# SPENT NUCLEAR FUEL SEPARATIONS AND TRANSMUTATION CRITERIA FOR BENEFIT TO A GEOLOGIC REPOSITORY

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### ABSTRACT

This paper describes the results to date from an ongoing study to establish the chemical separations and transmutation criteria for commercial spent nuclear fuel that would be of benefit to a geologic repository, as measured by the allowable reduction in repository size for a given capacity. The bases for determining the chemical elements to be separated are described, and include both thermal and dose rate criteria. The benefit to a repository is quantified as a function of the separation of certain chemical elements and the efficiency of the chemical separation, along with subsequent transmutation of some of the chemical elements. The proposed repository at Yucca Mountain is used as an example of a geologic repository for the purposes of illustrating the magnitude of the benefits that are possible and the implications for repository size and operation. This work is being done in support of the U.S. Department of Energy Advanced Fuel Cycle Initiative, where numerous reactor, processing, and recycling strategies are being examined to determine the impact on issues important to the viability of nuclear electricity generation, including the impact of spent nuclear fuel and nuclear waste.

#### INTRODUCTION

Geologic repositories, such as the one proposed at Yucca Mountain, are being considered for the direct disposal of spent nuclear fuel and high-level nuclear waste. Such repositories are being designed to safely store hazardous radioactive materials in order to limit releases to the environment for long periods of time, so that the peak dose rates associated with such releases are within limits specified by regulations. In general, to successfully perform this mission, a geologic repository needs to satisfy a number of technical requirements concerning the state of the repository and the materials it contains so that projections about the overall performance of the repository have a reasonable probability of being accurate. The technical requirements are usually related to the tested corrosion, degradation, and release characteristics of the relevant repository materials for ranges of environmental conditions, including temperature and water chemistry. Technical requirements can take the form of specifications for peak allowable temperatures for the repository, specification of materials to be used in the construction of disposal containers (packages), etc. In the example of a repository at Yucca Mountain, for the waste package materials currently specified, a number of temperature limits are imposed for various parts of the repository system. These limits in turn specify the maximum decay heat for each waste package at the time of placement, and the maximum average linear heat rate for the array of waste packages in a repository drift (tunnel). [1]

## **Repository Design: Dose Rate and Thermal Limits**

The goal of any geologic repository is to separate potentially hazardous nuclear materials from the environment, which has been interpreted as limiting the releases of radioactive materials to levels that are not considered a threat to human health. For a repository at Yucca Mountain, the U.S. Nuclear Regulatory Commission (NRC) has established a peak dose limit of 15 mrem/year for people living in the vicinity of the repository, with no more than 4 mrem/year from certain radionuclides in the groundwater. These limits are based on U.S. Environmental Protection Agency (EPA) standards set for individual radiation protection. As part of the consideration, a 10,000 year compliance period was selected for

satisfying quantitative limits on dose rate, with the additional requirement to calculate the peak dose rate using performance assessments if the peak dose rate is expected to occur after 10,000 years. [2]

Peak dose rate can be limited by restricting the rate at which radionuclides can enter the environment and be conveyed to the individual. This can be accomplished in several ways, including limiting the radionuclide inventory and constructing a robust engineered system. For a repository at Yucca Mountain, this approach led to the plans for a repository system with specifications on waste package materials and design, along with loading and operating conditions, to ensure that the repository system is capable of satisfying the dose rate limits. Since the spent nuclear fuel and high-level waste can generate heat for long periods of time, and the performance of the repository can be affected by temperature, many of these specifications are in the form of temperature limits for the various parts of the repository, including the waste packages and the mountain itself.

For example, the requirements for the current reference operating mode of a repository at Yucca Mountain, the high-temperature operating mode (HTOM) of the "cold" repository, include a temperature limit that specifies that the rock temperature midway between the drifts must always remain below 96 °C. This specification ensures that any water flowing downward through the mountain will be able to move through the repository at all times, preventing the retention of a large volume of water above the repository that could flood the repository as it cools. A temperature limit of 200 °C is also imposed on the rock at all times to prevent alteration of its crystalline structure. These temperature limits provide greater certainty about the conditions in a repository at Yucca Mountain, and increased the reliability of the assessments of repository performance. Other temperature limits for a repository at Yucca Mountain apply to the emplaced materials and the waste packages to limit the degradation rates that lead to releases of radioactive materials.

Temperature limits act as constraints on the design and operation of a geologic repository. Meeting these limits can be accomplished by a variety of methods, including controlling the amount of waste in any given area of the repository and actively cooling the repository for an extended period of time. In the case of a repository at Yucca Mountain, the emplacement drifts are 81 m apart, peak and average linear heat loads in the drift are specified, and the repository drifts are cooled by forced ventilation for a period of at least 50 years after completion of waste placement. Since a limit of 70,000 MTHM (metric tons of heavy metal) is legally mandated during the first phase of operation (prior to the opening of a second repository), this amount of spent nuclear fuel and waste will cover approximately 1150 acres, with the area ultimately being determined by the decay heat characteristics of the emplaced waste. The studies described in this paper have been conducted to evaluate the potential increase in drift loading, and therefore the potential reduction in the size of a repository, that would result from processing and recycling spent nuclear fuel.

## Approaches to Benefit a Geologic Repository

In the Advanced Fuel Cycle Initiative (AFCI) program sponsored by the U.S. Department of Energy, numerous reactor, processing, and recycling strategies are being examined to determine the impact on issues important to the viability of nuclear electricity generation, including the disposal of spent nuclear fuel and nuclear waste. As part of this program, studies are being performed to determine if processing spent nuclear fuel to separate certain chemical elements, followed by transmutation of these elements, would be of benefit to geologic repositories by altering the decay heat of the emplaced waste.

For most, if not all, geologic repositories, the material being disposed originates with commercial spent nuclear fuel (CSNF). In the following section, the mass and decay heat characteristics of CSNF are described. Next, the interactions of CSNF decay heat characteristics and repository temperature limits are described and illustrated through the use of detailed thermal models of a repository at Yucca Mountain.

The results identify the temperature limit that controls the drift spacing and loading, and identify the chemical elements in CSNF that contribute to approaching this temperature limit.

Once the chemical elements are identified, it is possible to examine the impact of removing and treating the chemical elements to quantify the associated potential changes in drift loading and repository size. After each processing step to remove certain chemical elements, analyses were performed to calculate the benefit to a repository and the results were examined to identify the next processing path that should be taken to provide further benefit. For those elements where transmutation is possible, the effects of transmutation and the recovery efficiency of the elements to be transmuted are also discussed, along with an evaluation of the impact on the peak dose rate.

# COMMERCIAL SPENT NUCLEAR FUEL DECAY HEAT CHARACTERISTICS

The decay heat generated by CSNF is determined by the mass and isotopic composition of the discharged fuel. In turn, the mass and isotopic composition depends on the discharge burnup of the fuel (GWd/MTIHM). For the example of a repository at Yucca Mountain, many types of CSNF need to be considered in planning the disposal strategy, including both PWR and BWR fuels. In this paper, only the results for PWR fuels are being described for the purposes of illustration. For PWR fuels, which constitute about 55% of the total MTIHM destined for a repository at Yucca Mountain, the average fuel is irradiated to about 41.2 GWd/MTIHM, and has an average age of about 23.1 years since discharge from the reactor.

Fig. 1 shows the decay heat generated by spent PWR fuel at 50 GWd/MTIHM discharge burnup. As shown in this figure, the decay heat drops rapidly after discharge for about the first 200 years. It is important to note that the decay heat is mainly generated by the decay of fission products for the first 60 years, with the contribution dominated by barium and yttrium as decay products of cesium and strontium. After 60 years, the decay heat is mostly from actinide elements, with the important actinide elements being plutonium and americium. Beyond about 200 years, the decay heat is caused almost entirely by the actinide elements plutonium and americium, out to at least 10,000 years. The slow decrease of the decay heat with time is due to the relatively long half-life of the isotopes Am-241, Pu-238, Pu-239, and Pu-240, as plotted in Fig. 1.



Fig. 1 Decay Heat Generated by Spent PWR Fuel Irradiated to 50 GWd/MTHM

In the analyses that follow, these decay heat characteristics are used with a detailed thermal model of a repository at Yucca Mountain to demonstrate the transient thermal performance of the repository. The effects of removing certain elements from the waste stream are then presented.

# THERMAL MODEL OF A REPOSITORY AT YUCCA MOUNTAIN

Yucca Mountain is a mountainous ridge built-up from layers of volcanic rocks (tuffs). The current natural environment is very dry. At the repository level (324 m below the surface and 344 m above the water table on average), the rock layer is fractured, typically about 14% porous, and about 80% saturated with water. The repository within the mountain is an array of parallel storage drifts (tunnels), each about 1 km long and 5.5 m in diameter, located in an approximately horizontal plane. As stated above, the separation between parallel drifts is 81 m for the HTOM. Current plans are for the emplaced waste to be cooled by forced ventilation for at least 50 years after final waste placement. Airflow enters the drifts from both ends and exits at the center. [1]

The thermal model of a repository at Yucca Mountain developed for these analyses simulates the geometry and operation of the repository both prior to and after closure of the repository. Major heat transfer modes of conduction, forced ventilation, free convection, and thermal radiation effects are included. Geometrically, the model describes a rectangular solid "unit cell" surrounding one half of a single storage drift that contains representative heat generating waste packages. Vertically, the model extends from the surface of Yucca Mountain to the water table, explicitly representing all of the rock layers with the corresponding thermophysical properties. Laterally, the model is defined by adiabatic boundaries that extend to one-half the drift separation on either side of the drift, so that the computed temperatures would be representative of values at locations near the repository center where temperatures are expected to be highest.

An explicit configuration of waste packages, drifts, and rock layers is more readily modeled as a network of modular components, boundaries, and connections rather than as a continuum. A single multi-node model spanning a wide range of length scales has been constructed and solved using the SINDA/G [3] thermal network analysis code system. Robust sparse-matrix implicit solvers are able to efficiently accommodate modeled components having very different length scales and time-constants in the same model. These solvers allow time-steps in the calculation to reflect the time-constants of the actual problem rather than that of the smallest node in the model.

Within the various rock layers present in the mountain, conduction is the principal heat transfer mode modeled. Details include specific saturation-dependent thermal properties for each layer. Thermal convection from surface water infiltration into the mountain through the porous rock is also included. Boiling within the rock near heated drifts is modeled in a simplified way by a reversible phase change of water contained in rock fractures or pores.

A centrally positioned 1.67 m diameter heat generating cylindrical surface within a drift represents idealized waste packages and serves as a placeholder for more detailed waste package models that are included as needed. (Drip shields as proposed by the Yucca Mountain Project are not currently modeled.) Waste package heating is assumed to be azimuthally symmetric and axially averaged. Heat exchange between waste packages and drift walls includes convection through the drift during the forced ventilation phase and natural convection within the drift after the forced ventilation phase, combined with thermal radiation interchange at all times.

It is noted that the repository thermal model used for these analyses is simpler than the complex models and schemes developed by the Yucca Mountain Project to simultaneously compute both thermal and hydrological behavior within a continuous porous and fractured rock medium [4]. Coupled thermal and hydrological models make detailed analyses at the scale of waste packages and drifts much more difficult. By contrast, the thermal model described in this paper decouples hydrologic effects by assuming key hydrologic conditions (e.g., rock saturation states and surface water infiltration rates) as measured input. This simplifying assumption enables a single inclusive model calculation that includes all relevant heat transfer modes in realistic ways to compute the entire time-dependent temperature field, from waste package details through Yucca Mountain rock.

Direct comparisons were made with results from the more complex thermal-hydrological models, showing that the present thermal model agrees well even for examples with significant boiling. The good agreement supports the assumptions made in developing the thermal model, and confirms the conclusion stated in Ref. 5 that boiling conditions in Yucca Mountain may strongly perturb hydrologic flow, but the amount of that perturbation will not strongly influence subsequent thermal development.

# THERMAL PERFORMANCE OF A REPOSITORY AT YUCCA MOUNTAIN

As discussed above, the design and loading of a repository at Yucca Mountain is governed by the transient thermal behavior and the appropriate temperature limits to ensure adequate performance of the repository. The following discussion begins with the current reference design, and then proceeds with identification of the chemical element(s) whose decay heat causes the temperature limits to be met. Subsequent sections illustrate the repository performance as groups of chemical elements are removed, and quantify the potential increase in drift loading, i.e. reduction in repository size, that would be possible with such a strategy.

#### Reference Case for a Repository at Yucca Mountain

The current plans for a repository at Yucca Mountain are described in detail in Ref. 1, and define the reference case for these analyses. Overall, the average drift loading of the repository is approximately 1.1 MTIHM/m to satisfy thermal constraints. For this reference case, the transient temperature behavior of the repository was calculated, with the results given in Fig. 2. The decay heat is shown as the corresponding linear heat rate in the drift, and is equivalent to the data in Fig.1. As Fig.2 shows, the waste package surface temperature is initially below the boiling point of water, while the drift is ventilated, but once forced ventilation is stopped (assumed to occur at 75 years after waste placement), the surface temperature increases rapidly to a peak of about 140 °C. The temperature then slowly drops over time, falling below boiling after about 3000 years. The drift wall temperature exhibits the same trends. The temperature midway between adjacent drifts responds much more slowly, since the only mechanism for heating the rock in this location is by conduction from hotter areas around the drifts. As the figure shows, this temperature peaks just below 96 °C, one of the temperature criteria for a repository at Yucca Mountain.

It should be noted that none of the temperatures in and around the drift are close to any of the relevant temperature limits. It is concluded that the temperature limit midway between adjacent drifts is the controlling limit for the reference case, and that the peak temperature in this location occurs at about 1500-2000 years after waste placement. Also, due to the extended time frame for heating this region of the repository, the temperature peak must be the result of the integrated decay heat over the time since the placement of the waste, rather than the value of decay heat at any particular time. Since the figure shows that substantial heating of the interior of the mountain does not commence until the forced ventilation has stopped, the responsible chemical element(s) would logically be those that have the highest integrated decay heat from the time when ventilation ceases until about 1500-2000 years.

Considering the decay heat data shown in Fig. 1, the decay heat is dominated by actinide heating for all times past about 60 years, indicating that one or more of the actinide elements is responsible for the decay heat leading to the temperature peak midway between the drifts. Examining the contributions from each of the actinide elements during the time period from 75 years out to 2000 years, it is observed that the dominant contribution is from the isotope Am-241. Since the isotopic composition of spent PWR fuel at the time of discharge shows very little Am-241, the Am-241 content must be caused mainly by the radioactive decay of Pu-241, with a small contribution from the decay of Cm-245. The Am-241 content peaks at about 80-100 years after discharge from the reactor, due to the 14.4 year half-life of Pu-241, but the 433 year half-life of Am-241 and its energetic decay are the reasons for this isotope's dominant contribution. During the time from 75 years out to 2000 years, the other contributors are the plutonium isotopes, Pu-238, Pu-239, and Pu-240. Fission product heating is essentially gone by 300 years.



Fig. 2 Transient thermal response of a repository at yucca mountain for reference loading conditions of spent pwr fuel and 75 years of forced ventilation

In summary, it would appear that the current plans for the design and loading of a repository at Yucca Mountain are constrained by the integrated decay heat mainly from Am-241, which arises from decay of Pu-241, with the remainder of the decay heat coming from other plutonium isotopes.

# Repository Behavior When Plutonium and Americium are Removed

With the identification of plutonium and americium as being the cause for one of the temperature limits to be reached in a repository at Yucca Mountain, it is possible to examine the effect on the repository of removing these two elements. While the plutonium and americium must be treated in some manner, the discussion of the impact of the disposition of the removed plutonium and americium is deferred until later in this paper.

Assuming that processing of spent PWR fuel is possible, and that the efficiency of removal for plutonium and americium is 99.9%, the transient behavior of the repository for the remaining materials can be studied. Fig. 3 shows the transient temperatures in the repository for this case, and it is apparent that the overall behavior is quite different from that observed for the reference case. The removal of plutonium and americium has made it possible to increase the linear loading in the drift from the reference value of 1.1 MTIHM/m to 5.9 MTIHM/m. (It is likely that in the processing spent PWR fuel, the uranium would be removed. The drift loading is for the waste associated with metric tons of initial heavy metal of the original fuel.) The loading increase can be interpreted as an increase in the area loading of the repository by a factor of about 5.4, which would allow a reduction in repository area by the same amount.



Fig. 3. Transient Thermal Response of a Repository at Yucca Mountain with Removal of Plutonium and Americium from Spent PWR Fuel with Increased Drift Loading

In the example given in Fig.3, it was assumed that the recovery efficiency of the plutonium and americium was 99.9 %. In studying cases with lower recovery efficiencies, it has been determined that the allowable increase in drift loading is a factor of 5.3 at 99%, and 4.3 at 90%. The relative insensitivity of the increase in drift loading to the recovery efficiency of plutonium and americium is caused by a change in the nature of the transient thermal performance, and recovery efficiencies above 99% may not be needed unless other separations are conducted.

Referring to Fig.3, it is seen that the drift loading is no longer limited by a peak temperature midway between the drifts, since this temperature peaks at only 75 °C at about 150 years after waste placement. Instead, the temperature limit constraining drift loading is the 200 °C allowed for the rock, and occurs at the drift wall. The peak temperature occurs at about 90 years, shortly after the end of the forced ventilation period at 75 years, when the repository is closed. This change in the nature of the transient thermal behavior of the repository indicates that the source of the decay heat responsible for reaching the temperature limit is not another actinide element, but would be caused by fission products, as shown on Fig. 1. Since the peak occurs rapidly after closure, this implies that the peak is caused by the integrated decay heat from 75 to 90 years.

Examining the remaining contributors to the decay heat during this time, it is clear that the decay heat is due almost entirely to cesium and strontium, and to their decay products barium and yttrium. These fission products provide almost the entire fission product portion of the total decay heat for times greater than 10 years after discharge of spent PWR fuel from the reactor, which is not placed into the repository until 25 years after discharge.

### **Subsequent Removal of Fission Products**

The fission products cesium and strontium, along with their decay products barium and yttrium, have been identified as being responsible for the decay heat causing the repository to reach one of the temperature limits once the plutonium and americium have been removed from spent PWR fuel. The half-life of barium at 2.5 minutes, and the half-lives of yttrium at either 3.2 hours or 2.7 days, are very short compared to the 30 year half-life of both cesium and strontium. This large difference in half-lives allows a separation strategy where only cesium and strontium are removed, since any barium and yttrium will rapidly decay away. As a result, the next step was to quantify the impact of removing the cesium and strontium, analyzed for various separation efficiencies as with the removal of plutonium and americium.

Sample analysis results are shown in Fig. 4, where the plutonium and americium have been removed with 99.9% efficiency, and the cesium and strontium were subsequently removed, also with 99.9% efficiency. The drift loading has been further increased to 47.0 MTIHM/m, a factor of 42.7 higher than for the reference case. As the figure demonstrates, the transient behavior of the repository has changed again, where in this case the drift loading is limited by the peak temperature of the drift that occurs immediately after placement of the waste, even with the forced ventilation of the repository. The decay heat causing the temperature peak is the result of short-lived actinide and fission product elements, mainly curium and europium.



Fig. 4. Transient Thermal Response of a Repository at Yucca Mountain with Plutonium and Americium Removal and Subsequent Removal of Cesium and Strontium from Spent PWR Fuel with Increased Drift Loading

The results are similar for lower recovery efficiencies of the cesium and strontium. For the same case of 99.9% removal efficiency of plutonium and americium, the drift loading can be increased to 44.0

MTIHM/m for 99% removal of cesium and strontium, and to 29.5 MTIHM/m for 90%. The sensitivity of the results to recovery efficiency indicates that the dominating importance of short-lived isotopes occurs with 99% removal of cesium and strontium, and higher removal efficiencies of these fission products may not be necessary unless other separations are done. While further processing of the waste might be considered, it is important at this point to consider the effects of realistic recycling scenarios for the separated plutonium and americium, and storage strategies for the cesium and strontium. Such scenarios have the potential to introduce more plutonium and americium into the waste stream, and could make further reductions in short-term decay heat irrelevant to repository loading constraints.

# **Recycling of Plutonium and Americium**

Processing spent PWR fuel to remove plutonium, americium, cesium, and strontium to separate them from the rest of the waste would not accomplish anything unless there was a method(s) for treating these elements to render them less hazardous, or to dispose of them in another way. In the AFCI program, various reactor and recycling options are being examined where the plutonium and americium are returned to a reactor and transmuted or fissioned into other elements. For the cesium and strontium, proposals have been made for sequestering the removed fission products in a separate area of the repository, or in alternate locations, since the decay heat problem for these elements only persists for about 200-300 years. At that time, these materials could be placed with no impact on the thermal management problem.

While in principle, it is possible to transmute or fission plutonium and americium in either a thermal or fast neutron spectrum, the recycling option chosen as an example in this study is a fast reactor, with the fast reactor spent fuel being reprocessed using molten salt pyroprocessing technology to recover plutonium and americium for further irradiation. A processing loss of 1% is assumed for the plutonium and americium at each recycle step. The cesium and strontium are separated and are stored separately.

The repository performance obtained from emplacing the resulting waste is shown in Fig. 5. As indicated in the figure, the transient behavior is now similar to that for the reference case, although the drift loading has been increased to 22.6 MTHIM/m, a factor of 20.5 greater than for direct disposal of spent PWR fuel. The drift loading is constrained by the temperature limit midway between adjacent drifts, with the peak temperature occurring past 2000 years, and implying that the integrated decay heat after closure was again responsible for limiting drift loading. Further study demonstrated that it was the same actinide elements, plutonium and americium, that dominate the integrated decay heat. Apparently, the processing losses from the fast reactor spent fuel processing are sufficient to cause the small amount of plutonium and americium in the waste to constrain the repository loading, and make further separations of other elements beyond cesium and strontium. In other words, the loading of the repository is now limited by the ability to separate the relevant chemical elements, where all processing steps must be considered.



Fig. 5. Transient Thermal Response of a Repository at Yucca Mountain with Removal of Plutonium, Americium, Cesium, and Strontium from Spent PWR Fuel, Recycling Plutonium and Americium in a Fast Reactor, with Increased Drift Loading

For cases with better separation of plutonium and americium at the processing steps for both the spent PWR fuel and fast reactor fuel, it should be possible to increase the drift loading up to at least the factor of 42 noted earlier. In principle, if the separation efficiency for both of these fuels were sufficiently high, it would then be useful to consider the separation of additional elements, notably curium. However, it should also be mentioned that the limit on increasing drift loading could be constrained by the ability to make waste forms dense enough to take advantage of the opportunity provided by the low decay heat of the resulting waste.

# IMPACT OF PROCESSING SPENT FUEL ON THE PEAK DOSE RATE

As discussed in the Introduction, the repository is ultimately designed to restrict peak dose rates below specified limits for a given period of time. This goal led to the identification of temperature limits as relevant to determining whether or not the repository would be able to meet this requirement. As a practical matter, given the 10,000 year regulatory time period for a repository at Yucca Mountain, and the robust engineered systems designed into the repository, the estimated peak dose rate is extremely low, around  $10^{-5}$  mrem/year. [6] Therefore, processing to reduce the peak dose rate is not currently relevant to a repository at Yucca Mountain. However, it is instructive to consider the effects of spent PWR fuel processing on longer term dose rates, as required by the NRC.

The peak dose rates from a repository at Yucca Mountain have been estimated at several hundred mrem/year, using detailed performance assessment modeling.[1] More recent analysis results show lower

estimates for the peak dose rate of about 100 mrem/year.[6] The peak dose rate occurs at about 250,000 years or later, and remains above 15 mrem/year for at least 1,000,000 years. Clearly, there is an opportunity to reduce the peak dose rate outside of the current regulatory period.

## **Removal of Actinide Elements**

As discussed in the preceding sections, certain actinide elements would be removed to increase the drift loading in the repository, specifically plutonium and americium. Considering the contributors to the peak dose rate, it is observed that the dominant isotope is expected to be Np-237, especially for times longer than 100,000 years.[1] In studying the isotopic composition of spent PWR fuel irradiated to 50 GWd/MTIHM, it can be seen that some Np-237 is present at discharge, about 1/3 of the eventual total amount. The remaining Np-237 is formed during decay of higher actinide elements, specifically Am-241 (from Pu-241) during the first several thousand years in the repository. If plutonium and americium are being separated and recycled for thermal management reasons, 2/3 of the total Np-237 would already been removed from the waste stream. One could propose a further separation of neptunium from the waste to almost entirely remove this element from the waste stream, depending on the efficiency of the separation. At that point, with uranium, plutonium, americium, and neptunium separated from the waste, there is very little residual dose from the actinide elements. Estimates of the remaining peak dose indicate that there may be a potential to limit the peak dose rate to less than 15 mrem/year at all times.

One could also consider elements which may dominate the dose at earlier times, such as technetium and iodine, but examining the peak dose rates attributable to these elements shows that they are already below the 15 mrem/year limit, and it may not be worthwhile to focus attention on separation and treatment to lower the peak dose rate.

### POTENTIAL BENEFIT FOR A LOWER TEMPERATURE REPOSITORY OPTION

The study has also been extended to the proposed low-temperature operating mode (LTOM) of a repository at Yucca Mountain. The LTOM is defined by imposing the additional limiting constraint that the temperature at the waste package surface must remain below 85 °C once the repository is closed. The technical basis for the 85 °C limit is the assertion that below this temperature, the C22 alloy utilized in Yucca Mountain waste packages will not corrode regardless of the relative humidity within the drift.[1] It follows that Yucca Mountain rock is never above boiling (96 °C) at any time in any location and ensures that the thermal impact of waste storage on the mountain's rock is minimal.

The analysis of cases for the LTOM has shown that the identification of dominant chemical elements and the respective increase in loading from removing such elements is basically unchanged from the results for the HTOM. Plutonium and americium are still the first elements that need to be removed from CSNF to provide a benefit, followed by the fission products cesium and strontium. The amount of benefit, as measured by the allowable increase in drift loading, based on the separation efficiency is similar, but may be somewhat larger in the LTOM case, although this may require moving the drifts closer together to achieve the maximum benefit.

In summary, the separations proposed to benefit a repository at Yucca Mountain for the HTOM are also effective for the LTOM. This result implies that the usefulness of strategies being investigated as part of the AFCI program is independent of some of the decisions being made about designing and operating the repository.

# SUMMARY AND CONCLUSIONS

The study of the response of a repository at Yucca Mountain to the planned disposal of CSNF has identified opportunities for increasing the drift loading of the repository and reducing the estimated peak dose rate. The analyses on the thermal response of the repository have resulted in the following principles that would be applied:

- 1. The dominant contributors to the thermal load of the emplaced waste in 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 drift loading of the repository. 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).
- 2. After the plutonium and americium have been removed, the next elements that need to be considered 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.
- 3. The next most important element is curium. However, in considering a realistic fast reactor recycling scenario for the plutonium and americium, it is observed that the drift loading can be limited by the losses of these two elements from the processing of the fast reactor fuel. In the case where 99.9% of the plutonium, americium, cesium, and strontium are removed from the spent PWR fuel, the 1% loss assumed in the processing of the fast reactor fuel reduces the potential increase in drift loading from a factor of 42.7 to 20.5. This emphasizes the need to reduce losses of plutonium and americium below the 1% assumed in this study before separation of curium will be effective.

In summary, it has been shown that removal of plutonium and americium has the potential for reducing the size of a repository at Yucca Mountain by a factor of 4.3 to 5.4. Combining this with removal of cesium and strontium allows for much greater reductions in size, upwards of a factor of 40, although the use of realistic recycling options for the plutonium and americium emphasizes the need to have very low losses for processing the recycled fuel, and would require the availability of waste forms that could be densely loaded with the remaining waste materials. Examination of the LTOM of a repository at Yucca Mountain shows that the same processing and recycling strategies would be effective for that case as well. This study has quantified the benefits to the geologic repository that would arise from certain spent fuel processing strategies, as would be investigated as part of the AFCI program, with these benefits relatively independent of the specific repository design decisions.

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