REPOSITORY BENEFITS OF PARTITIONING AND TRANSMUTATION

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Abstract

Geologic repositories such as the one proposed for Yucca Mountain are designed to both safely store hazardous nuclear materials for very long periods of time and to minimise the release rate of these materials to the environment. To succeed in this mission, these repositories need to satisfy a number of design and operational constraints which usually take the form of temperature limits, restrictions on inventory, etc. In general, the ability of a repository to satisfy the constraints depends on the nature and characteristics of the emplaced materials. This paper reports the results of several studies into the benefits to a geologic repository that can be obtained by processing and partitioning spent nuclear fuel, and recycling certain chemical elements in nuclear reactors to transmute them into less hazardous materials. The modest benefits obtained using limited recycling in LWRs is discussed, along with the far greater benefits possible when continuous, or repeated, recycling approaches are employed.

Overview

Geologic repositories such as the one proposed for Yucca Mountain are designed to both safely store hazardous nuclear materials for very long periods of time and to minimize the release rate of these materials to the environment. To succeed in this mission, these repositories need to satisfy a number of design and operational constraints which usually take the form of temperature limits, restrictions on inventory, etc. In general, the ability of a repository to satisfy the constraints depends on the nature and characteristics of the emplaced materials, most of which is commercial spent nuclear fuel (CSNF) in the United States. The emplaced materials can be altered by processing the spent nuclear fuel prior to disposal, removing certain chemical elements, and re-irradiating these elements to transmute them into materials more suitable for disposal in the repository.

The first part of the problem is to identify the chemical elements (isotopes) that are responsible for the applicable repository design limits being reached. In particular, in the case of a repository at Yucca Mountain, the temperature limits imposed on the engineered systems and on the mountain result in drift loading limits for the direct disposal of spent nuclear fuel. The measure of repository benefit has been defined as the allowable increase in repository drift loading consistent with satisfying all repository thermal design limits, since loading of a geologic repository at Yucca Mountain is currently limited by temperature constraints. Such an increase in drift loading can be used to either reduce the size of a repository of given capacity, or to increase the capacity of a repository of a given size. Any changes in estimated peak dose caused by the resulting alteration in the radionuclide inventory of the repository have not been evaluated, but are the subject of a separate ongoing study.

The following section of this paper reports the results of thermal analyses of a geologic repository at Yucca Mountain which show that removal of plutonium and americium is essential as the first step towards better utilization of the space available in repository drifts. With 99.9% removal of plutonium and americium, the repository benefit, as measured by the increase in drift loading, can be a factor of 5-6 over the direct disposal case, using anticipated repository operating conditions of disposing of spent fuel 25 years after reactor discharge, and ventilating the repository for 75 years after waste placement. It is then demonstrated that by additional processing of the spent fuel to remove cesium and strontium, further increases to the repository drift loading are possible. The results show that it is possible to increase the drift loading by up to a factor of 100 with 99.9% removal of TRU and cesium and strontium, for the same repository operating conditions, and assuming that suitable waste forms are available to take advantage of such potential.

The last part of the discussion examines possible recycling scenarios for the recovered plutonium, americium, and other elements. The need for ongoing processing of the spent fuel is presented, along with the requirement that the recovered plutonium and americium must be recycled repeatedly if large increases in repository space utilization are to be possible. Such repeated recycling can be achieved with either thermal or fast neutron reactors, although some elements are more easily transmuted in a fast neutron spectrum. Overall, the results indicate that substantial drift loading benefits can only be achieved by keeping the contents of TRU in the waste (or any other emplaced materials) as low as possible, and by separately storing the cesium and strontium.

Processing and partitioning requirements for commercial spent nuclear fuel

The direct disposal of CSNF is the current reference case for a repository at Yucca Mountain.[1] The repository drifts are 81 m apart, about 1 km in length, and 5.5 m in diameter. In this study, the repository is assumed to be ventilated for 75 years after repository closure to further reduce the importance of shorter-lived isotopes. This is longer than the minimum specification of 50 years, but within the operational envelope being considered by the Yucca Mountain Project. The reference operating mode, the high-temperature operating mode (HTOM) of the "cold" repository, is calculated to have an extended time period where the temperature of the rock surrounding the waste storage drifts will be above the boiling point of water. One temperature limit specifies that the rock temperature midway between adjacent drifts must always remain below 96 °C to ensure 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 once the temperatures have dropped. Another temperature limit is that the rock temperature must remain below 200 °C at all times to prevent alteration of the crystalline structure of the rock. Both of these temperature limits are used to provide greater certainty about the conditions in the repository, increasing 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 rate that leads to release of radioactive materials.

In a previous study, the repository benefits of separating certain chemical elements from spent fuel and recycling them to alter the characteristics of the waste stream destined for a repository at Yucca Mountain were quantified. [2] The conclusions from that study are as follows:

1. The dominant contributors to the thermal load from the emplaced spent fuel or waste in a repository at Yucca Mountain that lead to reaching one or more of the temperature limits are plutonium and americium. Removal of these chemical elements, and recycling and transmuting them to reduce the inventory of these elements in the materials placed in the repository, is essential to increasing the repository drift loading. The benefit ranges from a factor of 5-6 in increasing the drift loading (or decreasing the repository size for a given capacity) with waste placed in the repository at 25 years after reactor discharge.

2. After the plutonium and americium have been removed, the next chemical elements that need to be considered are cesium and strontium, mainly for the heat produced by their short-lived decay products, barium and yttrium. Removing cesium and strontium, and sequestering them in a separate area of the repository or in another facility, would allow a substantial increase in the repository drift loading, up to a factor of 40-50 greater than the direct disposal case, for 99.9% removal of plutonium, americium, cesium, and strontium, and when the waste is placed in the repository 25 years after reactor discharge.

3. The next most important chemical element is curium. However, in considering a realistic recycling scenario for the plutonium and americium, assumed to be irradiation in a fast reactor, it was observed that the drift loading can be limited by the losses of these two elements from processing the spent fast reactor fuel. The 1% loss assumed in the processing of the fast reactor fuel reduced the potential increase in drift loading from a factor of 43 to 21, and dominated the decay heat generation. This emphasises the need to reduce recycling losses of plutonium and americium below the 1% assumed in this study before separation of curium will be effective.

4. The issue of peak dose rate can be addressed by removing and recycling the plutonium, americium, and neptunium, as these chemical elements are the dominant contributors to the peak dose rate. It should be noted that the estimated peak dose rate of 100 to 400 mrem/year is occurring at times well beyond the current 10,000 year regulatory period, about 250,000 years.[1] Current estimates of repository performance indicate that the peak dose rate within the regulatory period is several orders of magnitude below the 15 mrem/year limit. Whether separations should be done to alter the estimated performance past the regulatory period appears to be an open question, although increased repository capacity would imply a need to evaluate the impact on the estimated peak dose rate to ensure that regulatory requirements are still satisfied.

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 5-6. 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 effectively transmute these elements and to have very low losses for processing the recycled fuel, regardless of the manner in which the recycled material is treated (thermal reactor, fast reactor, etc.) To take advantage of such a potential increase in drift loading requires the availability of waste forms that could be densely loaded with the remaining waste materials.

Examination of an alternative low temperature operating mode (LTOM) for a repository at Yucca Mountain shows that the same processing and recycling strategies would be effective for that case as well, yielding similar benefits. In summary, the study quantified benefits to a geologic repository that arise from certain spent fuel processing strategies, as would be examined as part of the AFCI program, with these benefits appearing to be relatively independent of the specific repository design decisions being considered for a repository at Yucca Mountain.

Transmutation of plutonium, americium and neptunium in LWRs

Given this background, specific recycling schemes using existing light-water reactors were examined, and the repository benefit quantified, using the following general guidelines:

- Spent PWR fuel irradiated to 51 GWd/MTIHM was used as the example of commercial spent nuclear fuel, as it represents the majority of material destined for a repository at Yucca Mountain, and is also packaged to the highest linear heat load in a repository drift.

- Plutonium and americium are separated from the spent PWR fuel with an efficiency of 99.9% to address the repository heat load issue. Neptunium is also separated from the spent PWR fuel for non-proliferation and radionuclide inventory concerns. Plutonium, americium, and neptunium are recycled for further irradiation in an LWR.

- The spent fuel is assumed to be processed at 5 years after discharge to minimize the buildup of americium during storage and to maximise the potential benefit of recycling in a thermal spectrum to reduce the long-term heat load. The effect of delaying processing and recycling of spent fuel, processing at 20 years after discharge, has also been considered for a single recycle in each strategy to determine the sensitivity of the calculated repository benefit to the age of the spent fuel.

- The number of recycles of plutonium, americium, and neptunium is a parameter in the study. After the last recycling, all materials are sent to the repository, including both processing waste and the spent fuel assemblies from the last irradiation cycle.

- Curium is not separated from the spent PWR fuel, but is sent directly to the repository in the process waste stream.

- Cesium and strontium are separated from the processing waste, and are stored separately, either in dry storage external to the repository or in a separate area of the repository.

- The neutronic analyses of each assembly type, whether enriched uranium fuel or fuel containing the recycled elements, was performed using lattice calculations. This is roughly equivalent to treating the assemblies as if they were in reactor cores of identical assemblies, i.e., homogeneous reactor cores. In some cases, this leads to difficulties with reactor safety coefficients and other issues that may result in these cases being impractical, requiring the use of heterogeneous reactor cores. However, based on past results, it is expected that the results of the lattice calculations for spent fuel compositions would be very close to the results obtained for heterogeneous reactor cores.

- Assemblies fabricated using recycled material are considered as substitutes for additional standard PWR assemblies, and produce the same integrated energy per assembly. Groups of assemblies are formed consisting of the assemblies containing recycled materials and the corresponding amount of processing waste from previous generations of assemblies, representing a certain amount of total integrated energy. Each group is then compared with a PWR group that produced the same total integrated energy. As a result, all determinations of repository benefit are performed on an *equal integrated energy basis*.

- The repository operation and design is consistent with the high-temperature operating mode (HTOM) of the cold repository. The thermal analyses calculating the response of a repository at Yucca Mountain are performed using the detailed 3-D model representing the central area of the repository where the highest temperatures are expected. The relevant temperature limits for the HTOM have been discussed above.

For the purposes of this study, three LWR recycling strategies have been examined, identified as MOX, CORAIL-PNA, and IMF:

- MOX – Mixed-oxide fuel. In this approach, the separated plutonium, americium, and neptunium are used to fabricate new fuel assemblies, in a fuel matrix of recovered uranium (enrichment slightly above natural uranium), all elements being present as oxides. To obtain sufficient fissile material for the first generation of MOX, provided mostly by plutonium, it is necessary to use materials recovered from a number of spent PWR fuel assemblies to fabricate one MOX assembly. Subsequent recycling with MOX uses the recovered plutonium, americium, and neptunium from the current MOX generation to fabricate the next generation of MOX assemblies. Again, to provide sufficient fissile material, a number of spent MOX assemblies from one generation need to be processed to fabricate one MOX assembly in the next generation. For each parametric case on the number of recycles, the last generation of MOX assemblies is directly disposed to the repository, along with the processing waste from all previous generations.

- CORAIL-PNA – This concept uses heterogeneous assemblies, where some of the fuel pins (about 1/3) are fabricated from the separated plutonium, americium, and neptunium, in a uranium matrix, and the remaining fuel pins (about 2/3) are fabricated from new enriched uranium, with all elements present as oxides. In the CORAIL-PNA case, the recovered plutonium, americium, and neptunium from one spent PWR assembly is used to make the 1/3 of the fuel pins for one assembly in the first CORAIL-PNA generation that contain these materials. After irradiation, the entire spent CORAIL-PNA assembly is processed to recover the plutonium, americium, and neptunium for the 1/3 of the fuel pins in a single assembly of the next CORAIL-PNA generation. The remaining 2/3 of the fuel pins are again fabricated from new enriched uranium, with the enrichment increasing with each CORAIL-PNA generation. For each parametric case on the number of recycles, the last generation of CORAIL-PNA assemblies is directly disposed to the repository, along with the processing waste from all previous generations.

- IMF – Inert Matrix Fuel. This approach is similar to MOX, but the fuel matrix is an inert material, zirconia, instead of uranium oxide. The recovered plutonium, americium, and neptunium from several spent PWR assemblies are used to make a single assembly in the first generation of IMF so that sufficient fissile material is provided. Subsequent generations of IMF also use several IMF assemblies of the previous generation to obtain sufficient fissile material. As with the other cases, the number of recycles is a parameter in the study, and after the irradiation of the last generation in each case, the spent IMF assemblies are directly disposed to the repository along with the processing waste from all previous generations.

Repository drift loading increases for LWR transmutation strategies

For each of these cases, detailed fabrication, separation, and irradiation histories had been calculated in a parallel effort by J.A. Stillman [3] and T.K. Kim [4] for a range of the number of recycles, with the assumption that all spent fuel was processed at 5 years after discharge to minimize buildup of americium from plutonium decay. As described in the previous section, the resulting isotopic compositions of the spent fuel assemblies and the processing waste were collected into the appropriate groups to preserve total integrated energy produced and compared with the reference case of direct disposal of spent PWR fuel. It should be noted that there are many issues associated with some of these strategies, including fuels development, designs of practical reactor cores, and fuel processing methods. These remain as open issues for further study and evaluation.

The potential increases in repository drift loading are displayed in Figure 1, with general results for each case as follows (where it is also noted that all cases shown in the figure may not be realizable in practice):

- MOX – The use of MOX fuel allows a steady increase in repository drift loading with each recycle of plutonium, americium, and neptunium, reaching a factor of 1.5 after 5 recycles of MOX fuel. The factor for the increase in drift loading is a combination of a very large factor for the process waste (~40) and a very small factor for the direct disposal of the spent MOX assemblies from the last recycle after the specified number of recycles (typically <0.10). At this time, it is not clear that the use of MOX to 5 recycles could be realized due to several difficulties, especially with reactor safety coefficients, but the use of heterogeneous reactor cores with assemblies of all generations may alleviate this problem.

- CORAIL-PNA – The use of CORAIL-PNA also allows a steady increase in drift loading with each recycle at a faster rate than for MOX, reaching a factor of 2.0 after 7 recycles of CORAIL-PNA fuel due to the favourable impact of using enriched uranium to provide fissile content rather than relying entirely on the recovered plutonium, americium, and neptunium. As with MOX, the factor for increasing the drift loading is a combination of a very large factor for the process waste (~40) and a very small factor for the direct disposal of the spent CORAIL-PNA assemblies from the last recycle after the specified number of recycles (<0.10). Unlike the MOX cases, though, the use of enriched uranium to supplement the fissile content in the CORAIL-PNA concept appears to allow recycling to reach an equilibrium state, where the charge and discharge amounts of plutonium, americium, and neptunium are equal and only process losses would go to the repository.

- IMF – The use of inert matrix fuel provides a factor of 1.8 for the increase in drift loading after the first recycling, and a factor of 2.1 after the second recycling. Further recycling of IMF is hindered by the rapid depletion of fissile material with each subsequent irradiation (especially for Pu-239), and makes it impossible to perform a third recycling to the same integrated energy for the assembly. Additional recycling using IMF fuel may be possible using heterogeneous assembly designs and blending with other fissile material, but this variation has not yet been evaluated. As with the other cases, the factor for increasing the drift loading is a combination of a very large factor for the process waste (~40) and a very small factor for the direct disposal of the spent IMF assemblies from the last recycle after the specified number of recycles (<0.10).

- In all cases - The recycling of neptunium appeared to be detrimental to increasing the drift loading in the repository, mainly through neutron capture in Np-237 to create additional heat-generating Pu-238. While not the dominant isotope, Pu-238 is second in importance to Am-241 in providing decay heat and limiting the drift loading of the repository. Some increase in benefit is expected if only plutonium and americium are recycled, but the amount has not been quantified at this time.

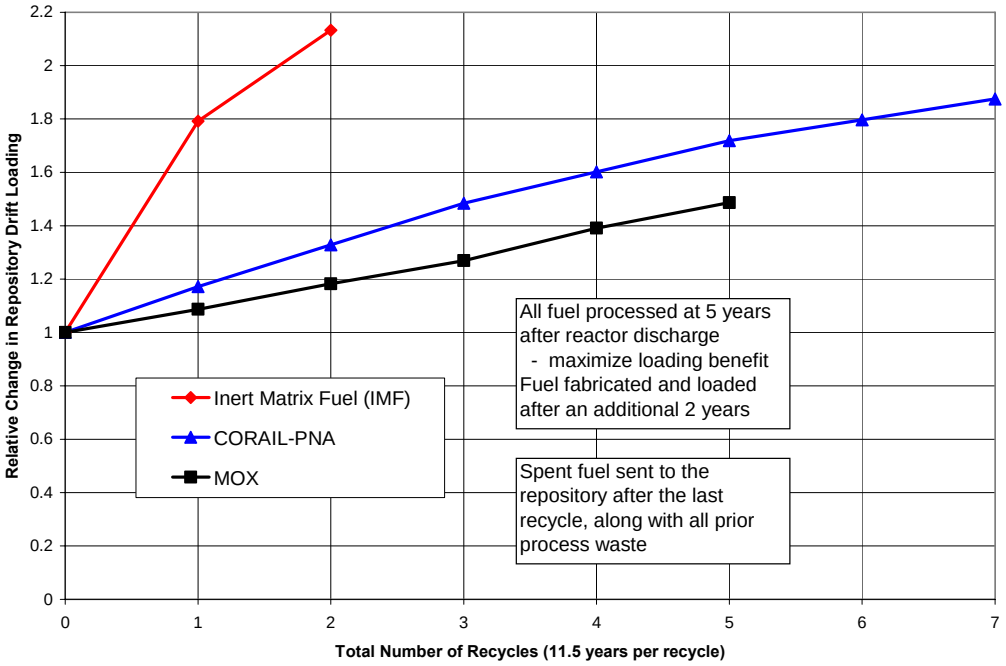

Figure 1. **Relative change in repository drift loading as a function of the number of recycles for MOX, CORAIL-PNA, and IMF recycling options, fuel processed at 5 years after reactor discharge, with spent fuel sent to the repository after the last recycle**

Sensitivity of repository drift loading increase to the age of the spent fuel

Since the repository drift loading for direct disposal of spent PWR fuel is controlled by the decay heat from americium (Am-241) arising from the decay of Pu-241 in the stored spent fuel after irradiation, the analyses reported in the previous section were performed for processing all spent fuel at 5 years after discharge to minimize the decay of Pu-241 and thus maximize the benefit to the repository. The benefit to the repository is also closely related to the total amount of Pu-241 (and Am-241) in the discharged fuel when the spent fuel is directly disposed in the repository. Higher enrichments are needed when older fuel is processed and recycled due to the decay of fissionable isotopes and the increase in isotopes that are more likely to capture neutrons in the thermal spectrum. The higher enrichment increases both the Pu-239 and Pu-240 content in the recycled fuel, and the net production of Pu-241 is increased, potentially reducing the benefit to the repository from processing and recycling spent fuel. To quantify the sensitivity of the repository drift loading increase to the age of the spent fuel, analyses were also conducted for one recycle in each case using spent fuel that had been stored for 20 years.

The effect of processing older fuel and recycling the recovered plutonium, americium, and neptunium in an LWR differs depending on the recycling strategy. The least impact is observed for CORAIL-PNA, where the use of enriched uranium reduces sensitivity to the changing isotopic composition of the recycled materials by allowing lower plutonium enrichment. The reduction in benefit as measured by the increase in repository drift loading is slightly less than 30%. The greatest impact is observed with IMF, since IMF depends entirely on the fissile content of the recovered materials, which is mostly plutonium, and more fissile is needed as the composition and isotopic distribution changes with age. In this case, the reduction in drift loading benefit is over 75%. The effect on MOX is between these two, with a reduction in the allowable drift loading increase of about 50%, due to the use of a uranium matrix that provides additional fissile material during irradiation. However, the overall effect is that for a single recycle step using older fuel, the use of IMF is comparable to using CORAIL-PNA, with the benefit of using MOX becoming minimal.

Remaining issues and considerations

As additional comments, it is useful to note that processing and recycling spent PWR fuel could also be used to lower temperatures in a geologic repository, maintaining the reference drift loading as currently planned for a repository at Yucca Mountain. This would allow more stringent temperature limits to be met, if needed, and could be used to provide greater assurance about the performance of the repository. Also, the reduction in the radionuclide inventory and the resulting alteration in estimates of the peak dose rate associated with releases from the repository have not been specifically addressed in this study, since this is not an issue for a repository at Yucca Mountain at this time as dose rate from releases does not constrain repository loading. However, some of the elements that are responsible for producing the decay heat in a repository (Pu, Am, Cs, and Sr), plus Np, are the main contributors to the dose rate. In a related ongoing study, quantifying the effects of the processing and recycling strategies using detailed repository performance assessment is showing that substantial dose rate reductions may be possible.

Systematic PWR fuel processing and recycling as evaluated in the current study would appear to offer other potential benefits for nuclear power, such as being able to densely dispose of process waste in a repository while greatly reducing the number of irradiated fuel assemblies that would need to be stored at reactor sites. Such possibilities emphasize the need to consider all relevant advantages and disadvantages when evaluating the usefulness of recycling in thermal reactors.

Conclusions

Results of this study have quantified the benefit to a repository as measured by increased drift loading of the repository; about a factor of 2 for the limited number of recycles considered. The results have also emphasized that approaches using a limited number of recycles will provide only a small fraction of the potential benefit (in excess of a factor of 40) that could be achieved with continuous recycling where the plutonium, americium, and neptunium remain in the fuel cycle, except for process losses. As discussed above, the general reasons for this have been determined as follows:

- In each case, the process waste is capable of being very densely loaded in the repository drift while still satisfying all thermal limits, about a factor of 40-50 greater than for spent PWR fuel. This is consistent with the previous results obtained for continuous recycling, since only process waste is sent to the repository in that case as well. (This can be increased to about a factor of 100 if curium were also separated from the process waste.)

- Direct disposal of the last assemblies in each recycling strategy requires most of the repository space, with allowable drift loading densities less than 5-10% that for spent PWR fuel. This is not unexpected, since all of the higher actinide elements from many assemblies have been concentrated in the assemblies containing recycled material.

- It is the combination of the increased drift loading density for the process waste and the reduced drift loading density of the last assemblies in each case that limits the overall benefit to the loading of the repository to a factor of about 2.

- Continuous recycling is essential for obtaining large increases in drift loading in the repository, and may be possible in a thermal spectrum using the CORAIL concept, or advanced MOX, IMF, or target strategies. The key is to prevent a large fraction (>99%) of the heat-producing transuranic inventory from ever being placed in the repository.

Given these conclusions, it would be useful to further quantify the impact of recycling scenarios using thermal neutron systems, where production of the higher actinide isotopes is enhanced, as compared with alternate scenarios involving irradiation in a fast neutron spectrum, where production of higher isotopes is limited. However, the results of this study should not be interpreted to mean that processing spent PWR fuel and recycling in LWRs is of little or no benefit:

- By processing the spent fuel and sending only process waste to the repository, substantial increases in loading (or corresponding decreases in repository size) are possible as soon as the activities are begun.

- As long as the recycled plutonium, americium, and neptunium are kept in the fuel cycle, the large allowable increase in drift loading, ~ 40 or more, is realized and maintained. The need to keep the plutonium, americium, and neptunium in the fuel cycle should not necessarily be viewed as a disadvantage, since in any version of a uranium-fuelled nuclear future where nuclear power generation is at least maintained, the eventual deployment of fast reactors appears to be inevitable and would provide the eventual destination for the remaining plutonium, americium, and neptunium. It should also be noted that in the absence of a nuclear future, processing of spent nuclear fuel from existing reactors is likely to be unnecessary, as sufficient repository capacity is likely to be available even within a repository at Yucca Mountain to store all of the spent fuel.

- The use of LWRs for recycling, where they are essentially plutonium burners, reduces the number of fast reactors needed to provide the continuing recycling of higher actinides for a given number of LWRs, as shown in previous work.[5]

In summary, it has been shown that processing spent PWR fuel and using limited recycling in LWRs is of modest benefit to a geologic repository where loading is determined by thermal constraints. In addition, consideration of older fuel virtually eliminates any advantage from novel fuel types such as inert matrix fuel, and that use of mixed-oxide fuel provides essentially the same benefit. However, it has also been shown that recycling in LWRs can be very beneficial as part of an overall strategy where nuclear power generation continues into the future.

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