IMPACT OF ACTINIDE REMOVAL ON WASTE DISPOSAL IN A GEOLOGIC REPOSITORY

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ABSTRACT

The presence of actinides in spent fuel destined for a geologic repository such as the proposed Yucca Mountain Repository causes a substantial long term heat load, causes the radiotoxicity of the waste to remain high for tens of thousands of years, and contributes significantly to the long-term dose rate once waste packages begin to fail. Examples, mostly based on the current design of the proposed repository, are considered to illustrate the potential impact of actinide removal on each of these factors. The analyses show that removal of 90 to 99% of the actinides may significantly increase in the capacity of a repository. In addition, the radiotoxicity of the waste may be reduced to a value less than that of the uranium ore from which the fuel was manufactured within the 10,000-year regulatory period. For example, removal of 99.9% of the actinides reduces the radiotoxicity to a level less than that of the original ore in less than 400 years. Finally, removal of 99% of the actinides could reduce the peak long-term dose rate, estimated to occur after about 270,000 years, by as much as a factor of 60.

INTRODUCTION

This paper quantifies the impact of actinide removal on the disposal of spent nuclear fuel (SNF). The actinides present in both Department of Energy spent nuclear fuel (DOE SNF) and commercial spent nuclear fuel (CSNF) cause the waste from these sources to remain highly radiotoxic for tens of thousands of years. Moreover, they also cause a long-term heat load and dominate the calculated long-term dose rate from a waste facility such as the proposed Yucca Mountain Repository. Removal of the actinides would significantly mitigate these issues and reduce the uncertainties related to the performance of the repository over geologic time scales. Actinide removal may also allow a geologic repository to accept more waste as a result of the lower initial heat load and the more rapid drop in decay heat.

There are several available processes, either chemical or electrochemical, which are capable of separating the actinides from the fission products in spent fuel. With these processes, most of the recovered material is low-enriched uranium, some or all of which may be separated from the other actinides. The recovered uranium is generally of low activity, and can be stored for future use. The remainder of the actinides, all transuranic elements, need to be treated, such as by irradiating them in a fast neutron spectrum where they will eventually fission, thus reducing them to shorter-lived fission products. The fast neutron spectrum can be produced either by an accelerator driven transmutation system or by a fast reactor.

IMPACT ON HEAT GENERATION

The relative importance of actinides and fission products on the heat generated by SNF depends on the level of burnup and the cooling time following irradiation of the fuel. This paper evaluates the impact of actinide removal on heat generation for generic LWR reactor fuel with 33 GWd/MTHM. Figure 1 shows the heat generation for this fuel as a function of time after discharge from the reactor. Separate curves are shown for various classes of elements. The curve labeled light elements refers to neutron activation products of non-actinide elements present in the structure and fuel at the time of manufacture. According to the figure, by a few hundred years after discharge, the heat output from the spent fuel is completely dominated by actinides and their decay products. At even earlier times, the actinides still account for a significant fraction of the decay heat, such that removal of 99% of the transuranics lowers the heat generation at 100 years by a factor of three.

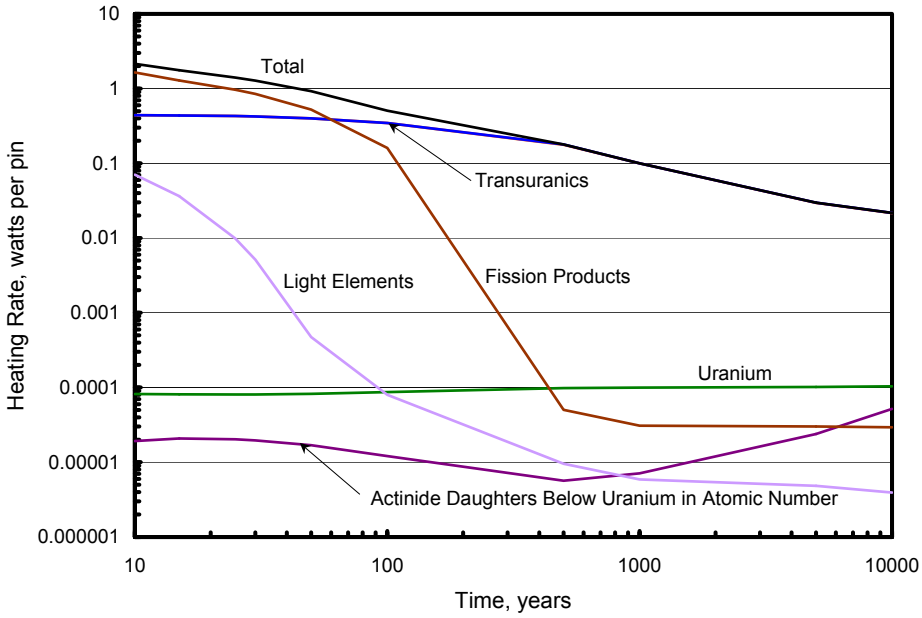

Fig. 1. Heating rate for various constituents of PWR fuel with a burnup of 33 GWd/MTHM.

In addition to lowering the heat generated from the spent fuel, the heat generation rate also declines more rapidly with time as greater fractions of the actinides are removed. Since the design of the repository is currently constrained by the initial heat generation of the spent fuel, any reduction in heat generation, such as from actinide removal, has the potential to change the capacity of a repository.

In the following discussion, the "linear loading" and the drift (tunnel) spacing characterize a given repository design. The linear loading is the number of metric tons of initial heavy metal that can be placed per unit length of drift (MTHM/m). If the fuel has not been treated, then the unit specifically refers to the number of metric tons of initial heavy metal in the fuel. In the case where spent fuel is treated in some manner, the unit refers to the number of metric tons of initial heavy metal in the fuel treated to generate the waste. The areal loading (MTHM/m^2) of the repository can be expressed as the ratio of the linear loading to the drift spacing. The total capacity of a repository is the product of the areal loading and the area. Removal of actinides can change the repository capacity or "footprint" (size) by changing the areal loading. The reduced heat load per metric ton of spent fuel and the more rapid decline in the decay heat allow either changing the linear loading, the drift spacing, or a combination of the two.

Several factors determine the potential impact of actinide removal on the areal loading of the repository. Among these are temperature constraints on the rock surrounding the waste package or on the waste package itself, the length of time the repository is ventilated following emplacement of waste, and the volume of waste after actinide removal compared to the volume before removal. These factors are in addition to the spent fuel burnup and the time after discharge when the fuel is emplaced, mentioned earlier.

Effect on Drift Spacing

The effect of actinide removal on drift spacing is examined, using the assumption that the spent fuel is packaged so that if placed in the repository 10 years after discharge from the reactor, the linear heating rate in each drift would be about 1.4 kW/m.* However, rather than place the fuel or the derived waste at 10 years after discharge, the packages were allowed to cool for an additional 50 years before placement. A distance of 81 m separates the emplacement drifts. Thermal effects were estimated by assuming that the

* This heating rate is about the value assumed for the high-temperature operation of the proposed Yucca Mountain Repository, but the fuel in the reference design is assumed to be about 25 years from discharge.[1]

repository is embedded in an infinite medium of tuff, where the tuff has a conductivity of 2 W/m/K and a thermal diffusivity of 9.8×10^{-7} m^2/s. These properties are characteristic of the tuff in the proposed Yucca Mountain Repository. The repository was approximated as an array of infinitely long line sources, laid out to a distance of 2 km on either side of the point where the temperature was to be estimated. Temperatures evaluated with this model most likely overestimate the actual temperature because of the infinite medium assumption and because potential evaporation and condensation effects are neglected even when the rock temperature approaches or exceeds the water boiling temperature.

For the present example, it is assumed that actinide removal from the spent fuel is used only to change the drift spacing in the repository. Temperatures were calculated as a function of time at a point midway between the drifts for fuel containing actinides and again for the waste that results after some fraction of the actinides were removed. The objective is to keep the peak midpoint temperature below the water boiling temperature. This temperature constraint is intended to allow water that boils in the rock close to the drifts to condense and drain downward between the drifts rather than return to the drifts as the repository cools. After removal of only 90% of the actinides, the calculations show that the spacing between drifts could be reduced by a factor of three with no significant increase in the peak temperature calculated at the midpoint. In the proposed Yucca Mountain Repository, such a reduction in drift spacing is probably feasible since the Viability Assessment considered a drift spacing of about 27 m as its reference case.[1] Since the linear loading is unchanged, this translates to a factor of three increase in the areal loading.

Effect on Linear Loading

The potential impact of actinide removal on the linear loading is examined as a function of the time after discharge when the spent fuel or the waste resulting from the spent fuel is placed in the repository. The objective is to maintain the peak temperature at the midpoint between drifts at the same value that would be obtained when untreated spent fuel is placed in the drifts. As in the previous example, this temperature is maintained below the water boiling temperature. However, in this case the drift spacing is held at a constant value of 81 m and the linear loading is allowed to change. Temperature calculations were performed using the computational model described in the previous example. As in the previous example, all waste is placed simultaneously, but 70% of the heat generated is removed for 75 years following placement to simulate a 75-year ventilation period.[2]

Table I
Ratio of Linear Loading With to
Linear Loading Without Actinide Removal

Time, Years after discharge	Fraction of Actinides Removed	
	0.99	0.999
25	4.6	4.7
50	7.8	8.0
75	13	13
100	20	21

The temperature calculations indicate that for 25-year old fuel with 99% or more of the actinides are removed, the linear heating rate (kW/m) in the drift must be about 3.3 times higher to maintain the same peak temperature in the rock between the drifts. This higher linear heating rate is permitted because heat generation declines more rapidly with increasing time when the actinides are removed. To achieve this increase in the linear heating rate, the linear loading (MTHM/m) must be increased by a factor of 4.6. The linear loading increases by a larger factor than the heating rate because the number of metric tons of initial heavy metal in the fuel must be higher to achieve a given initial heating rate when actinides are removed. As the time of placement, expressed in years after discharge, is increased, the linear heating rate producing the same peak midpoint temperature between the drifts decreases. This is because, as shown in Fig. 1, the heat output from the actinides decreases more slowly with increasing time than the heat output from fission products. For a given initial heating rate, the larger the initial fraction of heat contributed by actinides, the larger the value of the resulting peak midpoint temperature.

Table I lists the ratio of the linear loading (MTHM/m) when 99% and 99.9% of the actinides are removed to the linear loading for untreated spent fuel emplaced at the same time after discharge. The results show that if one waits until 75 or more years after discharge, there is the potential to achieve more than a factor of 10 increase in the linear loading and, hence, in the repository capacity. Whether or not such an increase is achievable depends on the properties of the waste forms that result when actinides are removed as well as on the details of waste package design and the placement of packages in the drifts.

The analysis performed to generate the results in Table I assumes that all waste behaves like generic LWR fuel with 33 GWd/MTHM burnup. However,

the proposed Yucca Mountain Repository is expected to contain a mix of PWR, BWR, and so called co-disposal waste packages. The PWR fuel is expected to have an average burnup of about 41.2 GWd/MTHM and the BWR fuel an average burnup of about 33.6 GWd/MTHM.[2] Analysis similar to that in the second example, but assuming a repository containing a mix of only PWR and BWR fuel in the same proportions as expected in the Yucca Mountain Repository, resulted in linear loading ratios similar to those in Table I. The analysis was repeated with a mix of PWR, BWR, and co-disposal waste packages as currently planned in the proposed repository. No actinides were removed from the co-disposal waste packages. Linear loading ratios at 25 years after discharge were about the same as shown in Table I. The ratio increased with increasing time after discharge and leveled off after about 90 years to a values between 11 and 12.

IMPACT ON RADIOTOXICITY

The impact of actinides on the radiotoxicity has been investigated previously by Hill.[3] Recently, the results have been updated using radiotoxicity factors based on ingestion of radionuclides. The results are expressed in terms of Hazard, evaluated as the number of cancer deaths caused by the ingestion of spent nuclear fuel, divided by the number of deaths caused by the ingestion of the ore from which the fuel was derived. Figure 2 shows the results for generic PWR fuel with 33 GWd/MTHM of burnup. Various classes of radionuclides contribute differently to Hazard. Not surprisingly, the results look similar to those shown in Fig. 1 for the heat output. Those portions of the curves above unity in Fig. 2 indicate a higher number of expected cancer deaths than would be expected from the ingestion of the original ore.

Figure 2 shows that the Hazard of those actinides and decay products with atomic number less than 92 (identified as sub-uranium) is never greater than the unity. However, it takes more than 100,000 years for the Hazard of transuranics to become less than unity. In contrast, the Hazard of fission products, (here also including activation products) becomes less than unity in only a few hundred years.

Table II shows the time required for the Hazard of the waste derived from the spent fuel to become less than unity when various fractions of the actinides are removed. If 99% or more of the actinides can be removed, the Hazard waste becomes less than unity within the 10,000-year regulatory period. Removal of 99.9% or more of the actinides can reduce the Hazard to less than unity within 400 years of discharge.

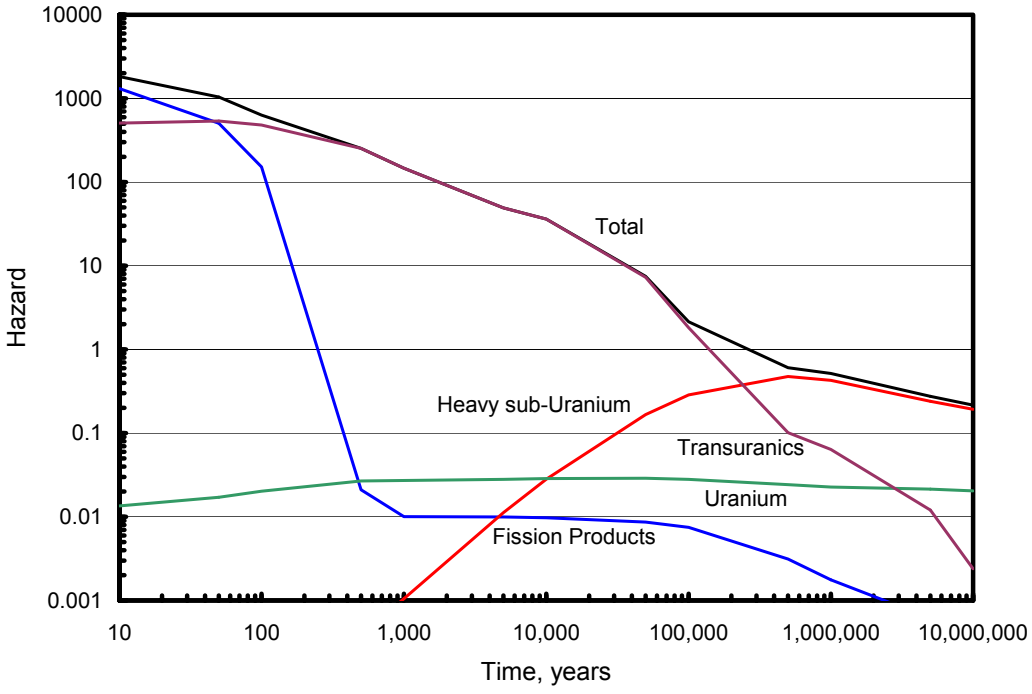

Fig. 2. Hazard for various constituents of LWR fuel with a burnup of 33 GWd/MTHM.

Table II
Time when Hazard
Becomes Less than Unity

Fraction of Actinides Removed	Time after Discharge, Years
0	260,000
0.9	46,000
0.99	1,900
0.999	370
1	270

For a mix of LWR fuels such as expected in the proposed Yucca Mountain Repository, the burnup for PWR fuel is about 25% higher than the burnup considered in constructing Table II. The burnup for the BWR fuel is about the same as considered for Table II, but the enrichment is about 25% lower. To a first approximation, considering both the PWR and BWR fuel, the number of years to reduce the hazard to that of the original ore would be slightly larger than listed in Table II. Nevertheless, the qualitative statements in the preceding paragraph would continue to hold. For a repository containing the mix of LWR fuel waste packages and co-disposal waste packages expected in the proposed repository, one can estimate that removal of 99% or more of the actinides from the LWR packages would leave a residual of about 10% of the total actinide inventory. In this case, about 50,000 years would be required to reduce the hazard to a value less than that for the original ore.

IMPACT ON LONG-TERM DOSE RATE

As a group, actinides tend to be relatively insoluble in water. In the extreme, if they were completely insoluble, removing actinides from spent fuel destined for a geologic repository would have no impact on the dose rate from water taken at a well 20 km from the repository. If solubilities are small, then the release of actinides will be controlled by the flow of water through the repository. Large solubilities will result in release rates controlled by the dissolution rate of the waste form. Note that if a continuous film of water exists in the porosity of the rock and the degraded waste form, then even if there is essentially no flow, radionuclides released from the degraded waste forms may be transported by diffusion. In this case, the solubility will determine whether the release of radionuclides to the environment is controlled by the diffusion rate through the film of water, or by the waste form dissolution rate.

The starting point for the present study is one of the models used in the total system performance assessment (TSPA) for the site recommendation (SR) of the Yucca Mountain Repository. The model, SR00_042nm6.gsm, was used to generate the results shown as Fig. 4-188 in the *Yucca Mountain Science and Engineering Report*.[2] The model is designed to be executed using the GoldSim simulation software.[4] One of the key parameters estimated by the model, and the parameter of interest for this study, is the dose rate resulting from use of water from a well located about 20 km from the repository.

The model was modified to eliminate the explicit modeling of radionuclide transport through the unsaturated zone (UZ) and saturated zones (SZ). The UZ is tuff with porosity partially filled with water and located above the water table while the SZ has porosity saturated with water and is located within the water table. As will be described below, the effect radionuclide transport in UZ and SZ resulting from changes in the actinide content in the waste was estimated by comparing the dose rate at the well with the release rates from the engineered barrier system (EBS).

In addition to eliminating the simulation of transport through the geosphere (the UZ and SZ), time-dependent failure distributions and waste package breach sizes were archived and reused rather than recalculated for each run. The modifications shortened the time required to evaluate a single time history by about a factor of 10. EBS release rates, evaluated with the modified GoldSim input file, agreed precisely with the corresponding rates in the original GoldSim model file.

Release from the EBS

To estimate the impact of actinide removal, the first step was to evaluate the impact on the release of actinides from the EBS. The results of this analysis are illustrated for ^{237}Np in Fig. 3. In the calculations, actinides were removed from all waste sources Solubility limits apply to radionuclide transport in the EBS. As a result, as shown in the figure, the effect of reducing the initial ^{237}Np inventory by even as much as three orders-of-magnitude is relatively small for most of the time period between first waste package failures and about 50,000 years. This is a result of the low solubility of ^{237}Np. By 100,000 years, the reduction in the release rate is about a factor of 260 when the initial ^{237}Np inventory is reduced by a factor of 1000. Although not shown in Fig. 3, by 200,000 years, a reduction in the ^{237}Np inventory by a factor of 1000 results in a factor of 1000 reduction in the ^{237}Np release rate from the EBS, indicating that the release is no longer solubility-limited.

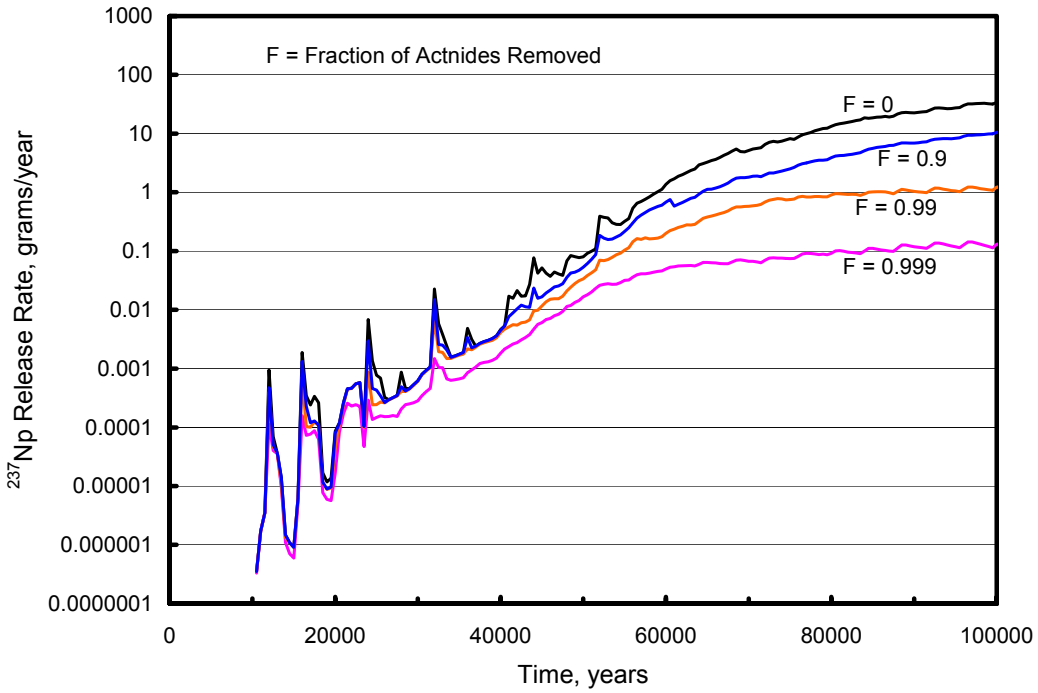

Fig. 3. Release rate of ^{237}Np as a function of the fraction of actinides removed from all waste forms.

Similar results to those for ^{237}Np were obtained for other important radionuclides, including ^{239}Pu and ^{242}Pu. However, when the initial inventory of uranium was reduced, the reduction in the release rates for the uranium isotopes was not the same as described for ^{237}Np. For example, at 1,000,000 years, a reduction in the initial inventory of ^{235}U by a factor of 1000 resulted in a reduction in the release rate from the EBS of only a factor of 190. For ^{238}U, the reduction was about a factor of 150. ^{235}U behaves differently from ^{238}U because of changes in the amount of ^{235}U produced in the EBS by the decay of ^{239}Pu. The differences between the behavior of uranium and other actinides may be due to the fact that there is much more uranium than other actinides. The larger amounts of uranium mean that solubility limits influence its transport in the EBS for longer times than for other actinides.

Estimation of Dose Rate

A simple procedure to estimate the change in the dose rate at the 20-km well was developed based on comparisons between the release rate from the EBS and the rate at which radionuclides cross a boundary located at the well. The controlling parameter appears to be sorption, i.e., the tendency for the element to become bound to the rock matrix. For moderately sorbing elements such as neptunium and uranium, the temporal shapes and magnitudes of the EBS release rates and the release rate across the 20-km boundary are remarkably similar. The primary difference is that the release rate at the 20-km well is delayed between 5,000 and 10,000 years. Similar comparisons for non-sorbing elements such as technetium and iodine show the same similarity in shape and magnitude, but the delay in the crossing rate at the 20-km well and the release rate from the EBS is only one or two thousand years. On a 1,000,000-year time scale, delays by even as much as 10,000 years can be considered to be negligible.

Differences between the EBS release rates and the release rate at 20-km boundary are much larger for strongly sorbing elements such as plutonium and thorium. Delay times between the two release rates range from a few ten thousand years prior to about 100,000 years to a few hundred thousand years at later times. However, for radionuclides irreversibly sorbed onto colloids, the release rates from the EBS are very similar to the release rates across the 20-km boundary, i.e., they behave like non-sorbing elements due to the mobility of the colloids.

Dose rates at the 20-km well, when various fractions of the actinides were removed, were

estimated by using the changes in the release rates from the EBS. For parent nuclei, the dose rate contributions were evaluated by assuming that the fractional change in the dose rate was the same as the fractional change in the EBS release rate. This would appear to be a very good approximation for moderately sorbing elements like neptunium and uranium. ^{233}U, ^{235}U, and ^{238}U were treated as parent nuclei even though their inventories are modified during repository operation by the decay, respectively, of ^{237}Np, ^{239}Pu, and ^{242}Pu.

The dose rate contributions for isotopes of the more strongly sorbing element plutonium were also estimated to change by the same fraction as its release rate from the EBS. Because the differences in the shape of the release rates from the EBS and the 20-km boundary are so large, this approximation is more suspect than for moderately sorbing elements. It may be justified in part by the fact that the fractional change in the release rate of plutonium from the EBS is nearly constant after a few hundred thousand years, the time period when the delay in the release at the 20-km well is largest.

Finally, the dose rate contributions for decay product nuclei such as ^{227}Ac, ^{231}Pa, and ^{229}Th were estimated to change by the same fraction as the release rates the nuclei from which they are produced. Each of these isotopes result directly or indirectly from the decay of an isotope of uranium. ^{227}Ac, with its very short half-life (22 years) must be in secular equilibrium with ^{231}Pa. ^{231}Pa has a fairly long half-life (33,000 years) and is probably not in secular equilibrium with ^{235}U. Nevertheless, its presence in the vicinity of the well must be strongly correlated with the presence of ^{235}U. ^{229}Th (half-life 7300 year) is probably tending toward secular equilibrium with ^{233}U.

Figure 4 shows the estimates, obtained as outlined in the foregoing paragraphs, of the changes in the dose rate when various fractions of the actinides are removed from the waste forms. This figure shows that removal of 99% of the actinides might reduce the peak dose rate by as much as a factor of 60. Removal of 99.9% of the actinides reduces the peak dose rate to only slightly more than the contribution to the peak dose rate by ^{99}Tc.

It should be noted that the foregoing estimates of the dose rate are based on the assumption that the waste form resulting from the process that removes the actinides is as effective in retaining radionuclides as the original spent fuel. As modeled in the TSPA, spent fuel is more effective in retaining radionuclides than,

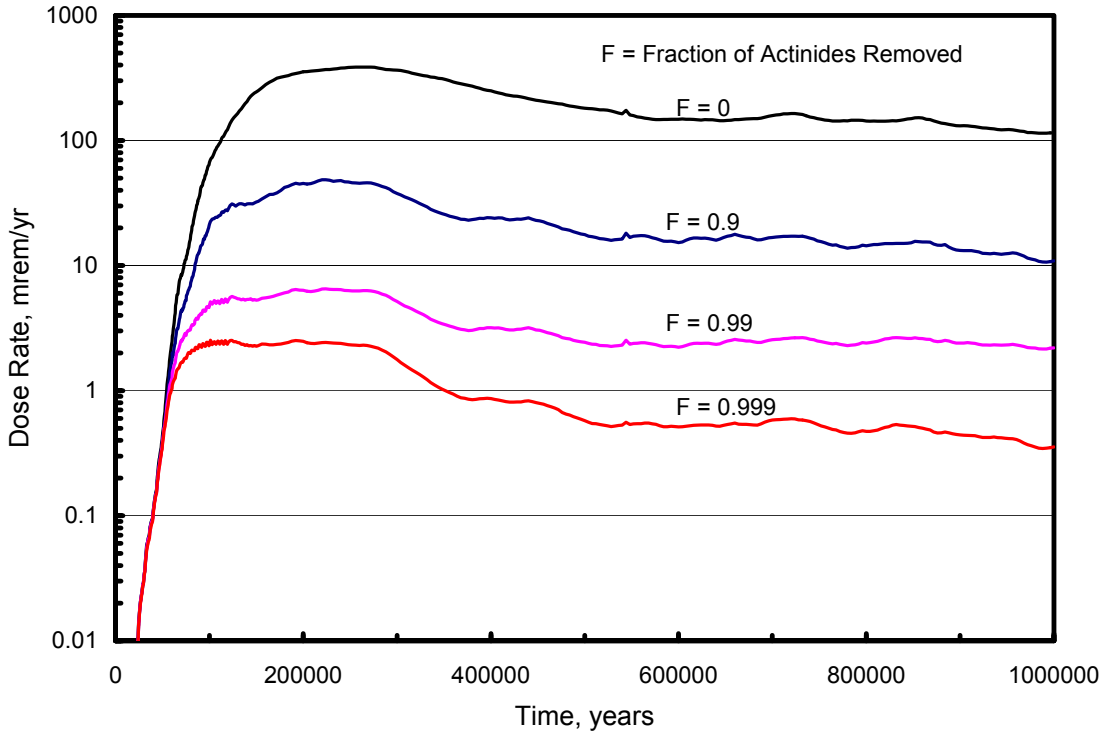

Fig. 4. Estimated dose rate at the 20-km well as a function of the fraction of actinides removed from all waste forms.

for instance, defense high-level waste glass. It remains to be demonstrated that a waste form as effective as this model assumption can be developed. Also, as stated earlier, actinides are removed from all waste forms. If actinides are removed only from the PWR and BWR commercial spent nuclear fuel, the reduction in the peak dose will not be as large as indicated in Fig. 4. This is because even though the actinide inventory is much lower in the non-commercial waste packages, in the current repository model the waste forms they contain are not as effective in retaining radionuclides once the waste packages are breached.

In connection with some of the points in the preceding paragraph, a few calculations have been performed using a simplified TSPA model developed by Golder Associates.[5] In one of the calculations, 99% of actinides were removed from all radionuclide inventories in the GoldSim model. The magnitude of the change in the dose rate at the 20-km well was essentially the same as shown in Fig. 4. Removal of 99% of the actinides from both the PWR and BWR spent nuclear fuel and DOE spent nuclear fuel (but not from defense high-level waste glass) resulted in a reduction in the peak dose rate by about a factor of 20. This is in contrast to the factor of 60 indicated in Fig. 4. However, when 99% of the actinides were removed only from the PWR and BWR spent nuclear fuel, the magnitude of the peak dose rate was reduced only by a factor of about 6.

CONCLUSIONS

Removal of actinides from the waste destined for the proposed Yucca Mountain Repository does not eliminate the need for such a facility. However, the foregoing analyses show that for each additional increment in the fraction of actinides removed, there is a corresponding benefit. The major benefits include increased repository capacity or reduced size because of the substantial reductions in the amount of waste heat to be dissipated, especially at longer times. In addition, there can be a substantial reduction in the long-term radiotoxicity of the waste and a reduction in the long-term dose rate resulting from the eventual failure of waste packages. While much of the analysis has used the proposed Yucca Mountain Repository to illustrate the various impacts of actinide removal, the qualitative results should be broadly applicable to any repository where spent fuel is to be interred.

ACKNOWLEDGEMENTS

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