





LABORATORY

# AFCI Storage & Disposal FY-06 Progress Report

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Advanced Fuel Cycle Initiative

## **AFCI Storage & Disposal**

## **FY-06 Progress Report**

September 18, 2006

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AJFCI

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## AFCI Storage & Disposal FY-06 Progress Report

Objective: AFCI Storage and Disposal participants at LLNL, ANL and INL provide assessment of how AFCI technology can optimize the future evolution of the fuel cycle, including optimization of waste management. Evaluation of material storage and repository disposal technical issues provides feedback on criteria and metrics for AFCI, and evaluation of AFCI waste streams provides technical alternatives for future repository optimization. LLNL coordinates this effort that includes repository analysis at ANL and incorporation of repository impacts into AFCI criteria at INL. Cooperative evaluation with YMP staff is pursued to provide a mutually agreed technical base. Cooperation with select international programs is supported.

#### FY-2006 Storage and Disposal Workscope:

Task 1 - Coordinate Storage & Disposal Activities - Internal to AFCI, the ability to store, transport and dispose of each product, waste or excess material must be understood. Ongoing waste/storage evaluation provides feedback regarding the output streams and material forms from separations activities. As part of the continuing interface between AFCI and the OCRWM repository program, Storage & Disposal participants at LLNL, ANL and INL will provide assessment of AFCI technology impacts on geologic disposal requirements. Evaluation of repository technical issues provides feedback on criteria and metrics for AFCI, and evaluation of AFCI waste streams provides technical alternatives for future repository optimization. LLNL will coordinate this effort, which includes repository analysis at ANL and joint AFCI/RW systems studies at INL. Input to other AFCI Systems reports or white papers on S&D issues will be provided as needed.

Task 2 - Cooperative Studies with OCRWM - 1) Participate as directed in the NE/RW Joint Steering Committee on Spent Fuel Management including working group activities. 2) Pursue cooperative technical evaluation with YMP staff of design and performance issues for evaluation of waste management beyond the YMP baseline.

Task 3 - International Cooperation - Cooperation with international programs will be supported as directed. The work package manager will also provide input to the Systems NTD for monthly progress reports (by the 10th of the following month) and quarterly technical summaries (by the 15th of the following month). FY-05 Carryover Completion of OECD-NEA report input.

#### Introduction

An important long-term objective of advanced nuclear fuel cycle (AFC) technologies is to provide improvement in the long-term management of radioactive waste. Compared to a oncethru fuel cycle, it is possible to generate far less waste, and potentially easier waste to manage, with advanced fuel cycles. However, the precise extent and value of these benefits are complex and difficult to quantify<sup>1</sup>. At the same time, advanced fuel cycles may create additional product and waste streams that require interim storage or extended storage. This document presents a status report of efforts within AFCI Systems Analysis to define and quantify the AFC benefits to geologic disposal, evaluation of AFC storage requirements, development of cooperative efforts with the US repository program, and participation with international evaluations of AFC impacts on waste management. The primary analysis of repository benefits is conducted by ANL, and waste stream analysis by INL. This year repository impact evaluations have included:

- Continued evaluation of LWR recycle benefits in support of scenario analysis.
- Extension of repository analyses to consider long-term dose reductions.
- Seeking opportunity for cooperation with the U.S. repository program.
- AFC waste and product stream analysis for storage and disposal requirements.
- International cooperation with OECD-NEA.

During the course of the year, the U.S. DOE announced a major new initiative in national and international development of advanced nuclear fuel cycles – the Global Nuclear Energy Partnership (GNEP). This initiative is based in large part on AFCI technology, AFCI Systems Analysis, and in part on prior Repository Benefits and current Storage and Disposal analyses. The Storage and Disposal staff contributed to GNEP planning and preparation for transition of AFCI to a future role as Advanced Fuel Cycle R&D under the GNEP program.

#### Summary of FY-06 AFCI Repository Impact Analysis

While analysis regarding recycle in current and Gen-III light water reactors continued in FY-06, there has been growing focus on fast-spectrum reactor recycle. Much of this driven by the GNEP move directly to full actinide recycle in fast reactors. Repository impact analysis results have been reported separately by ANL and other systems analysis reports.

Developments in the Yucca Mountain Project also impact the focus of AFCI waste management evaluations. Delay of the license application continued to defer cooperative interaction, while the proposed revision of the EPA regulation<sup>2</sup> (40 CFR 197 proposed revision) has focused attention on long-term dose from AFCI scenarios.

#### **Repository Benefit Analysis (ANL)**

Evaluation of repository impacts of advanced fuel cycles continued at ANL. Thermally controlled repository capacity analysis continued with a parametric study of differing levels of separations efficiency for actinides and short-lived fission products. In addition, evaluation of dose rate impacts has begun for several AFC scenarios using a simplified performance model for a Yucca Mountain-like repository.

Just after completion of the FY-05 version of this Summary Report an ANL AFCI Systems Analysis report, "Update Report on the Repository Benefits of AFCI Options," ANL-AFCI-161, by R.A. Wigeland, T.H. Bauer, and E.E. Morris<sup>3</sup> was completed. This report summarized the latest results on potential loading increase with removal of all minor actinides, an update on the potential recycle strategies, and an initial analysis on the potential impact of AFCI strategies on the peak dose rate from the repository. Some results from that report are summarized below:

#### Potential Increase in Utilization of Repository Space from Processing Spent Fuel (ANL):

As has been shown in the past, the loading of the Yucca Mountain repository is constrained by the decay heat generated in the emplaced spent nuclear fuel, since the design and operation of the repository must satisfy a set of imposed temperature limits to ensure adequate performance of the repository system. It had been determined that the dominant contributors to the thermal load of

emplaced spent LWR fuel in a repository at Yucca Mountain that lead to reaching one or more of the temperature limits are plutonium and americium. Removal of these elements, and recycling them to reduce the hazard, is essential to increasing the utilization of space in the repository by allowing greater drift loading. The benefit ranges from a factor of 4.3 to 5.4 in increasing the drift loading (or decreasing the repository size for a given capacity) depending on the separations efficiency, as shown in Fig. 1. It should be noted that for approaches that do not remove americium and plutonium, but only propose to separate fission products cesium and strontium, there is no potential increase in the drift loading.

After the plutonium and americium have been removed, the next elements that need to be separated to further increase utilization of the repository space are cesium and strontium. Removing these elements, and sequestering them in a separate area of the repository or another facility, would allow a further substantial increase in the drift loading of the repository, up to a factor of 42.7 greater than the direct disposal case for 99.9% removal of plutonium, americium, cesium, and strontium. Removal of plutonium, americium, cesium and strontium. Removal of plutonium, americium, cesium and strontium at 99% efficiency provides almost the same benefit as using 99.9% separations efficiency, due to the growing importance of other chemical elements remaining in the waste stream.

The next most important element is curium, as has been noted in the past, but the benefit of subsequent curium removal had not been previously quantified. Assuming that the curium can be removed along with the other actinide elements with the same efficiency, even greater increases in utilization of repository space can be realized, as shown in Fig. 2. In this case, the use of 99.9% separation efficiencies for all of the removed chemical elements results in a potential loading increase of 225. In addition, there is now a substantial difference between separation efficiencies of 99.9%, indicating that the effects of the other elements in the waste stream are still relatively unimportant in generating decay heat. It is also useful to note that by including curium removal in the separation process, the factor for increasing loading rises from 39 to 91 at only 99% separation efficiency.



Figure 1. Potential Repository Drift Loading Increase as a Function of Separation Efficiency for Plutonium, Americium, Cesium, and Strontium<sup>3</sup>



Figure 2. Potential Repository Drift Loading Increase as a Function of Separation Efficiency for Plutonium, Americium, Curium, Cesium, and Strontium<sup>3</sup>

These results show that there is a range of options for achieving a certain increase in repository loading, which can be used to advantage in guiding the design and goals of any planned separations facilities.

## Potential Impact of AFCI Processing and Recycling Options on the Dose Rate for Releases from a Geologic Repository (ANL):

The results described in this section summarize the evaluations performed to date on the potential for AFCI technology options to affect the dose rate associated with releases from spent nuclear fuel or other radioactive wastes destined for disposal in a geologic repository such as the one proposed for Yucca Mountain. As discussed in the following, results are calculated using the DOE Simplified Model, based on the Total System Performance Assessment (TSPA) models for the Site Recommendation phase of the Yucca Mountain project. This work was started during FY05 at a modest level; with plans for increasing the effort during FY06 to provide a comprehensive analysis of the impact of AFCI recycle options on the potential dose rate. It is expected that this work will lead to similar guidelines for actinide and fission product removal to benefit dose in the same manner as has been done for heat load.

#### **Dose Rate Estimates (ANL):**

In this section, the impact of some AFCI recycling strategies on dose rate from a geologic repository is examined. As described in the previous section, processing spent UO2 fuel and recovering and recycling elements (or isotopes) that contribute to the radiotoxicity can have an impact on the overall radiotoxicity by reducing the inventory of these isotopes. However, in the setting of a geologic repository, the relative importance of each element or isotope is modified by dissolution and transport effects, such as solubility limits and colloid formation. In estimating the dose rate associated with releases from a geologic repository, such effects are extremely important.

Previous results have shown that it is possible to greatly increase the utilization of repository space by processing the CSNF and removing those elements responsible for the majority of the decay heat. The resulting consolidation of the process waste causes the radionuclide inventory in the repository to be substantially different than for direct disposal of spent fuel. For example, with 99% removal of plutonium, americium, neptunium, and curium, along with 99% removal of cesium and strontium, it is possible to densify the remaining materials by a factor of 91 as compared to the direct disposal of spent fuel. While this brings the inventory of the actinides and cesium and strontium almost back to the values for direct disposal, the inventory of elements that were not removed has increased by almost a factor of 100. This can be especially important for potentially hazardous radionuclides that were present in small quantities in spent fuel. Examples are technetium and iodine, which are not necessarily important to the peak dose rates expected for releases from the Yucca Mountain repository at this time, but could be if their inventories were greatly increased relative to the actinide elements.

#### **Estimated Dose Rate for Increased Loading of the Repository (ANL):**

Calculations were performed where the spent fuel was processed, and the actinide elements were recovered with an efficiency of 99.9%. The cesium and strontium were also removed with an efficiency of 99.9%, so the potential increase in utilization of repository space is about a factor of 225. However, to keep the estimated peak dose rate the same as for the direct disposal of spent fuel, an increase of a factor of 100 was used, with the results shown in Fig. 3. Again, the dose rate is normalized to the peak dose rate that would be obtained with the direct disposal of spent fuel.

As can be seen in Fig. 3, the dose rate is dominated by technetium and iodine at the time of peak dose rate. The remaining actinide elements are still important, though, controlling the dose rate after about 350,000 years. Removal and treatment of the technetium and iodine would be necessary to reduce the dose rate for the process waste in this case, although it should be emphasized that the estimates of dose rates for releases from the Yucca Mountain repository are subject to changes as the science and modeling of the repository continue to evolve. This result emphasizes that the overall effect of processing and recycling strategies is to lower the inventory of certain elements in the waste stream, lowering both the decay heat and the projected peak dose rate. The lower decay heat allows for increased utilization of the repository. However, if the repository loading is increased, the projected dose rate will also increase. The decision on how much to increase the loading of the repository is a question of the tradeoff between the acceptability of the resulting peak dose rate and the cost of designing and constructing additional repositories.



Figure. Estimate of the Dose Rate for Releases from the Yucca Mountain Repository with Processing of Spent Fuel to Remove 99.9% of the Actinides, Cesium, and Strontium, and the Resulting Waste Densified by a Factor of 100 and Placed in a Glass Waste Form.<sup>3</sup>

More analysis, discussions and conclusions can be found in the full ANL report (ANL-AFCI-161)

#### **Repository Performance Assessment Analysis (ANL)**

Several performance assessment calculations were carried out to investigate the impact on estimated peak dose rate from the combined effect of actinide removal and repository drift loading increase using spent PWR fuel with a burnup of 51 GWd/MTIHM. In the performance assessment calculations, it was assumed that the spent fuel is processed to remove 99.9% or 99%

of the actinides. The process waste is immobilized in borosilicate glass and drift loading increase factors of 10 and 100 were considered. As a reference case, the spent nuclear fuel was processed and the entire radionuclide inventory was immobilized in borosilicate glass, resulting in the peak value of the mean dose rate (based on 1000 realizations) increasing by 43% relative to the direct disposal of the spent PWR fuel.

Removal of 99.9% of the actinides reduces the peak dose rate relative to direct disposal by a factor of 100, where the largest contributor to the peak dose rate is <sup>99</sup>Tc. Since previous work had shown that removal of this quantity of the actinide elements would allow a potential increase in repository drift loading of up to a factor of 225 based on thermal considerations, several drift loading increases were studied. Using a factor of 10 for the repository drift loading increase raised the estimated peak dose rate by a factor of 10, as compared to the case with 99.9% actinide removal and no increase in drift loading. Using a factor of 100 increased the peak mean dose rate by a factor of 100 results in an estimated peak dose rate approximately equivalent to the peak dose rate resulting from the direct disposal of the spent PWR fuel. In addition, the effect on the estimated peak dose rate appears to be directly proportional to the repository drift loading increase.

In the case where actinide removal is only 99% with no increase in repository drift loading, the peak mean dose rate relative to direct disposal of spent PWR fuel is reduced by a factor of 40. Increasing the repository drift loading by a factor of 10 led to a peak dose rate that is about a factor of 5 lower than for direct disposal of the spent PWR fuel. A further increase in repository loading of a factor of 100 (about what would be allowed based on thermal considerations) led to a peak dose rate about 40% higher than for direct disposal. In contrast to the previous case, the largest contributor when 99% of the actinides are removed is <sup>237</sup>Np.

#### Processing Requirements Analysis (ANL)

Evaluation of repository impacts produced a report: "Processing Requirements for PWR Spent Fuel to Reduce the Estimated Peak Dose Rate Associated with Releases from a Geologic Repository" by R.A. Wigeland and E.E. Morris, ANL-AFCI-166<sup>4</sup>. This report examines the potential reduction in long-term dose of removing individual radionuclides, at varying levels of efficiency, from the inventory of a geologic repository.

#### **Dose Rate Reduction from Reprocessing and Recycling**

The proposed Yucca Mountain repository is used as an example of a geologic repository for the purposes of evaluating the impact of reprocessing and recycling on repository performance. The modeling approach created by the Yucca Mountain Project for assessing the performance of a geologic repository is the Total System Performance Assessment (TSPA). TSPA models typically include all phenomena related to the degradation of waste packages and contents as well as solubility and transport phenomena to arrive at estimates of dose rate for affected individuals. For this study, a simplified TSPA model developed for DOE was used, based on the TSPA model for the Site Recommendation phase of the Yucca Mountain project.

A reference case for this study was established, based on direct disposal of 70,000 MTIHM of spent PWR fuel. All subsequent calculations reflecting the processing and removal of certain chemical elements from the waste are normalized to the peak dose for the reference case. Since the peak dose rate is determined by several actinide isotopes, as well as isotopes that result from the decay of actinide isotopes, evaluations were made for removal of individual chemical elements as well as groups of elements from the waste. The results are summarized in Table 1.

## Table 1.Estimated Peak Dose Rate Normalized to the Direct Disposal of PWRSpent Fuel; Elements Separated from Spent PWR Fuel at 99.9% Efficiency4

	Normalized	Peak Dose Rate
Element(s) Removed	Peak Dose Rate	<b>Reduction Factor*</b>
None	1.550	0.65
Uranium	0.852	1.17
Plutonium	0.835	1.20
Americium	1.473	0.68
Neptunium	1.525	0.66
Uranium &Plutonium	0.199	5.03
Uranium & Americium	0.765	1.31
Uranium & Neptunium	0.823	1.22
Plutonium & Americium	0.749	1.33
Plutonium & Neptunium	0.806	1.24
Americium & Neptunium	1.395	0.72
Uranium, Plutonium & Americium	0.092	10.9
Uranium, Plutonium & Neptunium	0.163	6.13
Uranium, Americium & Neptunium	0.700	1.43
Plutonium, Americium & Neptunium	0.666	1.50
All Actinides	0.011	91.0

\*The reduction factor is for the peak dose rate in each case compared to the peak dose rate for the reference case of direct disposal of PWR spent fuel assemblies. Reduction factors less than 1.0 indicate that the peak dose rate is higher than the reference case.

As shown in Table 1, in order for reprocessing and recycling to have a significant impact on the estimated peak dose rate, it is essential for all actinide elements to be removed from PWR spent fuel, including uranium. In principle, with 99.9% separation efficiency of the actinides from spent PWR fuel, a reduction in estimated peak dose rate of a factor of 91 is possible.

Given that it is possible to greatly reduce the estimated peak dose rate from the repository by reprocessing and recycling the actinide elements from PWR spent fuel, there are several options for using this capability. While Yucca Mountain was used as the example of a geologic repository for this study, the conclusions should apply to other repositories of a similar nature, especially where the same radioactive isotopes dominate the dose rate.

First, it is possible to decide to take advantage of the much lower peak dose rate to decrease the hazard associated with the geologic repository that would contain the waste from 70,000

MTIHM of PWR spent fuel instead of 70,000 MTIHM of PWR spent fuel assemblies. The estimated peak dose from such a repository would be about 100 times lower than for the direct disposal of PWR spent fuel. The desirability of this approach would depend on any benefit that might accrue for siting such a geologic repository as compared to siting a repository for the direct disposal of PWR spent fuel assemblies.

Alternately, if the estimated peak dose rate from a repository with the direct disposal of 70,000 MTIHM of PWR spent fuel assemblies is acceptable, one could take advantage of the much lower peak dose rate associated with the process waste, along with the much lower decay heat, to greatly increase the amount of process waste that could be placed in the repository. Based on the example listed in Table 1 with 99.9% separation efficiency, it would be possible to dispose of the waste from approximately 7,000,000 MTIHM of PWR spent fuel (approximately 3500 years of nuclear power operation at the current generation rate) and have essentially the same peak dose rate as for 70,000 MTIHM of PWR spent fuel assemblies. The repository temperatures would be slightly lower for the case of disposing of the process waste from 7,000,000 MTIHM as well.

A third option would be to take some advantage from both possibilities, providing for some increase in the amount of waste while also reducing the estimated peak dose rate. As an example, one could place the waste from about 700,000 MTIHM of PWR spent fuel and place it in the repository, while reducing the estimated peak dose rate by about a factor of 10.

#### Summary of FY-06 AFC Waste Stream and Product Storage and Disposal Analysis (INL)

A systematic waste analysis of the fuel cycle began at midyear, to assess each process step and associated facility for waste streams. The types of waste (LLW, LLW-GTCC, TRU, HLW, and SNF) produced are identified at each step, for each major fuel cycle alternative. This identification includes chemical and physical form of the initial waste stream, estimated mass and volume of the initial waste stream, potential waste treatment and contaminant loadings in the final waste form, estimated mass and volume of the final waste form, and expected waste classification, packaging, and type of disposal facility for final waste form. The technical maturity of the treatment options, uncertainty in the estimates and expected costs (when available) is being considered. This work is still in progress.

A brief illustration of the analysis approach being used in this study is presented below. The example selected is a Cs waste stream from aqueous separations; the data provided here represents an initial sketch from a partial literature review<sup>1</sup>. The analysis presented here will be enlarged and refined as more data are collected.

## **Example** Analysis<sup>5</sup>

<u>Select Facility, Reactor, and Fuel Type</u> - Aqueous separations facilities are expected to generate a wide range of wastes (LLW, LLW-GTCC, TRU, and HLW), the relative amounts,

<sup>&</sup>lt;sup>1</sup> The example presented here relies heavily on a recent report: Evaluation of the Use of Synroc to Solidify the Cesium and Strontium Separations Product form Advanced Aqueous Reprocessing of Spent Nuclear Fuel, INL/EXT-06-01377, March 2006, by Julia Tripp and Vince Maio<sup>5</sup>. This report was based on an extensive review of the literature.

compositions, and forms of the waste streams will depend on the types of fuel processed and the processes used for separations. Used uranium oxide fuel from a light water reactor was evaluated assuming five years of storage after removal of the used fuel from the reactor and a burn-up of 51 GWt-day/tonne-HM. The elemental mass fractions in the used fuel (excluding cladding) going into separations would be approximately as shown in the Table 2.

<b>Element or Group of Elements</b>	Mass Fraction 5 Years After Discharge
U	0.934
Np	0.000621
Pu	0.0116
Am	0.000638
Cm	0.0000780
Other Actinides	0.000463
Sr	0.00123
Тс	0.00114
Ι	0.000359
Cs	0.00395
Other Fission Products	0.0459
Total	1.00

Table 2 Mass Fractions of Selected Elements in Used UOX Fuel 5 Years after Discharge<sup>5</sup>

<u>Identify Potential Waste Streams</u> - There will be a large number of different waste streams from the aqueous separations facility, including: burned uranium (LLW), actinides (HLW), Cs/Sr (HLW or LLW-GTCC), Tc/I (HLW or LLW-GTCC), process chemicals (LLW, mixed waste, hazardous waste), QA/QC samples (LLW), personal protection equipment (LLW), and equipment replacement (LLW-GTCC, LLW, or industrial). Separated Cs and Sr will be a significant waste stream from aqueous separation and will be the focus of this example.

<u>Identify Disposition Options for Waste</u> - There are several possible disposition options for Cs/Sr including: immediate disposal as HLW, immediate disposal as LLW-GTCC, store as HLW or LLW-GTCC until it decays and reclassify it as LLW for disposal, store for reuse, and transmute in a reactor.<sup>5</sup> Given the short half-lives and small transmutation cross-sections, transmutation is not a likely disposition option for Cs/Sr. Given current waste classification regulations, the Cs/Sr waste will be considered HLW, but storing it as HLW until it decays and can then be reclassified as LLW for disposal is a possible option. The main Cs and Sr isotopes have half-lives on the order of 30 years, so after approximately 300 years of storage the treated waste could potentially be reclassified as LLW or LLW-GTCC. For this example, waste forms that support direct disposal of Cs/Sr as HLW or storage of Cs/Sr as HLW with subsequent reclassification and disposal as LLW will be explored.

<u>Identify Potential Waste Forms</u> - There are several possible waste forms for Cs/Sr waste including: ion-exchange of Cs/Sr in zeolites followed by immobilization in a cement matrix, immobilization of Cs/Sr in sodium zirconium phosphate, steam reforming of Cs/Sr to produce a mineralized product and immobilization of Cs/Sr in Synroc<sup>5</sup>. The waste form will be required to

immobilize the Cs and Sr and maintain physical integrity for approximately 300 years. In this example, Synroc is examined.

The exact composition of the Cs/Sr waste stream from the aqueous separation process will depend on the specific form of Uranium Extraction Plus (UREX+) process chosen. The waste stream will likely contain organics as well as other inorganic components in addition to Cs/Sr. The organic components will likely have to be removed before the waste stream can be put in a final waste form.

<u>*Quantify Waste Loading and Final Waste Form Properties*</u> - Synroc is a crystalline ceramic composed of natural titanate minerals. There are several forms of Synroc.

"The main titanate minerals in Synroc-C are hollandite  $(BaAl_2Ti_6O_{16})$ , zirconolite  $(CaZrTi_2O_7)$  and perovskite  $(CaTiO_3)$ . Zirconolite and perovskite are the major immobilization hosts for long-lived actinides such as plutonium (Pu) and the rare earths, whereas perovskite is principally for Sr. Hollandite principally immobilizes Cs, along with potassium (K), rubidium (Rb) and barium (Ba)."<sup>5</sup>

Focusing on just Cs for the remainder of this discussion, Cs decays into Ba so the same mineral phase (hollandite) in the Synroc waste form is compatible with the parent and daughter isotope, this favors the long-term durability of the Synroc-C waste form for Cs.

A range of waste loadings have been reported in the literature<sup>5</sup>, including: 1.26 wt% Cs<sub>2</sub>O in Synroc, 5 wt% Cs in hollandite, 7.5 wt% Cs in hollandite, 5.1 wt% Cs<sub>2</sub>O in Synroc-C. A limit of 9 wt% Cs in hollandite has been theoretically calculated. In addition to compositional limits, the maximum waste loading of Cs in Synroc-C may also be limited by the heat generated by Cs as it decays or the heat generation term may place requirements on the physical geometry of the final waste form. A calculation for a 10 wt% Cs<sub>2</sub>O in Synroc for a 100 kg mass with a diameter of 0.35 m yielded a centerline temperature of 300-400°C.

Using the 5.1 wt% Cs<sub>2</sub>O loading in Synroc-C from the Tripp/Maio report<sup>5</sup> and the 3.95E-03 mass fraction of Cs in used fuel 5 years after discharge, the mass of immobilized waste per tonne of U in fresh fuel can be estimated<sup>5</sup>. Assume 1 tonne U in fresh fuel; this yields 3.95E-03 tonne Cs in used fuel 5 years after discharge. If all the Cs ends up in one waste stream then there would be a 4.18E-03 tonne Cs<sub>2</sub>O waste stream. Assuming a 5.1 wt% Cs<sub>2</sub>O loading in Synroc-C would yield 82 kg of Synroc-C Cs waste form per tonne of U in fresh fuel.

As additional data is collected the Synroc-C Cs waste form will further quantified with respect to volume, leachability of Cs, technical maturity of process, and estimated cost. The process briefly presented above will be repeated for each branch of each waste stream for each facility and fuel type as available data and schedule permit, priority is being placed on waste streams with higher mass and more restrictive disposal requirements.

#### FY-06 AFCI Storage and Disposal Analyses & Support to Other Systems Studies

AFCI analyses for Storage and Disposal in FY-06, primarily conducted at ANL, included repository thermal/capacity evaluations in support of other systems studies (and reported elsewhere by those activities). This year includes extension of thermal analysis to 3-D modeling and development of capability for simplified repository performance analysis to examine long-term dose benefits from AFCI scenarios.

#### Support to AFCI 2006 Report to Congress:

The Repository Impacts staff contributed to development of the AFCI March 2006 Report to Congress through development of fuel cycle scenarios objectives as well as supporting repository capacity evaluations.

#### Technical Coordination with DOE-RW/YMP (INL)

There had been significant progress in the cooperation between the DOE-NE AFCI program and the DOE-RW Yucca Mountain Project. This included creation of a joint "Spent Fuel Management Steering Committee" to provide a venue for cooperative analysis of waste management issues for advanced fuel cycles.

Activities of the Steering Committee have been on hold since the summer of 2005 following management changes at DOE-RW. This stoppage was due in part to a perception by RW management that NE activities toward development of an advanced fuel cycle could complicate efforts to achieve repository licensing. While we have always stressed the need for one repository while indicating reprocessing and transmutation could delay the need for additional repositories by a century or more, other parties have misinterpreted a closed fuel cycle as eliminating the need for the first repository. Even without this confusion, changes in the nature of materials requiring geologic isolation could create a "moving target" for licensing.

These difficulties must be overcome. The rapidly improving prospects for industry growth drive a need for NE and RW to work together to understand the impacts of disposal options presented by advanced fuel cycle technology with the joint goal of maximizing benefit to the nation. This includes joint assessment of which materials should be considered for reprocessing and which should not, what degree of separations provides the best cost/benefit balance, which waste forms provide best performance considering volume, radiotoxicity, durability, dose contribution, heat load contribution, storage, transportation and disposal operations, and economics, as well as how repository and fuel cycle operations can best be coordinated.

While planned joint work on this type of effort has been on hold for over a year, recent actions provide an opportunity to restart cooperative activities. Most NE analyses of potential repository impacts have been based on models associated with the total system performance assessment (TSPA). As RW moves forward with the license application (LA), they have substantially updated their models. RW has recently initiated an activity at ANL, to assess likely changes if closed fuel cycle impacts were assessed using the LA models. At the same time, GNEP Systems has initiated development of a comprehensive waste management strategy at INL, which will examine a full range of disposition options for materials which may be separated during spent

fuel treatment and associated waste form options. These two reexamination efforts provide renewed opportunity for collaboration in addressing nuclear waste issues.

Collaboration by technical contributors is only one aspect of the needed RW-NE relationship. Efforts are also needed to expand general communications to ensure both organizations understand each other's drivers and objectives so that reports and public statements are mutually supportive. This is a first step needed to improve trust before true joint development of waste strategies can occur. Communications opportunities continue to be explored at various levels, but require expanded emphasis. These can include personal visits, informal sharing of draft reports, cross-briefings, etc.

#### Yucca Mountain Technical Analysis (LLNL)

A process for interaction with Yucca Mountain project staff at the technical working level had been developed prior to suspension of cooperative actions between DOE-RW and AFCI. This established a pathway for technical information exchange at the working level. It is desired to reopen this activity as an ongoing venue for 'beyond the YMP baseline' technical cooperation between AFCI and YMP.

#### **International Cooperation (LLNL)**

OECD-NEA has an international experts group conducting a study "Impacts of Advanced Fuel Cycles on Waste Management Policy". DOE-NE (AFCI) and DOE-RW (OCRWM) are supporting U.S. participation in this study, primarily by AFCI Repository Benefits staff. This study selected fuel cycle scenarios for analysis, developed flowsheets and defined waste streams from these flowsheets. The group determined to use fuel cycle cost as a common metric to evaluate the variety of waste management impacts. For this analysis, the group developed unit costs for each process and mass flow in the scenarios. Other metrics include repository design issues such as capacity, and repository performance represented by long-term dose calculations.

A dozen scenarios were evaluated including open cycle, single cycle MOX, multi-cycle MOX, Pu+Am LWR recycle, Pu+Am+Cm recycle in fast spectrum metal cooled and gas cooled reactors and recycle with ADS options. Processes include UREX, PUREX and Pyro, and metal, oxide and nitride fuels. Several scenarios include processes directly related to AFCI R&D including UREX+ and Pyro processing, and metal, oxide and nitride fuels containing minor actinides. U.S. participation assured that the results of the evaluation are consistent with AFCI and DOE-RW perspectives. With joint support from AFCI and DOE-RW, LLNL AFCI staff served as Chair of Working Group #3, which is responsible for evaluating the impact of the AFC waste streams on geologic disposal.

The NEA study selected four representative fuel cycle scenarios for most detailed evaluation including repository impacts. These scenarios are:

- Scenario 1a PWR, open cycle, UO<sub>2</sub> fuel (this is the reference scenario)
- Scenario 1b PWR, PUREX reprocessing, single recycling of Pu as MOX
- Scenario 2a PWR, PUREX reprocessing, multi-recycling of Pu as MOX
- Scenario 3c variant 1 GCFR, pyro-reprocessing, carbide fuel.

Because repository impacts vary for different repository types, it was desired to include evaluation of the selected scenarios for four different repository types: bedded salt, saturated granite, saturated clay and unsaturated volcanic rock.

The NEA study was completed during FY-06 and the final report has been published as "Advanced Nuclear Fuel Cycles and Radioactive Waste Management" (NEA 66 2006 05 1P)<sup>6</sup>. The final report addresses the waste, cost and resource impacts of different fuel cycle, including the impact of performance and capacity of four different repository types, including an unsaturated volcanic rock repository. A summary graphic from this report shows the relative scoring of four scenarios against 11 metrics. It can be seen that repository impacts and resource utilization vary significantly, while fuel cycle costs vary by only a small amount, and total costs vary even less.



Comparison of main indicators, once-through cycle (1a) is used as reference. <sup>6</sup> 1b: full LWR park, spent fuel reprocessed and Pu re-used once, 2a: full LWR park, spent fuel reprocessed and multiple re-used of Pu, 3c(1): full fast reactor park and fully closed fuel cycle.

"Advanced Nuclear Fuel cycles and Radioactive Waste Management" (NEA 66 2006 05 1P).

#### Summary of Storage and Disposal Basis for GNEP and Transition to AFC R&D (LLNL)

Midway through FY-06, DOE announced the Global Nuclear Energy Partnership. This is a major new direction for the future evolution of nuclear energy in the US and globally. GNEP reflects the results of AFCI Systems Analysis and technology development in Fuels and Separations in setting a path toward light water reactor fuel recycle and reuse of the Pu and minor actinides in fast reactor fuel. This enables a highly sustainable nuclear energy future at large scale and long term, with minimization of wastes in the nearer term and eventual maximization of resource utilization in the longer term. The results of AFCI Repository Benefits and now Storage and Disposal analyses appear to be factored into the GNEP planning.

During preparation for GNEP announcement, Storage and Disposal staff prepared several versions of a white paper on "Global Nuclear Futures Initiative - Waste Management in a Closed Fuel Cycle - Implications for Yucca Mountain and Other Options". One version of this document is included as Appendix A of this report. In addition, as the FY-06 ends, Storage and Disposal staff are leading the preparation of a GNEP Basis Document on "Integrated Strategy for Nuclear Material Transportation, Storage & Disposal Strategy Under the Global Nuclear Energy Partnership" (in review and revision).

As AFCI prepares to transition to the new role as Advanced Fuel Cycle R&D under GNEP, the Storage and Disposal task is preparing to become the more focused Transportation, Storage and Disposal analysis for GNEP product and waste streams. The objective of the work will be to understand the requirements for GNEP nuclear material handling (packaging, transportation, storage and disposal), and define solutions or pathways to solutions in support of GNEP implementation decisions and eventual GNEP facilities.

#### Summary and Path Forward

Systems analyses of potential Storage and Disposal requirements for Advanced Fuel Cycles have been conducted and coordinated with other systems studies and AFCI elements. The repository thermal capacity analysis and repository dose calculations continue to be used to guide technology selection, planning and management decisions. Cooperation with DOE-RW has been put on hold at DOE. The results from recent years waste management analysis served as part of the basis for a major DOE initiative in the Global Nuclear Energy Partnership. This will serve to focus future analysis, while creating more urgency to demonstrate solutions to nuclear material transportation, storage and disposal to support near-term decisions to proceed with GNEP technologies.

In FY-07 the Storage and Disposal activities will be expand to include transportation issues, and will focus on waste and product streams for GNEP technologies.

#### References

- "AFCI Repository Impact Evaluation Report: Systems Analysis Progress Report FY03", W. Halsey, UCRL-ID-155287, Lawrence Livermore National Laboratory, Livermore, CA,
- 2) "40 CFR 197 Public Health and Environmental Radiation Protection Standards for Yucca Mountain, Nevada,; Proposed Rule", US Environmental Protection Agency, August 2005.
- "Update Report on the Repository Benefits of AFCI Options," ANL-AFCI-161, by R.A. Wigeland, T.H. Bauer, and E.E. Morris<sup>3</sup>, Argonne National Laboratory, Argonne, IL. September 2005.
- 4) "Processing Requirements for PWR Spent Fuel to Reduce the Estimated Peak Dose Rate Associated with Releases from a Geologic Repository" by R.A. Wigeland and E.E. Morris, ANL-AFCI-166, Argonne National Laboratory, Argonne National Laboratory, Argonne, IL. April 2006.
- 5) "Evaluation of the Use of Synroc to Solidify the Cesium and Strontium Separations Product form Advanced Aqueous Reprocessing of Spent Nuclear Fuel", INL/EXT-06-01377, March 2006, by Julia Tripp and Vince Maio, Idaho National Laboratory, Idaho Falls, ID. 2006.
- 6) "Advanced Nuclear Fuel Cycles and Radioactive Waste Management" (NEA 66 2006 05 1P). Nuclear Energy Agency, Paris, France, December 2005.

#### APPENDIX A White Paper:

## Global Nuclear Futures Initiative Waste Management in a Closed Fuel Cycle Implications for Yucca Mountain and Other Options

## **Executive Summary**

An important factor in consideration of advanced nuclear energy technology is the waste management implications. With the projected increased use of nuclear energy for longer times, there will be more spent fuel to be managed. However, with closure of the fuel cycle through reprocessing and reuse of spent fuel, there is tremendous potential to minimize nuclear waste, and optimize the use of geologic disposal capacity. Analysis indicates that for a closed nuclear fuel cycle, with efficient fuel reprocessing, use of fast-spectrum reactors and optimized management of the different waste streams, it is possible for one geologic repository to manage the high-level waste from a growing U.S. nuclear energy enterprise for the rest of this century. This represents a large increase in the value of a repository to society.

A sustainable closed nuclear fuel cycle produces wastes that are quite different from a disposal perspective than our current open cycle. While a closed cycle still requires geologic disposal, the nature of the waste management challenge is much different. There is much less long-term heat output due to fissioning of actinides as recycled fuel in reactors and short-term heat output can be managed separately, and long lived fission products and residual actinides can be disposed of in waste forms optimized for long-term isolation.

The current Yucca Mountain repository is designed for effective disposal of both spent nuclear fuel and high-level radioactive waste. It is important for the anticipated growth of nuclear power in the U.S. that the repository be licensed, built and operated as aggressively as possible. Fortunately, the repository site and design appear to have flexibility to evolve to efficiently accommodate the wastes from a closed cycle - if and when these technologies are deployed. With continued growth of nuclear energy use, through 2100 there could be a ten-fold increase in the generation of spent fuel over the current legally defined capacity for the repository. With an efficient closed fuel cycle, it appears the technical capacity of the repository, as measured by repository thermal design goals, long-term performance, and design volume, could easily be increased by ten-fold, and probably much more.

Therefore, it is compelling to pursue parallel but complementary paths to A) follow through on the current Yucca Mountain baseline and B) develop and deploy advanced nuclear technology including a sustainable closed fuel cycle.

## 1.0 Current Waste Management and Yucca Mountain Design Basis

Current U.S. waste management practice is once-through use of oxide fuel, interim storage at reactor sites, and eventual transport to a mined geologic repository for permanent disposal.

#### 1.1 Current SNF and HLW waste streams and waste form characteristics

YM is being designed to dispose of 63,000 MTHM of PWR and BWR spent nuclear fuel assemblies that have been used in a reactor, stored underwater or in dry casks for decay of short half-lived radionuclides, and then transported to YM without any reprocessing. YM is also being designed to dispose of 7,000 MTHM-equivalent of US Navy reactor fuel (not reprocessed), DOE-owned spent nuclear fuel (not reprocessed, primarily from research reactors), and high-level waste glass (containing radionuclides remaining after reprocessing commercial spent nuclear fuel at West Valley and reprocessing DOE production reactor spent fuel at Savannah River and Hanford, and processed naval reactor fuel at INL).

The 70,000 MTHM capacity of YM was established by the Nuclear Waste Policy Act (NWPA). Waste beyond the legislated capacity is currently designated for disposal in a second repository, although Congress could increase the YM capacity by amending the NWPA. Using the same design and waste characteristics, the YM site with additional characterization could dispose of about twice the legislated capacity. The next ridge to the north (Jet Ridge) has similar characteristics and with additional characterization could potentially have a capacity similar to YM. If Jet Ridge proved equally suitable for geologic repository purposes, there is a potential to raise the overall site capacity to 200,000 to 300,000 MTHM.

	Design Basis 21-PWR	Nominal 44-BWR	5-HLW-Short
Emplacement	11,800 W	7,380 W	2,980 W
50 yr	4,950	3,020	871
100 yr	2,930	1,710	323
300 yr	1,410	800	32
1000 yr	599	356	6
10,000 yr	147	94	2
100,000 yr	12	7	0.5

The thermal power of the waste decreases with time due to radionuclide decay:

## **1.2 YM repository – configuration, design bases, performance**

The YM repository layout is configured to meet the design thermal goals shown below, cost effectively, with the design basis waste stream described in Section 1.1. The YM repository design's usage of characterized disposal volume is determined by thermal limits (design goals). The current YM thermal design goals, and the rational for each, are listed below and illustrated in Figure 1.

- 350°C peak sustained cladding temperature to limit creep rupture
- 300°C peak sustained waste package temperature to limit Alloy-22 transitions to less-corrosion-resistant phases
- 200°C peak drift wall temperature to limit rock mineral transitions to morevoluminous phases (that increase rock stress)
- 96°C rock pillar temperature to preserve the site's capability to drain infiltrating water between the waste emplacement drifts)



#### Figure 1: Thermal Design Goals for YM Repository.

Thermal evolution of the repository is a transient heat and mass transfer process that is dominated by thermal conduction. Peak temperatures are limited by ventilation during the time the short-lived radionuclides have not yet decayed.

Subsurface construction costs are reduced by emplacing waste packages end-to-end in drifts. The end-to-end emplacement reduces complexity because the heat source is similar to a line source, reducing the importance of 2D near-field processes and allowing 2D analyses (in the plane perpendicular to the drift axis) for a number of repository submodels. Waste package costs are reduced by using large capacity packages, which can hold up to 21 PWR or 44 BWR spent nuclear fuel assemblies. To meet the design thermal goals with end-to-end emplacement in drifts, the drifts must be widely spaced.

Dose performance for a 10,000-year regulatory period is based on a Total System Performance Assessment (TSPA) using three probability-weighted scenario classes (nominal, igneous, and seismic). There are two probability-weighted modeling cases within each scenario class (early waste package failure and corrosion failure, igneous intrusion and volcanic eruption, and seismic ground motion and seismic fault displacement, respectively). The seismic ground motion and igneous intrusion modeling cases produce most of the dose, with significant margin to the 15 mrem/yr standard during the first 10,000 years.

The recent proposed amendment to the EPA standard, in response to a U.S Court of Appeals ruling in 2004 requires that the Yucca Mountain site be held to a standard that is protective of human health for the next 1,000,000 years (to a time beyond the peak of the dose for the current design). This proposed standard is based on background radiation levels, rather than the lower values used for the 10,000-year standard. Significant contributions to the dose are expected from actinides such as Np-237, Pu-239, Pu-242, Th-230, and Ra-226. Model predictions are being updated to ensure that predictions are valid for this longer time frame and that the latest information relevant to assessing the peak dose is fully considered. The beneficial impacts of a

closed fuel cycle on the peak dose performance and the reliability of these very long-term predictions is discussed below (Section 3.3).

## 2.0 Fast-reactor Closed-cycle Waste Streams

With a mix of light-water reactors, fast-spectrum reactors and fuel reprocessing, the requirements of radioactive waste management are fundamentally changed.

#### 2.1 Nuclear energy scenario for sustainable fuel cycles

While many future nuclear energy scenarios are possible, a moderate growth scenario can be used to illustrate the important issues. This scenario includes growth from current 100 GWe to 300 GWe by 2050. It increases the nuclear energy contribution modestly from the current 20% of US electric generation. The majority of reactors are assumed to be Gen-III LWRs with once-through oxide fuel with 50 MWday/ton burnup. Fuel is cooled 10-25 years before reprocessing. If nuclear growth continued at this modest growth rate, this scenario would imply approximately 750,000 tons of reactor fuel used by 2100.

The scenario assumes 99% efficient UREX+ reprocessing of spent LWR fuel, such that

99% of short-term-heat-producing Cs and Sr are extracted for separate management (Section 5). The minor actinides and 99% of the Pu are extracted for use in fast reactor fuel. Uranium is recycled in fast reactor fuel, stored for future use in fuel, or disposed of separately (Section 5). The scenario also assumes that sufficient fast reactors are built to use the fissile radionuclides extracted from LWR-fuel reprocessing. The fast reactor fraction in new construction and the fast reactor conversion ratio can vary to accommodate evolution of the reactor mission, the nuclear growth rate, and the recycle versus uranium supply economics. Finally, the scenario assumes that fast reactor fuel could be metal, oxide or nitride depending on reactor design. It would include uranium plus fissile radionuclides from the reprocessed mix of LWR and FR fuel. The same reprocessing assumptions apply for FR fuel as for LWR fuel, although process details depend on FR fuel type. Note that FR fuel will contain more minor actinides than LWR fuel; therefore, the same separation efficiency will result in some increase in fissile loss to disposal. Relative efficiencies are not yet known.

#### 2.2 Closed-cycle waste streams and waste form characteristics

Waste streams from the closed fuel cycle are significantly different from the current oncethrough cycle. Many important components of spent fuel are made available for separate, optimized management or are reused until consumed. The removal and reuse or separate disposal of uranium removes 94% of the mass and most of the volume of spent fuel. The separate management of Cs and Sr removes more than 90% of the short-term heat output that dominates current early time repository loads. The reuse of most actinides removes most of the long-term heat, removes most of the long-term dose contributors, and eliminates all criticality issues. The waste forms can be optimized for different components of the waste, potentially providing long-term performance improvements. For example, long-lived fission products and residual actinide reprocessing losses can be immobilized in glass, ceramic, or metallic waste forms.

#### 2.3 Potential waste streams from a proliferation resistant, bring-back fuel cycle

If the U.S. supports a global nuclear energy approach with a secure fuel supply and proliferationresistant enhancement of a 'user-supplier' paradigm, then additional capacity for storage, reprocessing and disposal of waste from a larger reactor base could be appropriate.

## **3.0 Geologic Repository Impacts of Closed Cycle**

Transition from an open to closed fuel cycle offers potential benefits in repository capacity, design options, and long-term performance. It is expected that disposal of closed-cycle waste will be within the same, or similar, regulatory envelope as direct disposal of open-cycle waste. Closed cycle waste offers the opportunity to expand the repository design envelope, to increase the repository capacity, and to improve the repository performance by reducing the radioactive dose rate at the accessible environment. However, some of these opportunities are mutually exclusive. For example, some design flexibility options and repository capacity expansions are mutually exclusive. The extent of this trade-off is complex and not yet evaluated.

#### **3.1 Expanding the repository design envelope**

Waste streams from a closed fuel cycle are separated into several components. Some, such as the Pu, selected minor actinides and much of the uranium, are recycled as fuel and converted into fission products. Other waste streams can be individually managed with optimized waste forms and a wide variety of thermal management alternatives.

With transition to waste streams representative of a closed fuel cycle:

- The cladding thermal limit is not applicable.
- Spacing of line-loaded drifts can be much less than for the current waste stream, and still meet the design thermal goals. A panel design, in which drifts within widely spaced panels are spaced as closely as possible (three drift diameters from a rock stability perspective), is feasible. In a panel design, the smaller pillars between drifts within the panel can be allowed to heat above the boiling point of water, while the larger pillars between panels are subjected to the sub-boiling design thermal goal to drain infiltrating water through the repository elevation.
- Waste packaging currently is long cylinders, based on the length of spent nuclear fuel assemblies. Because reprocessed waste can be in other geometries that can include square cross sections and shorter lengths, it may be feasible to modify the in-drift design to more completely fill the drift with waste than for the current long-horizontal waste-package concept. This flexibility could compensate for the likely lower quantity of waste radionuclides that could be placed in a current waste package.
- Probability-weighted repository doses at the accessible environment are dominated by low probability (infrequent) igneous intrusions and by low probability (infrequent) large-

energy seismic events. Reduced early-time thermal output opens the opportunity to mitigate igneous and seismic event consequences by backfilling the emplacement drifts. For backfilled drifts, the post-closure drift wall temperature limit is not applicable (since rockfall is of no dose consequence with backfill) and the extent of elevation of the waste package temperature with respect to the drift wall temperature (compared to the no-backfill design) is decreased due to the lower waste package thermal power. Backfill would reduce the extent of flow of magma along drifts intersected by a volcanic dike. Finally, backfill would dampen waste package motion due to seismic accelerations, reducing mechanical impacts between translating waste packages that could lead to high residual stresses in the waste package and ultimately to stress corrosion cracking and waste package failure

#### **3.2 Potential for expanded repository capacity**

Recycle of most of the actinides from spent fuel dramatically reduces the long-term integrated heat output from the waste. Furthermore, if it is decided to remove and separately manage the Cs and Sr, then the majority of the short-term heat output is also eliminated. The decrease in both the short-term and long-term thermal loads of closed-cycle waste greatly reduce the intensity of the repository thermal transient and create the potential for dramatic increases in the capacity of the repository in a given area or volume without exceeding repository design thermal goals.

Due to the complex nature of a repository, there is no single, simple measure of technical capacity. However, with reasonable working assumptions, the capacity potential can be estimated for waste streams characteristic of closed fuel cycles. The AFCI program has conducted and published a number of such analyses in recent years. The important conclusions are summarized here, and illustrated graphically in Figure 2. The drift wall temperature limit is dominated by early-time heat from Cs and Sr, and the center-pillar (mid-way between drifts or panels) temperature limit is dominated by long-term heat from actinides.

With removal of only Pu and minor actinides, capacity increases (per unit of repository area) of a few times are possible, while the decay heat from Cs and Sr continues to require extended ventilation and creates a significant early time thermal transient. With removal of Pu, Am and Cm, and separate management of Cs and Sr, capacity increases of tens of times are possible. With high-efficiency removal of all actinides and of Cs and Sr, capacity increases of over one hundred times are conceivable.



Figure 2: Example Calculation of the Potential Increase in Repository Drift Loading as a Function of the Efficiency of Removal in Reprocessing of the Actinides (Plutonium, Americium, Curium), and Short Lived Heat Producing Fission Products (Cesium, and Strontium). (ANL-AFCI-161)

It is important to note that such capacity increases are theoretical, and require further work to be fully understood. However, considering a geologic repository as a valuable national asset, such increases represent a tremendous potential increase in repository value to society.

It appears plausible that a closed fuel cycle can readily accommodate the waste from a growing nuclear energy program (perhaps 750,000 tons of fuel throughput) until the end of this century in a single geologic repository. However, it should be noted that direct disposal of even limited amounts of intact spent fuel on an ongoing basis within future fuel cycles can reduce the capacity increase significantly. Fast reactor fuel and recycled LWR fuel (MOX-if used) contain larger actinide content and would be the most limiting if disposed without reprocessing. Older LWR once-through fuel likely to be initially emplaced at YM would be the least limiting.

#### **3.3 Improvement in long-term repository performance**

A closed fuel cycle removes most of the uranium and fissions most of the actinides normally present in spent fuel. This dramatically reduces the repository source term and offers potential improvement in long-term repository performance, in addition to the improvements in performance driven by a backfilled design, as discussed in Section 3.1.

As with capacity, there is no single simple metric to fully describe the performance impacts from changes in waste stream. The regulatory requirements and the licensing safety case are based on estimates of future potential doses to a population about 16 km from the repository. This estimate is radionuclide-specific and is developed from complex performance assessment

calculations involving mechanistic and stochastic representations of the features, events and processes envisioned for the future evolution of the repository following permanent closure. For screening purposes and general guidance, a simpler set of metrics and analyses may be used to bound the potential performance benefits, as discussed below.

Tc-99 (followed closely by I-129) dominate dose up to about 50,000 years, with the onset and shape of the initial dose rise determined primarily by waste package lifetimes of well over 10,000 years and by the relatively fast rate of mobilization of the waste after waste package failure. By contrast, the transport times of these radionuclides through the natural barriers between the repository and the downstream accessible environment will probably be relatively short (on the order of 1,000 to 10,000 years). Therefore, a key to improved long-term performance is to reduce the mobilization rate of these radionuclides and to minimize their escape from the engineered barrier system. To reduce mobilization rates in the context of a closed fuel cycle, these two separated fission products could be placed into an optimized, robust engineered waste form. If the degradation rate of this waste form is significantly slower than for intact spent fuel, a significant performance benefit would result. To first order, reducing the mobilization rate would provide a one-for-one reduction in the predicted dose contribution of these radionuclides. Similarly, the use of specialized backfill materials focused on these fission products could beneficially impact performance.

The peak dose, which is predicted to occur more than 100,000 years after emplacement for the current waste stream and repository design, is dominated by Np-237 (followed by Pu-242 about a factor of 5 to 10 lower). A closed fuel cycle eliminates much of the Np-237, including its precursors Am-241 and Pu-241. Use of the majority of these actinides as fast reactor fuel enables a potential reduction in the long-term peak dose. At the limit of complete elimination of these actinides, the peak dose will be controlled by the fission products Tc-99 and I-129 that dominate the early dose, and the steps discussed above to mitigate the releases of these radionuclides will also reduce the peak dose. At longer times and lower doses than from Np, decay chain daughters such as U-234, Th-230 and Ra-226 become significant dose contributors. In the closed fuel cycle, these are reduced in the repository along with their precursors, primarily U-238 and to some extent Pu-238. It should be noted that the separate management of separated uranium will have to consider these radionuclides to the extent that uranium is disposed rather than utilized. With reduction of many of the problematic radionuclides with long half-lives, the resultant simplification of the prediction problem may also enhance the credibility of the result, which is a critical benefit, given the unprecedented time frame under consideration, particularly with respect to the new proposed peak dose standard. Rather than having to project models of environmental transport of actinides such as NP-237 and Pu-242 out over time periods of a million years, the modeling problem becomes one of ensuring that the mobilization rates for the long-lived, relatively mobile fission products are reduced at the source. This development would alleviate one of the program's most vexing problems, namely the development of credible predictive models for extraordinarily long time scales.

There are a number of other potential benefits from a closed fuel cycle that could influence longterm isolation performance. These include optimization of waste forms, heat management, waste package design optimization, the number of waste packages needed, criticality control, and density of repository emplacement (influences the total water flux contaminated). Specific performance based metrics have not yet been defined for these issues.

## 4.0 Technology Timing and Evolution to a Sustainable Fuel Cycle

There is a mismatch between the schedule for repository development and deployment of a closed fuel cycle. The repository is nearly ready for license application while deployment of significant fuel reprocessing and fast-spectrum reactors will take 1-2 decades. However, this does not imply disruption to either program. It is quite feasible for both programs to proceed, and evolve together smoothly as they mature.

The repository program can proceed on current baseline, with added analysis to assure that nothing precludes transition to disposal of closed fuel cycle wastes. The YMP mission can be amended as advanced technology is deployed. Proceeding with YM facilitates both deployment of new reactors and reactor types, and assures the developing reprocessing program of a destination for its wastes. As reprocessing is deployed, the repository mission can transition from direct fuel disposal to disposal of reprocessed waste. Retrieval of spent fuel already placed in the repository is an option for future decision-makers, and can serve to both increase repository capacity and to provide a source for fast reactor fuel. Fast reactor deployment could initially follow the availability of fissile material from reprocessing to fabricate initial cores. If greater growth rate is desired, either more LWRs and reprocessing plants can be built, or fastspectrum reactors can evolve to higher conversion ratios to breed additional fissile fuel. Future economic drivers can control this long-term evolution.

#### 4.1 The Need for Geologic Disposal, What and When

We have seen in the sections above that geologic disposal of closed fuel cycle wastes is quite different, and in many ways more flexible and efficient, than direct disposal of spent fuel. However, there remains a vital necessity to have an assured geologic disposal pathway for the reprocessing wastes that contain long-lived fission products and residual actinides (losses during reprocessing). The timetable for six parts of a future fuel cycle often result in confusion. A logical progression for these 'timing events' are:

- Introduction of new LWRs
- Opening a geologic repository
- Beginning LWR fuel reprocessing
- Geologic disposal of reprocessed wastes
- Introduction of fast-spectrum reactors
- Introduction of true 'breeder' reactors.

One key barrier to deployment of significant numbers of new LWR reactors is resolution of the waste management issue. The creation of a repository that can accept LWR fuel resolves this, even if little or no spent fuel ultimately ends up in the repository. Such a repository serves as both 'proof of disposal principle' and 'backup' in case the closed fuel cycle does not materialize. Commercial deployment of new reactors is an important driving force for evolution to the closed fuel cycle, for both resource utilization and waste minimization reasons. Thus, having new

reactor deployment supports the fuel cycle evolution, and the fuel cycle R&D program supports reactor deployment. Fast reactor deployment should logically follow and keep pace with fuel reprocessing – as that is where the fissile material for the FR fuel comes from. High conversion ratio breeder reactors are not needed until the economics of uranium supply drives their deployment, and they then build on the technology of simpler, lower conversion ratio fast reactors that are deployed earlier driven by the fissile material flow within the evolving fuel cycle. To facilitate fuel cycle evolution, the initial repository design and operation should not preclude or complicate future change to disposal of wastes from the closed fuel cycle. It appears that the current Yucca Mountain baseline is quite compatible with future evolution to a closed fuel cycle, but further evaluation should be conducted while the repository is still under development.

## 4.2 Fuel Cycle Optimization

There are many variables in optimization of the fuel cycle, and how it evolves. As a result, there are opportunities buried within such evolution, such as co-location of other fuel cycle facilities near the repository. A national fuel cycle center with interim storage of fuel and wastes co-located with the repository could offer reduced transportation and enhanced security. It would require further analysis to determine if reprocessing and fuel fabrication were more efficiently located near reactors or the repository. The present Yucca Mountain Project resides within the Nevada Test Site, and is surrounded by the Nellis Air Force Range. Secure rail lines are presently being designed to carry high-level nuclear waste (HLW) to the site.

The drifts of the repository could be redesigned to handle specific waste forms and thereby reduce costs of various engineered barriers. Waste forms such as old, cold Cs and Sr (emplaced after interim storage to allow for sufficient decay) could be earmarked for designated drifts with specific engineered requirements (perhaps lesser due to the short half life). The same principles could apply to Tc and iodine waste forms, with enhanced (but more expensive) engineered barriers. Other portions of the repository could be allocated as hardened, secured, storage for waste forms of non-proliferation interest. Integration of the Fuel Cycle and Repository programs could result in a design that could smoothly transition from a pre-closure storage of once-through waste, conversion to store cooler closed-cycle waste, and conversion of the early-storage waste to fuel and re-use of that storage area for closed cycle waste.

## 5.0 Management of Additional Wastes and Other Waste Management Options

## 5.1 Separate management of Cs and Sr

To gain the largest repository capacity increase from a closed fuel cycle, it may be desired to manage the short-lived heat generating fission products Cs-137 and Sr-90 separately. The relative ease of separation, and the 30-year half-life of these radionuclides offers several options for management. Surface storage and decay for 100 to 300 years would permit most of the short-lived radionuclides to decay. The residual Cs would contain long-lived Cs-135 and would require eventual geologic disposal. Development of a custom geologic repository optimized for high-heat, short-lived waste, either at Yucca Mountain or elsewhere, should be considered as part

of the development of the closed fuel cycle. Potential beneficial use of the decay heat from these radionuclides is another aspect that should remain under consideration.

## 5.2 Management of separated uranium

The UREX family of fuel reprocessing is designed to provide a very 'clean' separated uranium stream. This facilitates reuse, storage or eventual disposal of this material as desired. The products of the UREX process can be stored for reuse as fuel in future breeder reactors. Some separated U could be used in fast reactor fuel fabrication; however, it could not all be used until the introduction of high conversion ratio breeder reactors. Custom geologic disposal in a greater confinement disposal facility (GCDF) has been suggested by potential regulators as a possible approach for large quantities of lightly contaminated uranium. It would require a statutory and regulatory framework, but could be as simple as re-interment of uranium in abandoned uranium mines. One GCDF was built and used for contaminated material at NTS.

#### 5.3 Other alternatives

Deep geologic repository disposal of spent fuel and high-level radioactive waste has been selected, and periodically confirmed as the pathway of choice. However, an advanced closed fuel cycle may cause consideration of other alternatives for specific waste streams.

Long-term storage, also referred to as extended or indefinite storage, has been suggested as an alternative to near-term disposal or reprocessing of spent fuel. To the extent that long-term storage is used to connect spent fuel generation to either disposal or reprocessing, it represents interim storage and can help smooth the transition to a sustainable fuel cycle. To the extent that long-term storage is used to justify deferral of either geologic disposal or reprocessing, it represents indefinite storage and does not represent a waste management solution. Interim storage can be used as a bridge from current technology to a closed cycle. Interim storage of spent fuel is already a fact at operating reactors. Expanded storage would provide further flexibility for phasing-in reprocessing in an efficient manner.

There have been suggestions that deep boreholes could be used for disposal of spent fuel, HLW, or specific waste streams. DOE conducted an investigation of deep borehole disposal of excess weapons Pu that offers insight into the strengths and weaknesses for this approach. The deep borehole is not well suited to dispose of wastes with either high heat generation or large volumes. Thus, neither intact spent fuel or HLW containing actinides are good candidates. However, the deep borehole could be an attractive alternative for wastes with modest volume and low heat generation, but high isolation priority. Tc and I are potential candidates that might be effectively isolated in deep boreholes in very tight rock and reducing water chemistry. Other specific waste could also be evaluated for borehole disposal.

## 6.0 Conclusions

Increased use of nuclear energy implies that there will be more spent fuel to be managed. However, with closure of the fuel cycle through fuel reprocessing, there is tremendous potential to minimize nuclear waste, and optimize the use of geologic disposal capacity. While a closed cycle still requires geologic disposal, the nature of the waste management challenge is much different. The current Yucca Mountain repository is designed for effective disposal of both spent nuclear fuel and high-level radioactive waste, and has flexibility to evolve to efficiently accommodate the wastes from a closed cycle if and when these technologies are deployed. With an efficient closed fuel cycle, it appears that the technical capacity of the repository, as measured by repository thermal design goals, long-term performance, and design volume, could easily be increased by ten-fold, and probably much more, and accommodate all U.S. wastes through the end of this century. Therefore, it is compelling to pursue parallel but complementary paths to A) complete the licensing of the current Yucca Mountain baseline and B) develop and deploy advanced nuclear technology including a sustainable closed fuel cycle.

Brief statement of conclusions:

- Increased use of nuclear energy will generate more spent fuel to be managed.
- The proposed Yucca Mountain repository is currently optimized for spent-nuclear fuel and high-level waste.
- A closed fuel cycle still generates long-lived wastes that require geologic disposal, but the nature of those wastes is much different, and in many ways easier to manage.
- The Yucca Mountain site and design can accommodate future evolution for disposal of wastes from a closed fuel cycle.
- Repository technical capacity can be extended greatly for disposal of closed cycle wastes. A factor of 10 times or more appears readily achievable with efficient fuel reprocessing, use of fast-spectrum reactors and optimized management of the differing waste streams.
- One geologic repository could serve the needs of a growing nuclear energy enterprise in the U.S. throughout the rest of this century, and perhaps much more.
- It is compelling to pursue parallel but complementary paths of completing Yucca Mountain while developing a sustainable closed fuel cycle.

**Note of Intent and Disclaimer:** This white paper is intended to explore the range of potential waste management impacts of postulated advanced nuclear energy systems including a closed fuel cycle with reprocessing. By the nature of this postulate, the paper considers technical issues beyond current U.S. policy. To provide an honest technical evaluation of potential impacts, it considers technical issues beyond the current legally established mission for a geologic repository in the U.S. It uses the Yucca Mountain repository as a working example – because that is the geologic disposal system that is best understood. The intent of the white paper is evaluation of technical possibilities, and not to evaluate or recommend policy. It is acknowledged that realization of the impacts discussed would require much more technical evaluation beyond the scope of this white paper, as well as future changes in U.S. policy and law.