mining to terminal storage of the radioactive waste in geological formations. The large amount of plutonium in the increased number of operating thermal power reactors and the development of a fast reactor technology with already two operating prototype reactors in Western-Europe require a well developed fuel cycle industry, especially for the reprocessing of the spent fuel, refabrication of the recycled fuel and waste disposal, areas which some years ago were considered to be of minor importance compared to reactor industry. In Germany, the design of a large scale reprocessing plant for LWR fuel of about 1500 t/yr through-put has been completed and is awaiting the licensing procedures. With the increasing plutonium amount another aspect has gained strong interest recently, i.e. safeguarding fissile material in order to prevent or at least reduce the possibility to divert fissile material from the fuel cycle for weapons fabrication. This aspect has started a world-wide effort to investigate the possibility of a fuel which is inherently safe against diversion (alternative fuel cycles) and, in parallel, has led to a narrowing of the requirements for reliable and timely detection of fissile material diversion.

These aspects form a background which requires a re-investigation of the physics aspects of the nuclear fuel cycle. This paper deals with the out-of-pile stages of the fuel cycle, with the processes involved, the present problems and the related nuclear data aspects. Because this conference is aimed at assessing the needs and status of nuclear data for reactors and other applied purposes, the more stringent conditions, imposed on fuel cycle aspects, necessitate to check whether new nuclear data requests have to be formulated although it has been indicated that the out-of-pile processes are not very sensitive to data uncertainties. An appreciation of any data request, and this is true also for reactor conditions, can be made only if a balanced consideration of the nuclear and non-nuclear aspects of the processes under investigation is performed in order to find out whether improved knowledge of nuclear data can help to decrease actual and potential difficulties or conservatism in the plant design. In the out-of-pile cycle, besides nuclear processes, fuel handling and chemical problems have to be discussed to that extent which is necessary to give meaningful data requirements. It is obvious that in the out-of-pile processes mainly the decay data of nuclei as half-lives, heat production, emission of α,β,γ-radiation as well as the fission product yields and the productions of neutrons via spontaneous fission and (α,n)-reactions play the dominant rôle. The reaction cross sections such as neutron fission and capture are important only during the reactor residence time of the fuel to predict the proper concentrations of radioactive nuclei, and in investigating criticality control of out-of-pile fuel.

In Fig.1 a simplified flow diagram of the fuel cycle is given. We will follow the various stages with main emphasis on the uranium/plutonium cycle of LWRs including recycling. The differences and the problems of the fuel cycles for the advanced reactors such as LMFBR and HTR with thorium as fertile material are discussed along with some alternate fuel cycle concepts.

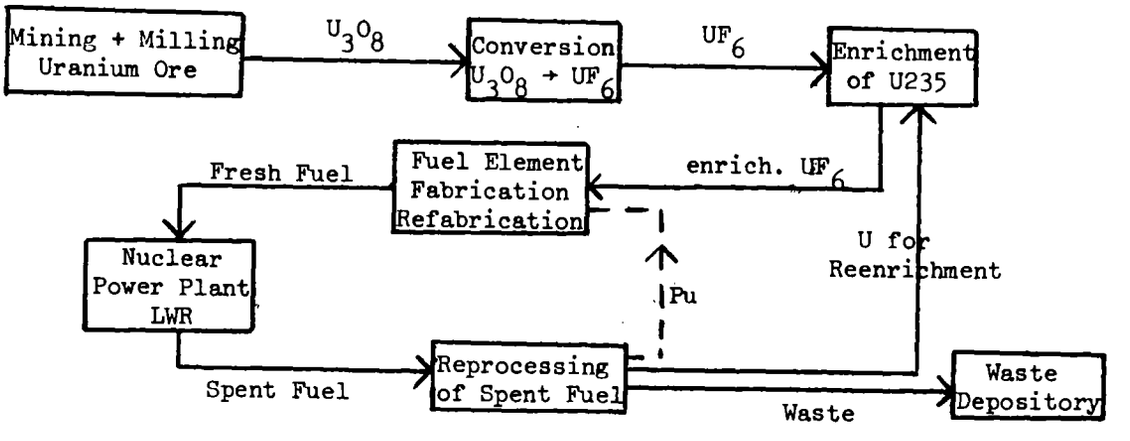

Fig.1 : Nuclear Fuel Cycle for LWRs

2. The Route of Unirradiated Fuel from Mining to Fabrication.

2.1. Mining and Milling

The problem of mining and milling uranium ore is connected with the huge amount of the waste produced in these processes. The waste originates from the removal of the waste rock to provide access to the ore body. Substantial amounts of contaminants are generally released from the waste rock piles only when they contain more than 1% sulphide mineral causing bacterial oxidation. Because uranium is extracted from the ore either by acid or alkali leaching, the list of the pollutants includes heavy metals, nitrate, phosphate, acidity and alkalinity as well as radioactive materials, namely the α-decay daughters of U238, i.e. Th230, Ra226 and Rn222. They can appear as contaminants in the waste water, seepage from the waste rock piles and from the mill tailings, contaminating finally the receiving ground and surface water. Radioactive air pollution is caused by uranium dust and Rn222. The consequences of the airborne releases are usually small, but the waterborne releases after many years of mine operation, dependent on location, may require treatment of the waste because of Ra226 activity ($t_{1/2}$ = 1602 yr) /3/. As indicated by Cohen /4/, the potential ingestion hazard of mill tailings formed to produce fuel for a certain number of 1000 MWe reactors exceeds the hazards of the waste coming from these reactors only after a period of about 250 yr. One should note that the reactor waste is much more securily stored than mill tailings (see section 4.2.3). The environmental impact of mining and milling uranium ore cannot be influenced or reduced by a better knowledge of the decay rates and radiation intensities of U238 and its decay daughters to Pb206.

2.2 Conversion of U_3O_8 to UF_6 and Enrichment of Fissile U235

The problem in the conversion process is connected with the corrosion of the components, because after the reduction of U_3O_8 to $U(IV)O_2$ with hydrogen, HF and F_2 are used for hydrofluorination to UF_4 and fluorination to UF_6. The gaseous waste contains large amounts of SO_2 and NO_x, a small amount of radioactivity (Ra226) is found in the liquid waste. Because of the high requirement of electrical energy in enrichments plants due to the low efficiency of the single enrichment steps eventually a large amount of waste heat is produced, which has to be dissipated to a river or as humidified air from a cooling tower. For a gaseous diffusion plant similar amounts of gaseous effluents SO_2 and NO_x are released as in the conversion process. As a nuclear aspect, for highly enriched UF_6 criticality has to be controlled. This is achieved by a suitable geometrical design. More accurate nuclear data are not requested, and as in all protective measures, safety margins are applied.

2.3. Fabrication of Unirradiated Uranium Oxide

The fabrication of UO_2 does not pose any problem due to the low radioactivity of U235 (α, spontaneous fission neutrons). Criticality control is assured by safe geometrical configurations. Environmental impacts arise from the chemical effluents (fluorine and nitrogen compounds) in the conversion process from UF_6 to UO_2 (e.g. by reaction of UF_6 and NH_3 + CO_2).

3. General Reactor Physics Aspects for Out-of-Pile Investigations

The description of the burn-up behavior during reactor operation including fuel management has been well developed. In general, this requires an adequate solution for criticality, reaction rate and flux distributions as well as for the neutron spectrum, the reactivity worth of control rods or blades, a proper treat-

ment of heterogeneity etc. The changes of the absorber rod positions, neutron spectrum and the related changes of the effective cross sections (due to spectral changes, nuclide concentrations, resonance selfshielding) during burn-up have to be taken into account, keeping k_{eff} unity during the evolution of reactor life. This relatively complicated procedure provides a reliable nuclide concentration at fuel discharge of each subassembly, provided the nuclear data used are accurate. For the out-of-pile behavior of the discharged fuel, this unloaded "nuclide vector" determines the amount of e.g. the inventory of radioactivity at any time after discharge assuming the decay rates to be known. However, for out-of-pile purposes the nuclide inventory and the deduced quantities need not to be known exactly for each space point in the reactor. Fuel bundles of different burn-up are mixed in the storage pond and in the dissolver tank. Therefore, only average nuclide concentrations for an unloaded fuel batch are needed to determine heating and radiation. But both these quantities originate from many radioactive fission products, structural material and heavy elements, most of those are usually not incorporated in the burn-up calculations. Therefore often one-energy-group fundamental mode calculations with all the isotopes of interest are applied also for the in-core description of build-up and decay of nuclides, neglecting the time dependence of effective cross sections, which is different in various zones of the reactor core (e.g. those in control-rod regions in the upper core compared to those in the control-rod follower region) or the different time evolution of core and blanket characteristics for fast reactors (it should be noted that the cell heterogeneity is taken into account by a proper definition of the effective cross sections). These simplified methods (e.g. /5/, /6/) can only be used as a guide-line for the calculation of the properties of spent fuel. In a licensing procedure, a pragmatic combination of accurate physics methods with the "zero dimensional irradiation programs" for the aspects in out-of-pile processes is required. Only if a sophisticated use of the simplified, but easily manageable fundamental mode codes to obtain equivalent results compared to those from calculations in a higher dimension is made (which is not possible in all cases and requires special attention in all recycling concepts), the application to calculate the in-core physics with these practical tools is justified.

With respect to the nuclear data which are necessary to describe the long-term behavior of irradiated fuel, all those reaction cross sections, fission product yields and spontaneous fission neutron yields, which lead to a radioactive nuclide and neutron radiation, respectively, of concern in the out-of-pile stages of the fuel cycle, are important.

In the Tables I-XIII the top ten nuclides at various times after discharge and in the waste are listed with respect to the production of thermal power, radioactivity, neutron production via spontaneous fission and (α,n)-reactions. In addition, the main gamma radiating nuclei are presented. Because at present the LWR fuel cycle is of primary importance, the numbers in the tables correspond to a 1000 MWe PWR, reaching 33000 MWd/t burn-up during 3 yr operating time; 1% Pu and U losses, appearing in the waste, are assumed in the reprocessing process. Because reprocessing on large scale may be delayed, cooling times after discharge up to 10 yr are considered, e.g. in interim storage ponds. As far as the accuracy of the nuclear data in question and the methods applied is concerned, many integral experiments have been performed and evaluated. These experiments relate to the in-core production of uranium and transuranium isotopes as well as of some fission products, see, also for additional references /7,8,9/. Because the data requests for the main isotopes present in the operating reactor have been clearly established for the prediction of reliable core performance, among the heavy elements of the U/Pu cycle mainly the in-core production rates of Am- and Cm-isotopes for out-of-pile investigations are to be considered here. At the advisory group meeting on transactinium isotope nuclear data in 1975 the requests for these data have been formulated /10/; they remain unchanged at present. To meet these requirements, an international working group has been established by the IAEA in 1977; work is in progress. As an example, the difference in the average capture cross section of Am^{241} by about a factor of two in fast reactors seems to be almost resolved, see also /11/.

The most striking success in the last years has been obtained in calculating reliably the shut-down decay heat from fission products in LWRs. Because the investigations cover a range up to 30 yr, the results are essential also for the out-of-pile heating problems and therefore should be regarded with special attention

here. According to /12;13/ the present uncertainty of the shut-down decay heat prediction of a LWR reactor is about ± 20% at 1 sec after shut-down decreasing to about 2-4% after 10 sec up to more than ten years. The initial uncertainty is not of high importance, because the cooling capability of a LWR in an emergency loss-of-coolant accident is not reduced to zero immediately after the break of a main primary coolant pipe. What is essential, is that the emergency core-cooling is fully operating at least before about one minute after initiation of a blow-down to prevent larger parts of the core from melting. Therefore, the presently achieved accuracy for decay-heat predictions in LWRs is sufficient. For fast reactors, a similar accuracy is not reached at present. The requests are between 5% and 10% for cooling times up to about 1 yr after shut down, relaxing to about 15% thereafter /13/. Due to the excellent cooling capabilities of sodium by natural convection in a loss-of-flow accident, the decay heat can be removed without difficulty once the reactor has been shut down successfully. If this action fails, then with subsequent voiding of the coolant a power excursion is initiated and decay heat is then of no importance.

For spent-fuel handling of fast reactor subassemblies (e.g. transportation, interim storage, waste packaging) an accuracy of about 10% would be desirable. The main aspects of beta- and gamma-heating from the spent fuel after discharge from the reactor in the various processing steps in order to avoid unreasonable data requirements, are discussed in the subsequent chapters. The questions arising from shipment of spent fuel or from waste are investigated after a discussion of reprocessing, refabrication and waste packaging because the corresponding problems at all these stages of the fuel cycle are similar.

As a concluding remark to this chapter, the application of the simple methods to predict the in-core and out-of-pile behavior of fuel and waste require (a) the check to more sophisticated reactor physics methods for the in-core description, (b) updating of the data libraries according to recent improvements, and (c), checking of these tools on measurements, e.g. by post-irradiation examinations of irradiated fuel pins and by mass-balance determination in the head-end step of the reprocessing stage. Unsatisfactory disagreement between theoretical and experimental results then may be removed by proper adjustment procedures until better data information is made available. Some of this information will not be declassified for commercial reasons.

4. The Out-of-Pile Processes for Irradiated Fuel from LWRs

Because the LWR fuel cycle is of primary importance at present, the various steps in handling and further processing of LWR fuel are presented in this chapter and the relations to nuclear data requirements are discussed in the context of process-procedures and process-improvements.

4.1. Interim Storage of Spent LWR-Fuel

In the next years it will be necessary to safely store larger amounts of the burnt fuel in water ponds, before reprocessing of fuel with high burn-up will be available on a large scale. Three main problems have to be solved under licensing conditions: (a) protection against radiation (neutrons and gammas), (b) providing cooling equipments to remove the decay heat, (c) to maintain subcriticality of the stored fuel under all circumstances. In the Tables I-XII the nuclides which are of importance for various cooling times after discharge, together with the corresponding effects of interest are listed. The storage ponds under investigation consist of compact cells of steel boxes containing about 1 ^{w}/o natural boron, into which a spent-fuel element is inserted; then the boxes are closed by a lid. The introduction of heterogeneous neutron absorbers allows (for criticality reasons) a reduction of the distance between neighboring fuel elements, which is important for economical reasons.

Boxes and pond water guarantee protection against radiation. Though the cooling conditions are rather weak compared to the in-pile situation, an overprediction of the actual thermal power requires e.g. larger box distances which again leads to higher costs. According to the discussion of shut-down decay heat prediction in

in chapter 3, no further data requirements are needed for this aspect. Differences in the results of decay-power may result from not up-dated libraries or from the application of different methods (e.g. simplified versus sophisticsted codes).

For criticality control in this simple type of storage ponds no further data requests arise, because they are covered by the requests for the in-pile reactor data if no exotic absorber materials are used (see also sections 4.2.4 and 4.4). As far as calculational methods are concerned, there should not arise a problem either. However, the configuration of the fuel elements in the steel boxes is different from any in-pile fuel element arrangement, and therefore the application of the usual recator codes for the super-cell configuration has to be verified, e.g. by Monte Carlo /14/ and/or other neutron transport theory models. As in all cases of protecting a facility, the operating personnel and the plant environment, additional safety-factors are applied to ensure that the radiation dose received by an individual is well below the permissible limit.

4.2. Reprocessing of Spent LWR-Fuel

From a 1000 MWe PWR, annually about 30 t of spent fuel with an average burn-up of 33000 MWd/t are discharged. After storage on the reactor site for some months the fuel contains about 280 kg of plutonium, ~15 kg of neptunium, ~4 kg of americium, ~1 kg of curium and ~1 t of fission products. These fission products are constituted by (guiding figures only) 17% noble gases and halogens (Kr, Xe, J), 32% rare-earth elements (Y, La, Ce, Pr, Nd, Sm, Eu, Gd), 15% alkaline and alkaline-earth metals (Rb, Cs, Sr, Ba), 11% of metals as Pd, Rh, Ru, 23% of transition elements (Zr, Nb, Mo, Tc) and about 2% of Ag, Sb, Te. Besides the release of C^{14}, as a special fission and reaction product tritium is produced.

The objective of reprocessing the spent fuel after cooling times up to three years is to recover uranium and plutonium to such a degree of decontamination that the residual activity of uranium approximately equals that of ore-made fuel (decontamination factors ≈ 10 to 10^7). This objective can be met by the chemical extraction process PUREX (plutonium and uranium recovery by extraction). Solution and extraction techniques are used since a long time in chemical industry, and the PUREX process is accepted as reliable also in large scale technology.

Reprocessing is performed in hot-cell bunkers with concrete walls of about 2 m thickness to ensure safe enclosure of the large amount of radioactivity and to attenuate radiation.

As in previous chapters we will describe the essential steps in the reprocessing process in order to obtain a better perspective of the importance of related nuclear data.

In Fig.2 a simplified flowsheet for reprocessing spent fuel is given.

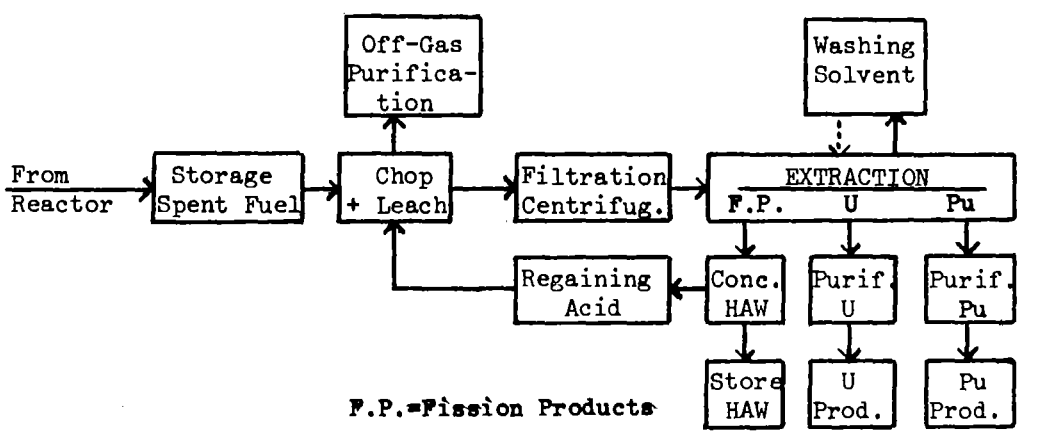

Fig. 2: Chemical Reprocessing of LWR Fuel

4.2.1. The Head-End of Reprocessing

The spent fuel elements are taken from the storage and are chopped, either single pins after disassembly or the bundle as a whole. From the 2-5 cm pieces the fuel is leached in boiling nitric acid. In this chop-and-leach-process about 64 isotopes of gaseous and volatile elements are released. The most important radionuclides are (the half-life is given in brackets): Kr85 (10.76 yr), H3 (12.45 yr), I129 ($1.57 \cdot 10^7$ yr), C14 (5736 yr). If the fuel had been stored longer than about 150 days, most of the active Xe131m (12d) and I131 (8.04d) is already removed from the fuel. Together with the gaseous effluents Pu, Am and Cm aerosols are released.

Kr85 is formed as a primary fission product and via the short-lived (seconds and minutes) precursors of mass number 85. Its hazard is mainly determined by the 151 keV gamma rays and also by betas in near-ground-level releases.

H3 is produced in the reactor mainly by (n,α)-reactions with boron in control rods and with soluble boron in the coolant (for reactivity control in PWRs), furthermore it is formed as a ternary fission product. The tritium probably is retained in the matrix of the control rods, but the tritium in the ceramic fuel escapes partly into the clad material.

The long-lived I129 is formed by its precursors Te129 and Sb129.

C14 is produced in LWRs mainly by (n,p)-reactions with N14 and by (n,α)-reactions with O17. Nitrogen originates from the fuel fabrication process (see section 2.3), with strong regulations not to exceed a certain level.

Hot nitric acid oxidizes to some amount the metallic fission product Ru, which is released as RuO_4 during the dissolution process. We take up this point in the next section. The licensing authorities require retention of gaseous radioactive effluents in reprocessing plants. Therefore the main technological problem associated with gaseous effluents is the development of effective filters for the decontamination of the off-gas. Krypton can be captured by cryogenic destillation, iodine by silver-loaded filters. Tritium can be converted to tritiated water THO. Further development work is necessary for all these methods. As another head-end process, voloxidation was considered. By this process the pins are treated prior to dissolution in an oxygen atmosphere at about 500°C - 700°C to release the gaseous fission products and tritium. This process is no longer considered, because there are indications /15/ that above 650°C insoluble PuO_2 is formed, which would increase Pu losses in the reprocessing stage.

As far as the nuclear data are concerned, good knowledge of the inventory of the gaseous and volatile effluents is necessary for the design of effective filters. The present accuracy in fission product yields is sufficient when compared to the uncertainty in the retention efficiency of filters. For tritium, the ternary fission yield has been re-measured and a value of 0.92/10,000 fissions is found, which compares with earlier values ranging from 0.5 to 1.08 (mean 0.93) /16/. The difficulty to isolate tritium on its way from the in-pile formation to the head-end process is definitely large. (n,p)- and (n,α)-reactions are not very well known from the nuclear physics point of view, but the uncertainties in these reaction cross sections are much less than e.g. those in the amount of nitrogen which remains in the fuel from fabrication.

4.2.2. Insoluble Fission Product Residues

The undissolved hulls are transported in a cascet to an interim storage, either under water or in a dry storage bunker. They are then treated as MAW waste (see section 4.3.2). The hulls are monitored especially for undissolved fissile material by the activity of Pr144 which is a daughter of Ce144 (284d).

In the leach process, fission products and heavy metals are not completely dissolved by HNO_3. It is necessary to treat the feed solution prior to the extraction process with filters or centrifuges to remove the insoluble residues after previous sedimentation. This is done in order (a) to avoid plugging of tubes or contactors in the subsequent process steps, (b) to allow a proper balancing of the homogeneous solution, and (c) to minimize radiolysis by the high active particles

in the feed residue. These residues are treated as waste and therefore heat generation and radiation level have to be known in order that an appropriate method for final repository can be provided (see section 4.3.3).

For 1 t of uranium with high burn-up about 1.5 to 3.5 kg of insoluble residues together with about 0.4 kg of Zr from the chop-process have to be treated. The sediment mainly consists of metallic fission product alloys of Ru, Pd, insoluble Zr and Mo compounds with low U- and Pu-content.

The problems connected with the insoluble residues require a check of the data for radionuclides with half-lives of about 1 yr and of their daughters with short half-lives, emitting beta- and gamma-radiation accompanied by heat production and radiolysis in the extraction process. From this point of view the production of Ru106 (1 yr), as primary fission product and via the beta-emitting precursors Tc, Mo and Nb is of special importance as well as the accompaning mean beta- and gamma-energies of Ru106 and, more important, of the short-lived Rh106 (2.2 h). The specific heat production from Ru106 is about 2 orders of magnitude less than the heat produced by Rh106 (2.2 h), leading to the stable isotope Pd106. Comparing yields, half-lives and the beta- and gamma-decay energies since 1965, as done in Table XIV, it can be seen that large differences in the short-lived isotope data exist. This is of no importance for this problem. But rather good agreement in the data exists for Ru106. The only important difference is indicated in the mean beta and gamma energy of Rh106, yielding about 6% less heat production in the insoluble residues. This is not of any importance for the filtering process and subsequent waste treatment. Furthermore, the uncertainties in the data for Ru are completely shadowed by the amount of insoluble Ru (part of Ru is released as RuO_4, see 4.2.1) or other constituents which form the residues; this is partly dependent on burn-up and the process procedure.

4.2.3. The Extraction Process

In the PUREX process, the aqueous solutions of uranium- and plutonium-nitrate are brought into contact with an organic solvent. The high-valued urania and plutonia are extracted, whereas the fission products, neptunium, americium and curium remain nearly unaffected. As organic solvent, tri-butyl-phosphate (TBP) is used, diluted with kerosene.

For storage times of about 1 yr or more, the amount of U237 (6.75d) and the strong radiation from Nb 95 (35. 1d) and Zr 95 (64d) have decreased and the extraction process can be optimized for Ru separation.

After the first extraction cycle a reducing agent (U(IV), Fe(III), H_2NOH etc.) is added to the U/Pu stream. By these agents the Pu(IV) is reduced to Pu(III), and Pu(III)-nitrate is insoluble in the organic phase and can be separated. In addition, electrolytic reduction has been applied successfully.
Because about 0.1% of the fission products, $\leq$ 2% of Np, Am and Cm remain with the U/Pu stream after the first extraction, the process is repeated two or three times to obtain the required degree of decontamination for uranium and plutonium. The extracted and separated U and Pu are concentrated by evaporating the aqueous constituents. Uranium is stored as nitrate which results from the treatment of the nitrate concentrate with ammonia (NH_3) and carbon dioxide. PuO_2 is precipitated with oxalate from the separated concentrate.

The main objective in the extraction step is related to minimize U- and Pu-losses and to separate radiolysis products from the solvent, because these products may plug the piping and may bind plutonium.

From this discussion it can be concluded that the nuclear data aspect in the extraction process is mainly related to the intensity of the beta- and gamma-radiation from the fission products and to the emission of alpha particles with 5 to 6 MeV from heavy nuclei mainly responsible for radiolysis. The process of radiolysis is not too well known for the special interaction with organic molecules, but an improvement of the existing knowledge of mean radiation intensities could not reduce the radiolysis problems. Another data aspect is discussed in the following section.

4.2.4. Criticality Control in Reprocessing Plants

Due to the high radioactive inventory in a reprocessing plant special safety measures have to be taken against accidents in the plant and against impacts from outside (e.g. earth quakes, airplane crash). All process-cells are bunkered and shielded against release of radioactive material. Special measures are taken against fires, chemical explosions and critical configurations of fissile material in the plant. Here only criticality control is investigated. For spent fuel with low burn-up, the neuralgic point in a reprocessing plant with respect to criticality, is the dissolution of the fuel and the subsequent uranium/plutonium separation. For this step a limitation of the fissile concentration is required. To improve the conditions in the head end and in the first extraction cycle, gadolinium is proposed as a homogeneous neutron poison (1-3 gr Gd/l). In the dissolver tanks and in the extraction columns also heterogeneous neutron absorbers are proposed. As an absorber, which probably can withstand corrosion and which can be fabricated, hafnium is under discussion.

For high burn-up fuel (> 20000 MWd/t), the effect of the neutron absorbing fission products, as seen in Table XIII is so large, that the solutions in nearly all the components of the head end and the first cycle are subcritical. Low burn-up fuel has to be separated from high burn-up fuel and has to be treated separately.

With respect to nuclear data and reactor physics methods, the subcriticality conditions of the solutions in the head end and the first extraction cycle can roughly be regarded under three aspects: fissile material concentration, the source of neutrons available from spontaneous fission and (α,n)-processes, moderation and reflection of neutrons by the aqueous solutions and the concrete walls of the bunkers, respectively. From this it is obvious that because of the complicated geometry situations the methodical questions seem to be more important than the nuclear data uncertainties.

If heterogeneous absorbers are used either in a lattice configuration or in a Pu- or Pu/U-solution, the uncertainty in the prediction of k_{eff} is increased. For k_{eff}, differences of 2 to 5% compared to experimental results are reported /17/. Most of the differences very probably are due to the representation of the complex undermoderated configuration. This is concluded from the fact that in these experiments no exotic heterogeneous absorbers were used, but rather boron, cadmium etc. Homogeneous poisoning with Gd could be treated to a 1% accuracy in k. A brief investigation of the data situation with respect to those absorbers which lead through (n,γ)-reactions to another absorbing isotope, showed that the thermal data and the resonance integrals for Gd and Hf are uncertain to about 5-10%. For europium the situation is worse. It is recommended that the evaluators should check the present accuracy of the data in question for these isotopes, preferentially Gd and Hf. From reactor physics point of view, further integral experiments with heterogeneous absorbers of Gd, Hf and Eu are recommended both for simple and more complex geometries. In addition, the absorption cross sections of the fission products in Table XIII should be checked against latest improvements. As far as the second aspect in criticality control is concerned, the neutron sources from spontaneous fission (Cm244, Cm242) and from (α,n)-reactions are important only when near-criticality in a cell or piping is reached. Usually $k_{eff} \leq 0.95$ must be maintained, and in the cases where sparse experimental information only is available, k_{eff} is kept below 0.9.

Moderation and reflection of neutrons are in these cases not of any concern for the purpose of evaluation of the nuclear data uncertainties. The organic extraction solvent TBP and the kerosene diluent do affect criticality only to a smaller amount compared to water, because the neutrons are mainly moderated by the hydrogen atoms. Concrete- instead of water-reflection reduces the critical diameter: e.g. for a plutonium solution by about 20%.

4.3. Radioactive Waste Management

Radioactive waste originates at all stages of the nuclear fuel cycle. High active waste (HAW) with an activity > 10^4 Ci/m^3 consists predominantly of fission products, non-recovered plutonia and urania, and the transuranium elements neptunium, americium and curium. Their radioactivity requires separation from the biosphere for a long period of time, especially because of the long-lived α-active waste of the heavy elements. Solid medium active waste (MAW) contains e.g. the hulls from

the head-end, filters solid residues from reprocessing and others. Liquid MAW mainly consists of the aqueous raffinates from the extraction process, the destillates from the HAW-concentrates and of tritium. The dominant constituents of MAW are alkaline sodium corbonate and acid nitrate.

Liquid low active waste (LAW) chemically is not very different from natural water. The contamination mainly arises from some dispersed or dissolved fission products. (It is noted that the terminology LAW and MAW is not unique with respect to the associated activity in Ci/m^3).

For LAW and MAW the main objective is to reduce the active volumes and to find a material which can incorporate the waste for safe terminal storage. Though also a reduction of the HAW volume of about 5 m^3 per one 1000 MWe year is desired, the main problem is **arising from the safe confinement** of the long-lived fission products and α-waste. The nuclides which are important in HAW, are listed in Tables XV-XX.

4.3.1. Treatment of LAW and MAW

Liquid LAW is well decontaminated to a sufficient degree that it can be released to the environment. The most effective way to reduce the large volumes of MAW is evaporation. By this way, a reduction factor of about 6 can be obtained. The concentrates are solidified either with cement/concrete or bitumen (asphalt). Compared to cementation, bituminization yields a final product which is smaller by about a factor of 5 compared to that of cement. The leach-resistance of bitumen is better than that of cement, but the radiation resistance (radiolysis) is worse. It has to be verified that the temperature in bitumen does not pass the softening-point of about 70°C, and that the radiolytic decomposition of bitumen with the release of H_2 does not lead to an ignitible gas/air mixture in the storage cavity.

Waste from the organic extraction phase is treated separately. By adding phosphoric acid, TBT can be separated from the diluent kerosene.

4.3.2. Treatment of HAW

The HAW nitrate solutions of fission products and actinides are concentrated by evaporation and then stored in cooled stainless steel containers for about 5 years. The concentrates are cooled and, in addition are stirred to prevent sedimentation. In order to prevent a release of the long-lived α-waste in a terminal storage (see below), it is the objective to solidify the HAW concentrate by calcination and vitrification at ~1000°C. The resulting glass-product must show resistance against radiation, temperature and leaching as well as mechanical and chemical durability. At present, vitrification is done with boron-silicate glasses. Radiation resistance has been investigated by implantation of Cm242 and Cm244. The main difficulty is to have a reliable extrapolation of the durability for a period of 10^4 to 10^5 years.

4.3.3. Waste Disposal in Geological Formations

The perpetual storage of HAW is considered in salt, free from groundwater. In Germany, the USA and in other countries waterfree salt domes exist since about 200 Mill. yrs. If no crystal water is released at higher temperatures (≥ 100°C), the burial of HAW in deep underground cavities offers to be a suitable method for terminal storage. Radioactivity and volumes of the final waste products from the various stages of the fuel cycle of a 1000 MWe LWR are listed in Table XXI /18/.

4.3.4. Nuclear Data Aspects in Waste Management

Two main aspects of waste management relate to nuclear data: (a) protection against gamma and neutron radiation during waste packaging, and (b) restriction of the temperature level and radiolysis effects in the solidified products. (It is self-understanding that the in-pile formation of the heavy elements and fission products and their out-of-pile decay determines the amount of radionuclides in the waste.

For thermal power production, gamma and neutron radiation in waste, the main nuclides are listed in Tables XV-XX.

Because the waste-concentrates are stored 3-5 years before solidification, the specific activity is reduced further. The vitrification of HAW is done in bunkers which are safe against external events like a plane-crash. The concrete of the walls is densified and includes heavy metal (e.g. Fe or Pb). Thus the shielding requirements during packaging HAW can easily be met without improved nuclear data knowledge. The question of temperature level and distribution in solidified products has been touched earlier. According to the obtained accuracy in decay heat prediction (see section 3), no further data requests need to be formulated. Uncertainties in data are shadowed by the irregular disposal of waste-drums in the cavity and also by the inhomogeneity of the heat-source distribution in the drums.

Radiolytic effects, especially in bitumen products, are not sensitive to the present uncertainties in radiation intensity. Thus nuclear data requests for waste management are mainly covered by the data requests for the build-up of radionuclides in the reactor, see chapter 3. The decay properties of nuclei (half-lives, spontaneous fission rates and average gamma and beta energies) are sufficiently well known for waste management aspects at present.

In what remains, up-dating of the various libraries in use has to be performed and, for the prediction of the radioactive inventory in the waste, a sophisticated use of simple reactor physics methods has to be applied to obtain reliable nuclide concentrations.

4.4. Shipment of LWR-Fuel and Waste

The safe shipment of fuel material between the various stages of the fuel cycle requires shielding against neutron and gamma radiation and criticality control. The latter is achieved by limiting the amount of shipped fuel material in critically safe configurations. Furthermore, measures are required to remove the decay heat from irradiated fuel and for mechanical stability of the fuel casks with respect to accidents during road or rail transportation. All containers have to be designed in such a way that they withstand drop tests from about 9 m height onto concrete (or rock), onto a strong pin or trunnions. They have to withstand a fire of about 30 minutes and about 800°C. Submerged into water, any leakage of container-inventory during 8 hours has to be prevented.

4.4.1. Shipment of Unirradiated Fuel

Three groups have to be distinguished:

(a) The shipment of UF_6 and UO_2, which does not involve additional measures compared to those mentioned above.

(b) Shipment of pellets, pins and fuel bundles. This heigh-valued material has to be protected during transportation against shocks, vibrations etc.

(c) Shipment of plutonia as oxide powder or nitrate-solutions for fabrication requires additional measures. Especially in nitrate solutions, the radiolysis of water by radiation leads to hydrogen and oxigen build-up resulting in pressurization of the container. Therefore all containers in which plutonium nitrate is shipped, have an inner pressure vessel.

4.4.2. Shipment of Irradiated Fuel

Spent fuel casks are designed as a compartment or basket to hold and position a number of fuel assemblies in a critically safe configuration. A leak tight inner containment and strong gamma (Pb or depleted U) and neutron shields have to be installed. The heat production in a LWR fuel assembly is between about 4 kW (BWR) and 10 kW (PWR) half a year after discharge. Heat removal is achieved by metallic fins on the container surface (small containers) and in addition by forced cooling (large containers, up to 70 t - 100 t weight for about 6 t of spent fuel) (Note:

though the large containers have a favorable service load to total weight ratio, the upper limit for the dimensions seems to be reached from the handling point of view).

4.4.3. Shipment of Radioactive Waste

LAW and MAW is shipped in drums of 200 l to 400 l capacity. HAW containers are under development; the vitrified waste is packed in about 1 m long stainless steel containers of about 20 cm diameter. For the gamma and neutron shields as well as for the shock absorbing covers and decay heat removal devices the same criteria hold as for spent-fuel flasks.

4.4.4. Nuclear Data Aspects

The nuclear data aspects in fuel and waste transportation are related to the amount of fission products and actinides in the shipped fuel or waste, the decay rates, beta and gamma energies (heat production, shielding) and the neutron intensity of spontaneous fission neutrons and neutrons from (α,n)-reactions (shielding). The dominant nuclides for these processes are listed in Tables I-XII for various times after discharge (which in this context have to be interpreted as those times when spent fuel or waste is shipped). Because protection of the operating personnel and the environment has to be guaranted, safety factors are applied to shielding and cooling devices as well as to geometry and amount of shipped material to ensure critically safe configurations. The neutron radiation requires special attention. It is desirable to know the neutron sources to about 10% accuracy. This is practically reached at present. Therefore, for spent-fuel or waste shipment no further urgent data requests seem to be necessary. For further details on nuclear data for shielding see /19/.

5. Recycling of Plutonium in LWRs

In order to preserve uranium resources, the recovered plutonium from LWRs will be recycled in LWRs in some countries until the preferential use in fast reactors will be possible in large scale. The recovered plutonium can be blended with natural or depleted uranium. In order to keep the fabrication costs low, the number of Pu pins should be kept as small as possible.

5.1. Fabrication of Plutonium Fuel

The fabrication process of plutonium fuel (see ref. /20/) is complicated by the activity of plutonium of about 1 Ci/gr. Technical quantities have to be handled in closed glove-boxes with low pressure to avoid the release of activity into the working hall. This especially is necessary because, due to the α-decay of the plutonium isotopes (except Pu241), plutonium aerosols are formed by α-recoil from the surface of ceramic fuel, bearing the danger of incorporation . Special attention has to be paid to the fact that PU(III)-oxide is hygroscopic and reacts already at room temperature with humidity. Therefore the pellets are dried in vacuum before being filled into rods, which are subsequently sealed by welding. By this procedure also care is taken for the removal of H_2 at high temperature, which is formed as a radiolysis product in humid glove-box atmosphere. This is extremely important because H_2 reacts with the clad material and may lead to clad-failure during reactor operation.

Another problem arises from the fact that Pu241 decays into Am241 which emits a 60 keV γ-ray. To avoid this complication, Am241 is separated from plutonium immediately prior to fabrication. Depending on the amount of Pu238, Pu240 and Pu242 in plutonium fuel, neutron radiation due to the relatively high spontaneous fission rates of these isotopes has to be attenuated.

The fabrication process itself has also to be considered with respect to the solubility of mixed oxide fuel in the head-end step of reprocessing. There are indications that the dry sintering process does not lead to sufficiently homogeneous

$(Pu\text{-}U)O_2$ crystals which may cause uncomplete dissolution in hot HNO_3; a wet fabrication process (sol-gel) would be preferable from this point of view.

The above mentioned problems have led to a mechanized processing with hands-on-maintenance. It is obvious that further increase in activity, e.g. in fabricating "proliferation resistant" fuel (low decontamination after the first extraction cycle), or transplutonium fuel elements for subsequent incineration, will lead to fully remote fabrication techniques, which are not available on an industrial scale at present (see chapter 8).

5.2. Repeated Recycling of Plutonium in LWRs

Multiple recycling of plutonium in LWRs has the effect that the plutonium concentration in the reactor is increased and, in addition, the isotopic plutonium composition is shifted to increase the higher Pu isotopes. While the Pu composition in reprocessed fuel from an originally U-fueled LWR is about (Pu238 : Pu239 : Pu240 : Pu241 : Pu242) ≈ (2 : 59 : 24 : 11 : 4), after some recycling steps this composition changes to about (3 : 38 : 28 : 18 : 13). Thus the spontaneous fission rates are increased and by this neutron and gamma radiation during fabrication of repeatedly recycled fuel. In addition, the temperature of the fuel is increased by the higher amount of the α-decaying Pu238. Therefore the above mentioned difficulties in the fabrication process are enlarged.

As a consequence of the high Pu concentration in LWRs after repeated recycling, Am and Cm is increased to more than an order of magnitude compared to the amount after the first cycle. By this the α-activity and the spontaneous fission rates are increased with the already discussed problems in shipment, reprocessing and waste disposal, requiring stronger gamma and neutron shields and also improved cooling capabilities. In addition, the larger amount of plutonium requires special attention to the criticality problem in all stages of the out-of-pile fuel cycle.

5.3. Nuclear Data Aspects

As far as nuclear data aspects are concerned, it is obvious from the discussion above that the relevant requests arise from the in-pile production of the radionuclides, especially for the transuranium isotopes and the fission products. Because the decay heat prediction is sufficiently accurate at present, especially the neutron sources from spontaneous fission and (α,n)-reactions (i.e. mainly the concentration of the neutron and α-emitters) should be known to a higher accuracy for repeatedly recycled fuel than for fuel of the normal LWR cycle. As already mentioned, work is in progress to improve the nuclear data situation of the higher plutonium isotopes (plus Pu238) as well as of the transplutonium isotopes Am and Cm. Nevertheless, sensitivity calculations are recommended to clarify the needed accuracy of the corresponding data for repeatedly recycled LWR-fuel with respect to radiation shielding (storage, shipment, refabrication, waste packaging) and criticality (storage, shipment, reprocessing, refabrication).

6. The Out-of-Pile LMFBR Fuel-Cycle

In a fast reactor neutron spectrum the higher plutonium isotopes Pu241 and Pu242 are burned much more effectively than in thermal reactors. Thus the core plutonium consists only of about 6 % Pu241 and of about 4 % Pu242 after 67000 MWd/t burn-up in a 1000 MWe plant. Yearly about 2 t of plutonium have to be reprocessed compared to 280 kg from an once-through LWR and about 1 t from a LWR with repeatedly recycled plutonium. As a consequence of the effective consumption of the higher Pu-isotopes in a LMFBR, the production rate of Am and Cm is less than in a LWR with recycled plutonium. Especially the Cm build-up (as the main source for neutron production) is even less than in the LWR once-through cycle (0.6 kg/yr compared to ~ 1 kg/yr). The fraction of the Pu238 (α- and neutron source) build-up (≤ 1 % compared to ≤ 2 %) is also decreased.

The yearly amount of about 20 kg Am from a 1000 MWe LMFBR is less by about a factor of 4 compared to a LWR with recycled fuel and larger by about a factor of 4

compared to the LWR once-through cycle. Those problems, which have been discussed in relation to Cm and Pu238 in the previous sections, are slightly decreased for Cm in fast reactor fuel even compared to the once-through LWR fuel cycle; for Pu238, due to the higher amount of reprocessed Pu, they are increased.

The main problems in out-of-pile fast reactor fuel cycle stages are associated with the large amount of plutonium and with the high burn-up in fast reactors. Research and development are related to /15/:

a. Pu-losses in the dissolution and extraction process should be minimized.

b. The addition of a reducing agent in separating Pu from U should not largely expand the process volume.

c. Critically safe configurations for the high Pu concentration have to be assured.

d. An efficient removal of the enlarged amount of insoluble fission product residues has to be achieved to avoid hydraulic disturbances.

e. Radiolysis has to be reduced (e.g. by reducing the contact times in the extractors) to avoid Pu losses in binding Pu to the radiolysis products.

As common reprocessing of core and blanket fuel elements is foreseen, the effect of high burn-up is reduced.

Nuclear data requirements are similar as in LWR fuel cycles, or even relaxed (see above and the discussion in chapters 4 and 5). However, because of the larger uncertainty of the data in the fast neutron energy region, a large effort is being undertaken to fulfill the requested accuracy for the actinides, put forward at the TND-consultants meeting at Karlsruhe 1975 /8/. No further nuclear data requests are necessary at present.

As already stated in section 3., the decay heat prediction for fast neutron fission is not as accurate as for thermal neutron fission. Due to the higher power density in spent fast reactor fuel, appropriate cooling devices have to be applied in the relevant out-of-pile stages (shipment, storage, evaporators, waste concentrates). A 10 % accuracy for decay heat prediction should be achieved.

7. The Thorium Fuel Cycle

The thorium high-temperature reactor (THTR or HTR) is under development in the USA and in Europe since about 15 years. Two HTRs of about 300 MWe have been designed, Fort St. Vrain in the US is operating, the German THTR is under construction.

THTRs operate on carbide-thorium and carbide-uranium particles, which are coated with pyrolytic carbon and silicon-carbide layers to retain the fission products.

7.1. Reprocessing of HTR-Fuel

A cooling-time for spent-fuel of at least 200 days is necessary to allow the Pa233 (27 d) to convert to U233.

The primary fissile inventory of HTRs is U235. If only the fissile particles U235 are coated with silicon-carbide, the fertile particles (together with the bred U233) can be separated from the fissile particles in the head-end process. The spent-fuel elements are mechanically crashed and then burnt to remove the fuel element graphite and the pyrolytic carbon. In the subsequent leaching process, the fertile particles (with U233) are dissolved, while the silicon coating of the fissile particles remains intact during burning and leaching. They therefore can be separated to be grinded, burnt and then to be leached. Both the streams are treated separately to extract the fuel from the fission products by organic TBP solvents. For the extraction of Th/U233, the THOREX process is applied, while the PUREX process is used to separate uranium from the fission products. Thorium is selectively washed out with nitric acid so that after final decontamination steps U233 is obtained.

The amount and composition of fission products are not essentially different from

those of a LWR. Besides Kr85 and I129, C14 and tritium in the dissolver off-gas have to be retained. Tritium is expected to appear as gaseous effluent, only little is found in the liquid waste. Therefore, as in the case of LWR fuel reprocessing, the off-gas decontamination is very essential.

An advantage of HTRs is, that the build-up of long-lived α-emitters is by far smaller compared to LWRs and LMFBRs. As an example, a 1000 MWe HTR which is fueled with U233 and U235 as fissile material, Th232 and a small amount of U238 as fertile material with the isotopic composition (U233: U235: U238: Th232) ≈ (5: 10: 1: 200) yields at a burn-up of 94000 MWd/t less about 2 kg of Pu (about 60% are Pu238) per ton of heavy material. There are about 45 gr of Am and about 20 gr of Cm per ton of heavy material at 1 yr after discharge. Thus the perpetual storage problem for times after about 1000 yr (when almost all of the fission products are decayed), is reduced.

Because the experience with HTR-fuel reprocessing is by far not as large as for LWR fuel reprocessing, the present process difficulties require intensive research, especially with respect to the extrapolation of experience obtained in laboratory investigations to technological plants.

7.2. Refabrication of HTR-Fuel

Fuel benefit from the thorium cycle can only be gained, if U233 can be efficiently separated in the reprocessing stage, and if subsequently U233 fuel can be fabricated. The main problem in the U233 fabrication process is related to the high activity from the decay daughters of U232 (72 yr), which is formed in-pile by

(a) $Th232\ (n, 2n)\ Th231 \xrightarrow[25.6\ h]{\beta} Pa231\ (n,\gamma)\ Pa232 \xrightarrow[1.3\ d]{\beta} U232$

(b) $Pa233\ (n,2n)\ Pa232 \xrightarrow[1.3\ d]{\beta} U232$

(c) U233(n,2n) U232

Route (a) is the dominant production chain of U232. The decay daughters of U233, mainly Bi212 and Tl208, emit very penetrating gamma rays of 0.4 to 2.1 MeV (Bi212) and 2.6 MeV (Tl208). The decay-chain is the following:

$$U232 \xrightarrow[71.7\ yr]{\alpha} Th228 \xrightarrow[1.9\ yr]{\alpha} Ra224 \xrightarrow[3.7\ d]{\alpha} Rn220 \xrightarrow[56\ s]{\alpha} Po216 \xrightarrow[145\ \mu s]{\alpha} Pb212 \xrightarrow[10.6\ h]{\alpha}$$
$$Bi212 \xrightarrow[1\ h]{\alpha} Tl208 \xrightarrow[3.1\ m]{\alpha} Pb208$$

(The branching from Bi212 to Po212 is not shown here, also the small contribution from the α-decay of Th232 ($1.4 \cdot 10^{10}$ yr) is neglected).

Because of the 72 yr half-life of U232 after separation of the decay daughters in the reprocessing step the radiation level will increase gradually, and for about 2 to 3 weeks after reprocessing U233 can be fabricated by glove-box techniques. After that time heavy shielding is required and fully remote fabrication is foreseen.

7.3. Nuclear Data Aspects

As in all other cases, discussed in previous chapters, the main data requests originate from the in-pile formation of radionuclides. Especially the (n,γ), (n,f) and (n,2n) reactions for Th232, U233, Pa231 and Pa233 are important for the operational characteristics of a HTR as well as for the out-of-pile stages of the fuel cycle. These data are not sufficiently well known at present (see also the corresponding contributions to this conference). Furthermore, evaluators should check the accuracy of fission product yields from U233 fission. Because there is no essential difference in the fission product sequence from HTRs compared to that from LWRs, there is no urgent need to investigate mean beta- and gamma-energies for thermal power production in out-of-pile fuel cycle stages.

8. Alternate Fuel-Cycle Concepts and Related Problems

Only a brief account of alternate fuel cycle concepts is given in this paper. As far as thorium is used as fertile material (to avoid a plutonium economy), the problems are similar to those arising in the HTR fuel-cycle. The same holds for the corresponding nuclear data situation in these systems. If other fuel concepts are to be invented, the technological difficulties by far dominate any drawback from related nuclear data uncertainties.

8.1. General Background

Because the demands for energy continue to grow (although the growth-rate is decreasing), the depletion of natural energy resources requires that in the nuclear power industry the breeding of fissile material instead of U235 extraction from ore has to be investigated more deeply. It is well known that the solution of this problem can best be obtained on the basis of fast power reactors. Since 1976 the question of proliferation of fissile material for weapons' fabrication plays an important role in the international discussions on nuclear energy. In safeguarding nuclear materials, the requests for effective and early detection of any diversion of fissile material from the nuclear fuel cycle were narrowed. To avoid an increasing plutonium economy, the thorium fuel cycle gained more interest and also concepts for burning plutonium and the long-lived α-waste from the U/Pu fuel cycle. Symbiotic systems with Th/U233 and U/Pu fuel were considered more intensively. It should be mentioned that some of the concepts presently discussed had been investigated already in the late fifties and early sixties and had been abandoned either for economic reasons or for associated unresolved technological problems.

It is not attempted in this paper to give an overall validation of the various alternate fuel cycle and corresponding reactor concepts. A brief description of the main features and problems of alternate fuel cycle concepts is given to render a balanced discussion of related nuclear data aspects. For further references see for instance /21/ and /22/.

8.2. Alternate Thermal Reactor Concepts Using the U/Pu-Fuel Cycle

These concepts aim at the preservation of uranium resources. Pu-recycling in thermal reactors has been discussed in chapter 5 and is technologically feasible. In order to improve the conversion to fissile material in LWRs, on very general physical reasons the parasitic neutron absorption has to be reduced. In the Spectral-Shift-Control Reactor this is achieved by the addition of D_2O to the H_2O moderator. By this procedure the neutron spectrum is hardened, the reactivity compensation at the beginning of reactor life is avoided and, in consequence, there is no need for having soluble boron as a neutron absorber at reactor start-up. During burn-up D_2O has to be replaced successively by H_2O. Technological experience is available (Vulcain program up to 1963 in BR3, Mol, Belgium). In order to improve further the conversion, the LWR lattice pitch can be narrowed. In PWRs the moderator-to-fuel-volume ratio is about 2, a reduction of this ratio to less than 1 yields a very tight lattice. If the control of criticality is not performed by the usual control rod concept but rather by movable fuel, the parasitic neutron absorption is decreased further. These LWR-High-Converters are not technologically available at present. The main physical problems are related to a proper prediction of tight-lattice quantities, especially a reliable description of space-energy self-shielding in the resolved resonance region. From the safety point-of-view, e.g. the operational behaviour of the reactor and the functioning of a tight-lattice emergency-core-cooling in an accident situation require intensive research work.

In a very special alternate reactor concept the burning of plutonium, produced in LWRs, is considered. If reprocessing of LWR-fuel is postponed, the idea is to burn spent LWR-fuel in heavy water reactors (TANDEM-concept) either by reassembling the LWR bundles to HWR-fuel elements (a difficulty arises from the different length of the fuel pins!), or, after removal of the clad and of the gaseous and volatile fission products, by refabricating the grinded active fuel to pellets to be assembled to HWR-fuel elements. The latter procedure is not feasible

technologically at present.

8.3. Alternate Reactor Concepts Using the Th/U Cycle

These concepts are aimed at avoiding an extensive plutonium economy and, by the use of Th and U233, preserving the uranium resources.

LWRs and HWRs (e.g. the Canadian CANDU-PWRs) in principle can be fueled with ThO_2 as fertile material instead of UO_2. Because U233 is not available, these reactors have to use at beginning of life U235 as fuel (e.g. highly enriched to reduce Pu build-up). U233 could be produced in Th-blankets of fast reactors and, after reprocessing and refabrication, could then be loaded into LWRs. The same procedure can also be considered in connection with U233 fueling of HTRs.

Optimization of these types of symbiotic systems with regard to fuel consumption in long-range reactor strategies is performed world-wide in the international fuel cycle evaluation (INFCE) program.

A special alternate concept has been developed to achieve breeding in LWRs with Th/U233 fuel. This Light-Water-Breeder Reactor /23/ went in operation in 1977 in the Shippingport plant in the USA. Control of the reactor is performed with the highly enriched fissile fuel-moduls (seed) moving relative to the surrounding Th-blanket elements (seed- and blanket-concept).

The main technological problems associated with the utilization of Th and U233 in power reactors are given by the lack of experience in reprocessing spent fuel and in the difficulties of fabrication and refabrication of U233 fuel. This has been discussed already in chapter 7. Economic questions are not touched here.

8.4. Alternate Fuel Concepts

Alternate fuel concepts are being investigated mainly to reduce the possibility of fissile-material diversion for weapons' fabrication and to reduce the potential hazards of long-lived fission products and α-emitters in the waste. Weapons' grade materials are plutonium with low Pu240 content (spontaneous fission!), highly enriched U235 and U233. The critical masses are (with U238 reflector): $\lesssim$ 10 kg ($\gtrsim$ 95 % of Pu239), $\sim$ 14 kg (70 % of Pu239), $\sim$ 21 kg (commercial PuO_2), $\sim$ 31 kg (U235), $\sim$ 11 kg (U233). As discussed in chapter 7, after separation of U233 from Th and fission products, during few weeks U233 can be handled in normal glove-boxes before the radiation level becomes too large.

An increase of the difficulties in fuel handling would decrease the possibility of diversion of fissile material from the fuel cycle. For this reason, the question of coprocessing of uranium and plutonium is discussed. One possibility is to commonly process U and Pu after the first extraction cycle, i.e. after separation of the fission products and Np, Am, Cm (CIVEX-Process). Because a decontamination factor of only $\lesssim 10^3$ is reached in this case, any further handling of the fuel requires heavy shielding, which renders diversion more difficult. However, besides the fact that fabrication of the hot fuel for further use in reactors has to be performed remotely (a technique which is not yet available on industrial scale), the U235/Pu239 concentration is by far too low to reach criticality; highly enriched U or Pu fuel has to be fabricated for spike-elements: only partial coprocessing is possible, the advantage of hot-fuel handling as a means against diversion therefore has decreased. Another possibility is to blend predecontaminated uranium and plutonium after the second extraction cycle (Pu/U separation) to the desired Pu:U ratio; this process is used in some reprocessing plants.

Contamination of fuel by means of Pu238 has also been discussed recently /24/. Because of the strong α-decay, 5 % of Pu238 in Pu-fuel may lead to surface temperatures of about 800°C; therefore chemical explosives triggering most nuclear weapons would melt already at 200°C. In thermal reactors, 5 % Pu may be reached already after 2 recycling steps, if Np237 and U236 are recycled likewise.

In reducing the α-waste hazard potential, recycling of the Am and Cm isotopes into reactors is discussed. Fast reactors would be suitable reactors for the transmutation of these isotopes to fission products of shorter half-lives /25/. The main technical problems are associated with the very high specific neutron and gamma activity, requiring additional shielding in reprocessing (necessary after the first recycling step), transportation and refabrication (fully remote). On the other hand, separation of Am and Cm from the fission products is rather difficult because of the chemical similarity to the lanthanides. Techniques for efficient actinide partitioning from waste will not be available for a long time. If part of the fission products were recycled together with the waste actinides, the difficulties would even be increased.

From the discussion above it is obvious that radioactive fuel decreases the possibility of diverting fissile material but increases the handling of the fuel for further use in reactors, associated with large technical and economical impacts.

Therefore, in another alternate fuel concept a dilution of U233 with U238 is envisaged with thorium and a U233:U238 ratio of about 1:6. This denatured fuel cannot be used for weapons' fabrication. In addition, U233 cannot be separated chemically from U238 (but the physical separation is easier than that of U235 from U238!). The denatured fuel cycle /26/ could be realized in the following way: All dispersed national reactors (e.g. LWRs or HTRs) are fueled with denatured fuel (U233, U238, Th232, no Pu). Irradiated fuel (including U233 and Pu) is shipped to a closed and internationally safeguarded fuel cycle center, where the hot fuel is reprocessed to uranium and plutonium products. Plutonium could be used as core-fuel of fast reactors with thorium blankets, where U233 is bred to be blended with U238 and bred U233 from the dispersed reactors. This denatured fuel again is fabricated inside the center and shipped out to national reactor plants. Waste is stored within the area of the center and plutonium does not leave the center.

This concept implies almost a complete change of the nuclear power and fuel cycle industry. The principal difficulty lies in the necessary coprocessing of Th/U/Pu mixed-oxide fuel, which is not available at present. The described scheme may require for large scale application about 20 years research and development. Related economic questions are not considered here.

8.5. Nuclear Data Aspects

As is obvious from the discussion in sections 8.2 to 8.4, an improvement of nuclear data uncertainties cannot reduce the technological problems associated with an implementation of alternate fuel cycles. As already mentioned in the introductory remarks to this chapter, almost all of the requests are already covered by the corresponding requests in thermal or fast systems using the U/Pu cycle, or by the requests for thermal Th/U systems as e. g. the HTR. As far as those concepts with transmutation of Am and Cm are concerned, the technical realization is far away; the data status is sufficient for the present stage of investigation.

9. General Conclusions

The balanced discussion of technical procedures and the related physics aspects in the out-of-pile stages of the nuclear fuel cycle shows that almost everywhere in the established fuel cycle for LWRs an improvement of the presently available nuclear data will not help to reduce the problems. Quite generally, the situation in the out-of-pile fuel cycle is different from the in-pile situation, where the nuclear reactions directly control the reactor performance (criticality, reaction rate balance, and power distribution, reactivity worth, burn-up and kinetics behaviour) while in the out-of-pile stages the nuclear aspects are mainly considered in connection with the protection of personnel and plant components against neutron- and gamma-radiation, heat production and criticality. The latter processes are not controlled by nuclear aspects. Data requests are relaxed out-of-pile also because additional safety margins are applied.

The main data requests for the out-of-pile stages of the fuel cycle originate from the formation of radionuclides in-pile. For the transuranium isotopes these requests were given at the Karlsruhe TND-Meeting in 1975 and for the fission products at the Petten-Meeting in 1977. In applying simplified methods to describe the in-pile and out-of-pile behaviour of nuclear fuel and fission products, only a sophisticated use of these methods can give reliable results. These have to be checked against more elaborate reactor physics methods and against experimental results, e.g. those of post-irradiation examinations. In this paper, a special recommendation is given to check the nuclear data of strong fission product absorbers, such as Eu, Gd, Hf, and to improve the data if necessary. Physics experiments are recommended for heterogeneous neutron poisons in reprocessing plants for complicated geometries. For repeatedly recycled fuel, higher accuracies might be necessary compared to those of the usual LWR fuel cycle, mainly for shielding and cooling purposes in the reprocessing, transportation and refabrication steps (e.g. because of the high Pu^{240}, Pu^{242}, Am and Cm contents as neutron sources). Sensitivity studies should be performed to establish the adequate needs. The decay heat production from fast neutron fission should be predicted to about 10 % accuracy, which is not reached in general. For the thorium cycle, (n,γ), (n,f) and $(n,2n)$ cross sections in-pile have to be improved for U^{233}, Th^{232}, Pa^{231}, Pa^{233}. Fission product yields for U^{233} fission should be reconsidered for intolerable uncertainties.

For alternate fuel cycles, the technical problems are dominating by far any drawback from the uncertainties in nuclear data. For the investigations done at present, the nuclear data status is sufficient.

As a final remark, users of data libraries for fuel cycle calculations should take care of updating their data-sets to the present knowledge. An internationally recommended standard of half-lives and fission product yields would be welcomed.

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Table I : Percentage of Therm. Power from Heavy Nuclides**
(Interim Storage and Reprocessing)

Nuclides	Time after Discharge 1yr	3yr	10yr
CM242	55.4	5.3	.1
PU238	19.6	40.8	40.
CM244	16.9	32.7	25.8
PU240	3.1	6.5	6.8
PU239	2.1	4.4	4.5
AM241	1.8	8.1	21.1
PU241	.9	1.6	1.2
AM243	.1	.3	.3
Rest	.1	.3	.2
Total Thermal Power*	473.	227.	220.

*in W/tHM, tHM = Metric Ton of Heavy Material

Table II: Percentage of Radioactivity from Heavy Nuclides
(Interim Storage and Reprocessing)

Nuclides	Time after Discharge 1yr	3yr	10yr
PU241	88.	93.	90.9
CM242	6.3	.3	
PU238	2.5	2.8	3.7
CM244	2.0	2.2	2.3
PU240	.4	.4	.7
PU239	.3	.3	.4
AM241	.2	.6	.2
Rest	.3	.4	.0
Total Activity*	113. $\times 10^3$	97.1 $\times 10^3$	71.4 $\times 10^3$

*in Curie/tHM

**Tables I - XX refer to a 1000MWe PWR reaching a burn-up of 33000 MWd/tHM in 1096 full-power days (3 years). The results presented in these tables are based on calculations with the latest version (1978) of ORIGEN (nuclear data basis of ENDF/B-IV)

Table III: Percentage of Neutrons from Heavy Nuclides by Spont. Fissions (Interim Storage and Reprocessing)

Nuclides	Time after Discharge 1yr	3yr	10yr
CM244	87.	97.4	97.8
CM242	11.4	.6	
PU240	.6	.7	.9
CM246	.5	.6	.8
PU242	.2	.2	.3
PU238	.1	.1	.2
Rest	.2	.4	.0
Total*	3.7×10^8	3.06×10^8	2.33×10^8

*in Neutrons/tHM

Table IV: Percentage of Neutrons from Heavy Nuclides by (α,n)-Reactions (Interim Storage and Reprocessing)

Nuclides	Time after Discharge 1yr	3yr	10yr
CM242	61.6	6.7	.1
CM244	16.4	36.5	29.4
PU238	16.4	39.4	39.3
PU240	2.2	5.4	5.6
PU239	1.5	3.6	3.8
AM241	1.5	8.	21.4
AM243	.2	.4	.4
Rest	.2	.0	.0
Total*	3.36×10^7	1.4×10^7	1.33×10^7

*in Neutrons/tHM

Table V : Percentage of Therm. Power from Fission Product Nuclides (Interim Storage and Reprocessing)

Nuclides	Time after Discharge 1yr	3yr	10yr
PR144	33.8	16.9	.1
RH106	27.4	20.5	.5
CS134	17.8	26.9	8.4
Y 90	4.3	12.2	34.1
BA137M	3.7	10.6	30.0
CE144	3.6	1.8	
NB 95	2.7		
CS137	1.6	4.7	13.4
EU154	.5	1.4	3.5
PM147	.4	.7	.4
Rest	4.2	4.3	9.6
Total Thermal Power*	10.3 $\times 10^3$	3.46 $\times 10^3$	1.04 $\times 10^3$

*in W/tHM

Table VI: Percentage of Radioactivity from Fission Product Nuclides (Interim Storage and Reprocessing)

Nuclides	Time after Discharge 1yr	3yr	10yr
CE144	20.4	9.5	
PR144	20.4	9.5	
RH106	12.2	8.6	.2
RU106	12.2	8.6	.2
CS134	7.9	11.2	2.6
CS137	4.8	12.7	27.0
BA137M	4.4	11.9	25.2
PM147	3.7	6.2	2.4
Y 90	3.4	9.1	19.4
SR 90	3.4	9.1	19.4
KR 85	.4	1.3	1.8
EU154	.3	.9	1.4
Rest	6.5	1.4	.4
Total Activity*	2200. $\times 10^3$	785. $\times 10^3$	315. $\times 10^3$

*in Curie/tHM

Table VII: Main Contributers for Photon Production (γ+ Bremsstrahlung) of Structural Materials (SM), Fission Products (FP) and Actinides (AC)

Average Energy [MeV]	SM - Time after Discharge 1yr	3yr	10yr	FP - Time after Discharge 1yr	3yr	10yr
0,3	SB125	-	-	CE144	CE144	CE144
				RH106	RH106	
				Y 90	Y 90	Y 90
				CS137	CS137	CS137
0,63	NB 95	-	-	CS134	CS134	BA137M
	ZR 95	-	-	BA137M	BA137M	CS134
	SB125	SB125	SB125	PR144	PR144	Y 90
	MN 54	MN 54	-	Y 90	Y 90	
	CO 58	-	-	RH106	RH106	
1,1	CO 60	CO 60	CO 60	RH106	RH106	
				CS134	CS134	CS134
Photons sec·tHM	$1,47 \cdot 10^{14}$	$5,2 \cdot 10^{12}$	$1,6 \cdot 10^{12}$	$3,4 \cdot 10^{16}$	$1,4 \cdot 10^{16}$	$4,2 \cdot 10^{15}$
Mean Energy Per Photon [MeV/Photon]	0,645	0,965	1,05	0,614	0,630	0,634

Average Energy [MeV]	AC - Time after Discharge 1yr	3yr	10yr
0,03	AM241	AM241	AM241
0,04	AM242	AM242	AM242
0,06	PU240	PU240	PU240
	PU242	PU242	PU242
	AM243	AM243	AM243
	AM241	AM241	AM241
0,1	PU240	PU240	PU240
0,15	CM245	CM245	CM245
	AM243	AM243	AM243
0,2	CM243	CM243	CM243
1,99	TL208		
Photons sec·tHM	$1,1 \cdot 10^{13}$	$1,6 \cdot 10^{13}$	$2,8 \cdot 10^{13}$
Mean Energy Per Photon [MeV]	0,097	0,085	0,073

Table VIII: Percentage of Therm. Power from Structural Materials (Interim Storage and Reprocessing)

Nuclides	Time after Discharge 1yr	3yr	10yr
NB 95	47.6		
ZR 95	24.9	.2	
CO 58	13.3	.1	
FE 55	5.4	38.6	21.1
CO 60	4.6	42.7	60.3
MN 54	2.4	5.3	
SB125	1.1	8.3	5.
NI 63	.3	3.8	12.8
TE125M		.5	.5
Rest	.4	.5	.3
Total Thermal Power*	19.6	1.63	.46

*in W/tHM

Table IX: Percentage of Radioactivity from Structural Materials (Interim Storage and Reprocessing)

Nuclides	Time after Discharge 1yr	3yr	10yr
NB 95	43.2	.1	
ZR 95	20.7		
FE 55	18.3	49.1	15.7
NI 63	8.8	39.8	78.1
CO 58	4.2		
CO 60	1.3	4.5	3.7
SB125	1.2	3.4	1.1
TE125M	.7	1.4	.5
Rest	1.6	1.7	.9
Tot. Activity*	4490	982	475

*in Curie/tHM

Table X : Nuclide Concentrations [gr./tHM] for Main Radioactivity Contributers of Structural Materials in Discharged Fuel

Nuclides	Time after Discharge		
	1yr	3yr	10yr
NB 95	$4.9 \cdot 10^{-2}$	$2.1 \cdot 10^{-5}$	-
ZR 95	$4.4 \cdot 10^{-2}$	$1.8 \cdot 10^{-5}$	-
CO 58	$6.0 \cdot 10^{-3}$	$4.9 \cdot 10^{-6}$	-
FE 55	$3.3 \cdot 10^{-1}$	$1.9 \cdot 10^{-1}$	$3.0 \cdot 10^{-2}$
CO 60	$5.1 \cdot 10^{-2}$	$3.9 \cdot 10^{-2}$	$1.6 \cdot 10^{-2}$
MN 54	$7.2 \cdot 10^{-3}$	$1.4 \cdot 10^{-3}$	$3.9 \cdot 10^{-6}$
SB125	$5.2 \cdot 10^{-2}$	$3.1 \cdot 10^{-2}$	$5.2 \cdot 10^{-3}$
NI 63	6.43	6.33	6.01
TE125M	$1.3 \cdot 10^{-3}$	$7.6 \cdot 10^{-4}$	$1.3 \cdot 10^{-4}$
Total	6.97	6.60	6.06

Table XI: Concentrations [gr./tHM] of Heavy Nuclides in Discharged Fuel.

Nuclides	Time after Discharge		
	1yr	3yr	10yr
U238	$9.43 \cdot 10^{5}$	$9.43 \cdot 10^{5}$	$9.43 \cdot 10^{5}$
U235	$8.03 \cdot 10^{3}$	$8.03 \cdot 10^{3}$	$8.03 \cdot 10^{3}$
PU239	$5.26 \cdot 10^{3}$	$5.26 \cdot 10^{3}$	$5.26 \cdot 10^{3}$
U236	$4.52 \cdot 10^{3}$	$4.52 \cdot 10^{3}$	$4.52 \cdot 10^{3}$
PU240	$2.16 \cdot 10^{3}$	$2.16 \cdot 10^{3}$	$2.16 \cdot 10^{3}$
PU241	$9.78 \cdot 10^{2}$	$8.90 \cdot 10^{2}$	$6.38 \cdot 10^{2}$
NP237	$4.77 \cdot 10^{2}$	$4.77 \cdot 10^{2}$	$4.80 \cdot 10^{2}$
PU242	$3.49 \cdot 10^{2}$	$3.49 \cdot 10^{2}$	$3.49 \cdot 10^{2}$
PU238	$1.66 \cdot 10^{2}$	$1.65 \cdot 10^{2}$	$1.57 \cdot 10^{2}$
U234	$1.23 \cdot 10^{2}$	$1.26 \cdot 10^{2}$	$1.34 \cdot 10^{2}$
AM243	$9.26 \cdot 10^{1}$	$9.26 \cdot 10^{1}$	$9.25 \cdot 10^{1}$
Total	$9.65 \cdot 10^{5}$	$9.65 \cdot 10^{5}$	$9.65 \cdot 10^{5}$

Table XII: Nuclide Concentrations [gr/tHM] for Main Radioactivity Contributors of Fission Products

Nuclides	Time after Discharge 1yr	3yr	10yr
PR144	$5.9\ 10^{-3}$	$1.0\ 10^{-3}$	$2\ 10^{-6}$
RU106	80.2	20.2	0.16
CS134	133.	67.5	6.3
BA137	71.3	126.	297.
CE144	1200.	1150.	976.
Y 90	0.138	0.131	0.111
NB 95	1.5	$6.3\ 10^{-4}$	-
EU154	45.5	41.7	30.8
PM147	89.1	52.5	8.2
Total	1761.	1481.	1318.

Table XIII: Proportions of Strong Fission Product Neutron Absorbers

Nuclides	Concentration [gr/tHM] 1 yr after Discharge		$\sigma^{*}_{n,\gamma}$/Res. Int. [barn]
CS133	1010	(Form. after Discharge)	29/415
TC 99	835		19/340
ND143	804		325/140
ND145	692		42/240
RH103	389	(via RU103 40d)	150/1100
EU153	129		390/1635
SM151	46		15000/3300
SM149	8		41000/3183
EU155	4		4040/ -
GD155	2	(via EU155 5 yr)	61000/1550

* at 2200 m/sec neutron velocity

Table XIV: Comparison of Production and Decay Data for Nuclides of the Feed Solution Residue

Nuclides	Yields from the Fission of									$T_{1/2}$ |sec|			$\bar{E}_\gamma + \bar{E}_\beta$ |eV|	
	$U235_{therm.}$			$U238_{fast}$			$Pu239_{therm.}$							
	ORIGEN-73	ENDFB-IV	IAEA	ORIGEN-73	ENDFB-IV	IAEA	ORIGEN-73	ENDFB-IV	IAEA	ORIGEN-73	ENDFB-IV	IAEA	ORIGEN-73	ENDFB-IV
Nb	0.	6.55_{-2} $\times 10$	-	0.	0.362	-	0.	$6 \cdot 10^{-2}$	-	0.	5.35_{-1} $\times 10$	1.0	0.	6.00_{6} $\times 10^{6}$
Mo	0.	0.275	-	0.	1.94	-	0.	1.68	-	0.	9.0	8.2	0.	1.79_{6} $\times 10^{6}$
Tc	0.38	4.68_{-2} $\times 10^{-2}$	-	2.83	0.422	-	4.57	1.73	-	37.	36.	36.	$2 \cdot 10^{6}$	3.89_{6} $\times 10^{6}$
Ru	0.	1.13_{-3} $\times 10^{-3}$	-	0.0	1.35_{-2} $\times 10^{-2}$	-	0.	0.81	-	3.17_{7} $\times 10^{7}$	3.19_{7} $\times 10^{7}$	3.17_{7} $\times 10^{7}$	$1 \cdot 10^{4}$	$1 \cdot 10^{4}$
Rh106	0.	3.5_{-7} $\times 10^{-7}$	-	0.0	5.6_{-6} $\times 10^{-6}$	-	0.	9.7_{-4} $\times 10^{-4}$	-	30.	29.9	29.9	1.77_{6} $\times 10^{6}$	1.64_{6} $\times 10^{6}$
Total	0.38	0.384	0.391	2.83	2.737	2.71	4.57	4.38	4.40					

ORIGEN73 Data from earlier than 1973

ENDFB/IV Data Status 1975

IAEA/PETTEN Conference Data Status 1977

Table XV: Percentage of Therm. Power from Heavy Nuclides (Waste)

Nuclides	Time in Waste 100yr	1000yr	10000yr
AM241	66.	64.2	.8
CM244	14.3		
PU238	8.8		
AM243	5.5	19.7	44.8
PU240	2.7	10.2	21.3
PU239	.8	3.8	28.3
Rest	1.9	2.1	4.8
Total Thermal Power*	12.2	2.99	.58

*in W/tHM

Table XVI: Percentage of Radioactivity from Heavy Nuclides (Waste)

Nuclides	Time in Waste 100yr	1000yr	10000yr
AM241	60.2	54.4	.6
CM244	12.4		
PU238	8.1	.2	
AM243	4.4	15.5	28.8
NP239	4.4	15.5	28.8
PU240	2.7	9.4	15.6
AM242	1.4		
PU239	.8	3.4	21.1
Rest	5.6	1.6	5.1
Tot. Activity*	400.	105.	25.

*in Curie/tHM

Table XVII: Percentage of Neutrons from Heavy Nuclides by (α,n)-Reactions (Waste)

Nuclides	Time in Waste 100yr	1000yr	10000yr
AM241	64.3	63.2	.8
CM244	15.7		
PU238	8.4	.2	
PU240	2.2	8.3	17.1
AM243	6.5	24.8	56.4
CM242	1.8	.1	
PU239	.6	3.0	23.0
Rest	.5	.4	2.7
Total*	7.62 $\times 10^5$	1.85 $\times 10^5$	3.6 $\times 10^4$

*in Neutrons/tHM

Table XVIII: Percentage of Neutrons from Heavy Nuclides by Spont. Fiss. (Waste)

Nuclides	Time in Waste 100yr	1000yr	10000yr
CM244	71.6		
CM246	20.	87.1	87.
PU240	.4	2.2	3.2
CM242	.3	2.4	
CM248	.1	.4	1.5
PU238	.1		
PU242	.1	.4	1.5
Rest	7.4	7.5	6.8
Total*	9.78 $\times 10^6$	1.94 $\times 10^6$	5.18 $\times 10^5$

*in Neutrons/tHM

Table XIX: Percentage of Therm. Power from Fiss. Prod. Nuclides (Waste)

Nuclides	Time in Waste 100yr	1000yr	10000yr
BA137M	37.		
Y 90	36.5		
CS137	16.5		
SR 90	8.1		
SM151	.9	7.	
EU154	.7		
SB126M		15.	16.5
SB126		31.	31.6
TC 99		43.	45.2
Rest	.3	4.	6.7
Total Thermal Power*	103.	.0224	.0206

*in W/tHM

Table XX: Percentage of Radioactivity from Fiss. Prod. Nuclides (Waste)

Nuclides	Time in Waste 100yr	1000yr	10000yr
CS137	29.3		
BA137M	27.		
SR 90	18.		
Y 90	18.		
SM151	1.5	2.	
EU154	.2		
TC 99		68.3	69.7
ZR 93		9.0	9.4
NB 93M		9.0	9.4
SB126M		2.5	2.5
Rest	6.	9.2	9.
Tot. Activity*	3.35 $\times 10^4$	20.8	19.8

*in Curie/tHM

Table XXI: Volumes and Radioactivity of the Final Waste Products from the Various Stages of the Fuel Cycle of a 1000 MWe LWR

Step in the Fuel Cycle	Waste		
	Type of Waste	Volume m^3/1000 MWe·yr	Radioactivity Ci/m^3
Chopping of Fuel Bundles	Fuel and Clad Parts	5	10^4
Dissolution of Chopped Fuel	Hulls; I, Kr H^3 in Off-Gas	15 (Hulls)	10^4 (Hulls)
Recovery of U and Pu	Fiss. Prod., N_p, A_m, C_m, non-recovered U, Pu	3	10^7
Separation of U from Pu Conversion of U→UF_6, Pu→PuF_6	Contaminated Solvents, Failed Equipment, Trash converted to Solid Waste	75	$<10^3$
Enrichment of U235-Content of Uranium	UF_6-Tailings	50	~1
Mixed-Oxide Fuel Fabrication	Trash, Contaminated Solvents, Failed Equipment converted to Solid Waste	10-40	~1
Fuel Exposure in the Reactor	Solid Waste, Solids, Resins converted to Solid Waste	600-900	1-10

After A.M. Platt: Process Operations and Wastes in the LWR Fuel Cycle, Honolulu Conf., 1977, p.451