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Plutonium destruction in a non-fertile, ZrO_2-based fuel - a reactor option for disposition of surplus plutonium

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Abstract

The United States and Russia are assessing options for disposition of surplus weapon-grade plutonium. This paper reviews the options under consideration by the US Department of Energy and suggests an additional option that fits within the framework of the environmental analysis provided in the draft PEIS (Programmatic Environmental Impact Statement). In addition to the burning of Pu in mixed U-Pu oxide fuel, we recommend consideration of a non-fertile fuel based on zirconia with inclusion of rare earth elements for phase stabilization and control of reactivity. The zirconia based fuel could also be used to burn plutonium generated in commercial reactor fuels, which represent a larger inventory of plutonium than the weapon-grade material. The increasing inventories of civilian plutonium potentially represent a larger threat with respect to diversion of weapons usable material than the stocks of weapon-grade material considered for disposition by the US and Russia. We discuss the use of zirconia-based fuel and pyrochemical processing of spent commercial reactor fuels as a means of decreasing world-wide plutonium inventories. The experience gained in burning weapon-grade plutonium in the new non-fertile fuel would shorten the time required to gain acceptance of the fuel for commercial reactor use.

Introduction

The end of the Cold War and the subsequent negotiation of treaties between the United States and Russia has resulted in the recall and dismantlement of weapons containing highly enriched uranium (HEU) and plutonium. The HEU, after dilution with depleted or natural uranium, could be used to manufacture conventional light water reactor fuel; the disposition of the plutonium presents a more difficult question.

Options for the disposition of surplus weapon-grade plutonium are being assessed in the United States and in Russia as part of a joint effort to reduce the risks of holding such material in storage. The US National Academy of Sciences recommended that surplus plutonium should be handled using spent commercial reactor fuel as the standard for safeguards and security requirements (NAS, 1994). This "spent fuel standard" is subject to considerable interpretation in the details of implementation, but generally means that the plutonium from surplus weapon stock should be no easier to recover and use for weapons than the plutonium in spent fuel. In addition, the NAS report emphasized that the measures taken to dispose of weapon-grade plutonium should not be more stringent than those used for spent fuel. This was done in recognition that spent fuel from commercial reactors constitutes a much larger reservoir of plutonium than the surplus weapon material. While there is a considerable difference in isotopic composition of the two types of plutonium, both present risks with respect to diversion of material to inappropriate uses.

Options under review for the disposition of plutonium

The National Academy report (NAS, 1994) identified two main options for disposition of surplus plutonium. They were to use plutonium in existing reactors as "once-through" fuel or to mix the plutonium with high level radioactive wastes and vitrify the mixture. A third option, to dispose of plutonium in deep boreholes, was also mentioned, but the report noted that it had been less thoroughly studied than the other two.

In response to the National Academy recommendations, the US Department of Energy began a formal study of disposition options for plutonium. The evaluation process is being conducted under the US DOE guidelines for compliance with the US National Environmental Policy Act (NEPA). The NEPA process involves an evaluation of various options, including a no-action alternative, to provide the means for deciding on the ultimate course of action. A draft Programmatic Environmental Impact Statement (PEIS) must be prepared that examines the environmental impacts of various options. The PEIS must then be issued for public comment; this was done in March 1996 (DOE 1996a). After the comment period and revision of the PEIS to reflect response to the comments, a final PEIS will be issued. That document will provide the basis for a programmatic decision by the US government concerning the disposition of surplus US weapon-grade plutonium.

The US DOE Office of Fissile Materials Disposition developed screening criteria for disposition options in order to select options for further evaluation. The screening criteria were as follows:

Resistance to theft and diversion by unauthorized parties;

Resistance to retrieval, extraction, and reuse by host nation;

Technical viability;

Environmental, safety, and health compliance;

Cost-effectiveness;

Timeliness;

Fosters progress and cooperation with Russia and other countries (establish standards for disposition of surplus weapons-usable fissile materials, support negotiations for bilateral and multilateral reductions in these materials, and allow for international inspections at all stages);

Public and institutional acceptance;

Additional benefits (including the ability to leverage the government investments for disposition of surplus materials to contribute to other national or international initiatives).

At the end of the screening process, nine cases for disposition were selected for further analysis. These cases were two options for disposal of plutonium in deep boreholes, with or without immobilization of the plutonium in a waste form; three options for mixing plutonium with fission product radionuclides and immobilization in an engineered waste form barrier in preparation for disposal in a high level waste repository; and four options that involved making surplus plutonium into mixed uranium-plutonium oxide fuel to be used in either light water or CANDU reactors.

The DOE PEIS (DOE, 1996a) contains analyses of the environmental impacts of implementing these options. Other supporting documents contain discussions concerning variations on the basic options, such as different geometric arrangements of wasteforms and different locations for implementation. In the following section we will describe these options briefly in the three generic categories and then present an alternative strategy that we believe offers significant advantages over those options presently included in the PEIS.

Assessment of options

Borehole Disposal Options

There are two deep borehole disposal options discussed in the PEIS. Both of these involve placement of the plutonium in canisters followed by lowering a string of emplacement canisters into a borehole that reaches 4 kilometers into the bedrock with the waste canisters confined to the bottom 2 km section of the hole. The upper 2 km of the borehole would contain backfilling materials and seals to divert water so that there would not be preferred vertical flow of water down the borehole. The two options differ in that in one of the options plutonium would be disposed of without immobilization into a waste form, while in the second plutonium would be chemically combined into ceramic in pellet form to immobilize the plutonium. The pellets would then be mixed with grout inside emplacement canisters.

In both options, the plutonium-containing canisters would be below the water table. For plutonium that was not incorporated into a waste form, the only barrier to dispersion into the environment would be the integrity of the emplacement canister. Since ^{239}Pu decays to ^{235}U, which is also fissile, the potential for criticality to occur through migration of fissile materials downhole or out of the hole must be addressed for a time comparable to the age of the earth. It does not seem reasonable to expect that a canister can be designed to withstand degradation for times of that order. Thus, assurance against criticality seems to depend on the use of a waste form with good resistance to dissolution. The ceramic pellets containing plutonium could provide this resistance. Furthermore, neutron absorbing materials could be added to the ceramic to further decrease the liklihood of criticality in the long term.

The borehole disposal concept has the advantage over other disposal concepts for plutonium that it does not require addition of other radioactive materials to the disposal medium in order to satisfy the requirements for rendering the plutonium "unattractive" to those who might want to retrieve it. This helps in the design of a durable waste form and also helps minimize the potential radiation dose to workers during preparation of the materials for disposal.

A disadvantage for the borehole disposal concept would be the difficulty in assessing the hydrogeologic characteristics of the intended host rock in a manner sufficient to do adequate performance assessments of the disposal concept.

Immobilization Options

The immobilization alternative study considered a wide variety of possible methods for immobilizing surplus plutonium in waste forms that meet the general screening criteria listed above. In addition to the general criteria, the immobilization methods must result in stable waste forms that are capable of meeting repository waste acceptance criteria (that have not yet been defined for waste forms that contain weapon-grade plutonium) and that meet the so-called "spent fuel standard". The intent of the "spent fuel standard" is to assure that the plutonium-bearing waste form is no more attractive for potential theft than spent nuclear fuel such as that from commercial light water reactors. The components of this "standard" include a fairly low plutonium concentration and a high radiation field that discourages unsophisticated handling operations with the objective of theft.

After down-selection based on the screening criteria, three main plutonium immobilization options remained, two of which included sub-options. These options are discussed in the Immobilization Alternative Technical Data Documents ((DOE, 1996b). There are four variations on incorporation in borosilicate glass along with high-level tank wastes, two variations on incorporation in a ceramic waste form along with ^{137}Cs from capsules now stored at Hanford (Jardine, 1995), and one option to use electrometallurgical treatment of plutonium concurrently with DOE-owned spent nuclear fuel to make a glass-bonded zeolite waste form.

The borosilicate option would be done using one of several possible methods, including (1) in conjunction with operation of the Defense Waste Processing Facility (DWPF) glass melter at Savannah River, (2) in conjunction with operation of the DWPF glass melter, but the glass would be poured into canisters that contained individual plutonium-containing cans mounted on a rack inside the canisters, (3) in a smaller adjunct glass melter located in an adjoining facility at Savannah River, or (4) in a green field (completely new) glass melting facility.

The ceramic option would use a waste form that is similar to the "Synroc" waste form, in which the plutonium is stabilized in oxide minerals, such as zirconolite, in a ceramic matrix. The two cases being considered for this option are (1) the use of pellets that would be put into small canisters and then treated in a manner similar to the glass "can-in-canister" approach and (2) a ceramic monolith where the radioactivity required to meet the spent fuel standard would be provided by incorporation of ^{137}Cs from the Hanford capsules. The second ceramic method could either be done as a green field facility or by modification of the Argonne National Laboratory-West (ANL-W) hot cells to include the plutonium residue treatment and hot-pressing equipment to make the ceramic waste form.

The electrometallurgical treatment option includes only one case, which is to incorporate the surplus plutonium into a spent fuel treatment process that would be done at the ANL-W hot cells. A variety of spent nuclear fuels would be treated, and the radioactivity from them would provide most of the activity required to meet the "spent fuel standard". The surplus plutonium would be incorporated, along with the transuranic (TRU) elements from the spent fuel, into the glass-bonded zeolite waste form from the electrometallurgical treatment of spent fuel.

The disadvantages of the Immobilization Alternative options are the probable difficulty of licensing high-plutonium waste forms for repository disposal, the large volume of waste required to handle up to 50 metric tonnes of weapon-grade plutonium thought to be surplus to requirements, and the short life of the protection offered by the "spent fuel standard", which decays along with the 30-year half life of ^{137}Cs. The licensing difficulty would arise from the greater difficulty of assuring the absence of post-emplacement criticality in a repository designed for wastes with less plutonium content. Neutron absorbers would be incorporated in the waste forms to prevent near-term criticality; however, it will be very difficult to assure that the neutron absorbers would migrate exactly the same way as the fissile materials in the geologic medium, particularly after the plutonium has decayed to ^{235}U. Any differences in dissolution, migration, or reprecipitation between the fissile materials and the neutron absorbers could result in concentration of fissile materials over geologic time separeted from the neutron absorbers, possibly leading to a criticality incident in the future.

The volume of waste is large, if plutonium concentration is kept to only a few weight percent, with about 500 to 2500 canisters required to contain 50 metric tonnes of plutonium, or about 20 to 100 kg of plutonium per canister. The number of canisters could be reduced at the cost of greater risk of future criticality in the repository.

The "spent fuel standard" protection would be quite short-lived. The half-life of ^{137}Cs is only about 30 years, so after about 100 years, it would be feasible to mine the fissile material with very unsophisticated equipment, such as modified excavation machinery without workers incurring high radiation doses.

Reactor Options

There are four plutonium disposition options that involve burning of mixed Pu-U oxide fuel (MOX) in reactors to reduce the plutonium abundance, to convert the plutonium into spent fuel with the isotopic composition reduced in ^{239}Pu, and to make use of some of the energy content of the plutonium by generation of electricity. The options differ in the type of reactor they would use. The four options examined are existing light water reactors (LWRs), partially completed light water reactors, new evolutionary light water reactors, and the Canadian heavy water moderated reactors (CANDU).

All of the reactor options share the advantages that they would generate energy while converting the plutonium into a spent fuel waste form that is similar to the spent fuel currently destined for deep geologic repository disposal. They would also convert the isotopic compositon of the plutonium into one that is less attractive for diversion than the original weapon-grade material. If conventional reactor-grade plutonium is used in mixed oxide fuel, the abundance of ^{239}Pu reduces from 61% of the plutonium content to 24 to 31%, depending on the type of reactor used (Paratte et al., 1995). The reduction in total plutonium content is about 33 and 25%, respectively for these cases. For burning of weapon-grade plutonium, which is greater than 90% ^{239}Pu, the reduction in both isotopic content and total plutonium abundance would be higher.

The disadvantage of using mixed oxide fuel for plutonium disposition is that it might be seen to indicate a government position that favors development of fuel cycles that include MOX fuels, which is contrary to the position of the present administration (White House Press Office, 1993). These fuel cycles are presently supported by PUREX reprocessing of light water reactor fuels to extract the plutonium to make the MOX fuels, and there is concern about the proliferation risk with separated plutonium.

A new, non-fertile fuel option

The PEIS for disposition of surplus weapon-grade plutonium did not evaluate non-fertile fuels for plutonium burning. This type of fuel has been investigated in other countries and is being evaluated for burning of plutonium separated during reprocessing of spent commercial reactor fuels. One of these new fuels for light water reactors is being developed at the Paul Scherrer Institute in Switzerland (Degueldre et al., 1996; Paratte et al., 1995). The fuel would consist of 80-90% ZrO_2, 7-14% PuO_2, and 3-6% Er_2O_3 (atom percent), with the potential addition of Y_2O_3 to aid in stabilization of the fluorite phase structure. This fuel was designed to function in conventional light water reactors, with the intent to burn plutonium that was separated from spent fuel, but also to provide a durable waste form after burning the zirconia-based fuel.

Paratte et al. (1995) calculated the performance of the zirconia-based fuel and mixed oxide fuel for burning reactor grade plutonium under the same reactor conditions for a burn-up of 52 MWd/kgHM (as measured for the MOX fuel). The initial loading of plutonium assumed in the fuels was 6.1 wt% Pu in natural uranium for the MOX fuel and 16.5 wt% Pu in the zirconia fuel (corresponding to 7.5 atom % Pu with 3.5 atom % Er). The density of the zirconia fuel would be 7.28 g/cm^3, while a typical MOX fuel would have a density of about 11 g/cm^3. Thus, the initial amount of plutonium per cubic centimeter would be 1.2 g in the zirconia fuel and 0.66 g in the MOX fuel. (Less plutonium is needed in the initial MOX fuel because more ^{239}Pu is bred and burned in the MOX during reactor operation.) Table 1 shows a mass balance for plutonium based on 100 atoms loaded into the fuel for each fuel at the beginning and end of the fuel cycle. The initial isotopic compositions of the plutonium assumed in each case were slightly different; the reason for this was not explained in Paratte et al. (1995), but the differences are small enough to allow comparison of the general effects of burning plutonium in the two fuel types.

Table 1: Comparison of plutonium burning in MOX fuel and zirconia-based fuel for the same amount of thermal energy generation. Initial plutonium content of 6.1 wt% for MOX fuel and 16.5 wt % for zirconia fuel. Figures represent numbers of atoms per hundred initial plutonium atoms.

Plutonium Isotope	MOX fuel		Zirconia fuel	
	Initial	Final	Initial	Final
238	1.3	1.3	2.7	1.96
239	60.8	23.9	54.5	6.49
240	24.7	21.7	22.8	15.07
241	8.7	12.3	11.7	9.06
242	4.5	7.6	8.3	10.23
Total Pu	100.0	66.8	100.0	42.80

As can be seen in Table 1, the fissile isotope ^{239}Pu under these conditions would be reduced to 12% of its original amount in the case of the zirconia fuel, but with the MOX fuel the end-of-cycle ^{239}Pu would still be 39% of the original value. Almost no neptunium would be created in the zirconia fuel case, while 0.4 units of ^{237}Np awould be created in the MOX fuel. The amounts of Am and Cm created in zirconia and MOX fuels would be comparable (Paratte et al., 1995).

Degueldre et al. (1996) calculated comparative cases for MOX fuel with approximately 0.6 g/cm^3 Pu (2.51 mole/L initial Pu), zirconia-based fuel with reactor grade Pu at 0.8 g/cm^3 Pu (3.36 mole/L initial Pu) and zirconia-based fuel with weapon-grade Pu (2.65 mole/L initial Pu). Calculations were done for solid fuel pellets and a full core load of each fuel for irradiation of 1300 days in a light water reactor to produce 4.2 x 10^6 MWd thermal energy. Table 2 compares the amount of plutonium in those fuels with that in standard light water reactor fuel after burning to the same level of energy generation. The abundance of ^{243}Am is also shown because this isotope eventually converts to ^{239}Pu ($^{243}\mathrm{Am} \rightarrow {}^{239}\mathrm{Np}$ + alpha $\rightarrow$ ^{239}Pu + beta).

Table 2: Comparison of the plutonium content of fuels after irradiation for 1300 days to produce 4.2 x 10^6 MWd thermal energy from a full core load. Initial plutonium content: MOX = 2.51 mole/L; Zirconia fuel with reactor grade (RG) plutonium = 3.36 mole/L; Zirconia fuel with weapons grade (WG) plutonium = 2.65 mole/L; UO_2 spent fuel = 0.

Final conc. mole/L	MOX	Zr-RG	Zr-WG	UO_2 spent fuel
Total Pu + ^{243}Am	1.76	1.22	0.64	0.46
^{239}Pu	0.70	0.11	0.086	0.23
^{240}Pu	0.52	0.42	0.26	0.12
^{241}Pu	0.31	0.24	0.15	0.072
^{242}Pu	0.18	0.37	0.11	0.033
^{243}Am	0.053	0.082	0.03	0.009

Table 2 clearly shows that the efficiency of destruction of both ^{239}Pu and total plutonium in the zirconia fuel would be far greater than for MOX fuel. This is because there would no uranium in the zirconia-based fuel, so no ingrowth of new plutonium would occur. The final concentration of plutonium in the case of weapon-grade plutonium in the zirconia-based fuel would not be much higher than in conventional light water reactor fuel and it would contain very little of the fissile ^{239}Pu isotope. This is a distinct advantage with respect to final waste disposal because it would simplify considerations related to the potential for criticality to occur in the repository over long time frames, where both ^{239}Pu and its fissile daughter product ^{235}U must be considered.

Zirconium dioxide occurs in nature as the mineral baddeleyite. This would provide the possibility to use the natural mineral as an analog to the zirconia-based fuel in the assessment of the long term performance of the fuel as a waste form. Baddeleyite is highly resistant to weathering, which suggests that the zirconia-based fuel should be a highly durable waste form. Provided that segregation of fission products in the fuel does not occur to a much greater extent than in standard uranium dioxide fuels, the zirconia fuel is likely to be simpler to assess as a waste form because there are no oxidation processes to consider for the zirconia. With uranium dioxide spent fuels, assessment of the potential for the spent fuel uranium (IV) to oxidize to the more soluble uranium (VI) is an essential part of the evaluation of long term performance for the waste form.

The chief drawback of the zirconia-based fuel is that it is in the early stages of development. This should not, however, be seen as an impossible drawback. The first achievement of a sustained nuclear reaction occurred on December 2, 1942, and the startup of the first commercial light water power reactor occurred exactly 15 years later at Shippingport (Glasstone and Sesonske, 1967). By comparison with the task of inventing the commercial nuclear power plant, the testing and certification of a new fuel type for an existing reactor should be seen as achievable within a shorter time period. For the MOX fuel options discussed in the draft PEIS for plutonium disposition, the start of reactor operations was given as 2008. That schedule would allow 12 years to develop and test the new zirconia-based fuel prior to the scheduled start of operations.

One potential difficulty with the zirconia-based fuel is the relatively low thermal conductivity of the matrix material. In order to keep fuel temperatures at desired levels in the reactor, it would probably be necessary to use annular fuel pellets. This would add to the complexity of the fuel fabrication in comparison to standard MOX fuel.

The zirconia-based fuel is expected to be highly resistant to radiation damage in the reactor (Degueldre et al., 1996); however, testing under actual operating conditions is needed. One issue that must be addressed during testing is the effect of possible high power peaks at the interface between zirconia fuel and standard uranium dioxide fuels in the test reactor. The performance under mixed core loadings of uranium dioxide fuel and zirconia-based fuel might be different from that for either fuel loaded alone in a reactor (Paratte et al., 1995). The possible effects should be explored through calculated simulations prior to fuel testing.

Evaluation of the zirconia-based fuel against the plutonium disposition project screening criteria

Resistance to theft and diversion by unauthorized parties.

In common with all disposition options under consideration, there is a vulnerability to theft and diversion of material while the plutonium is in a pure state as either metal or oxide. Degueldre et al. (1996) recommend fabrication of the fuel using coprecipitation of plutonium and zirconium oxides (together with the rare earth elements used in the fuel) from nitric acid solution. This would be followed by pellet forming and sintering to the required density. Once the pellets are sintered, the resistance to diversion would be comparable to that of mixed uranium-plutonium oxide fuels.

Resistance to retrieval, extraction, and reuse by the host nation.

After the plutonium is incorporated into ziconia-based fuel pellets, but before irradiation, the plutonium would be harder to retrieve from the zirconia-based fuel than from mixed uranium-plutonium oxide fuel or from glass waste forms. It would be comparable in difficulty to retrieve from zirconium titanate ceramic waste forms. After irradiation, the weapon-grade plutonium is altered in isotopic composition to the extent that up to 95 % of the fissile ^{239}Pu is destroyed. This makes the residual plutonium much less attractive for weapon manufacture. Plutonium that is immobilized in glass or ceramic waste forms could be recovered by the host nation with varying degrees of difficulty and used for weapons after chemical separations were performed. Plutonium in MOX fuel after irradiation has an isotopic composition that is less attractive for weapons fabrication than weapon-grade plutonium or reactor grade plutonium because of the high abundance of ^{240}Pu relative to ^{239}Pu. The zirconia-based fuel after burning has an abundance of ^{240}Pu relative to ^{239}Pu that is even greater than in MOX spent fuels. (See Table 2).

Technical viability

All manufacturing steps needed to produce the ziconia-based fuel have been developed for other purposes, so production of the fuel should be possible without extensive research and development. Calculations of fuel performance have not revealed potential problems with the exception of low thermal conductivity. This can be handled by using annular fuel. Fuel integrity and fission product segregation need to be evaluated through irradiation tests in a light water reactor. There is no intrinsic reason to believe that the zirconia-based fuel would have substantially inferior performance to uranium dioxide or MOX fuels, so long as annular pellets were used.

Environmental, safety, and health compliance.

Environmental, safety, and health compliance during fuel fabrication for the ziconia-based fuel should be similar to that for mixed uranium-plutonium dioxide fuel fabrication. In some respects the environmental, safety, and health impacts of the zirconia-based fuels may be lower because handling of large amounts of uranium will not be necessary. After burning in the reactor, the amount of plutonium in the spent fuel and the proportion of the plutonium that is fissile are much less in the case of the zirconia-based fuel than for the mixed uranium-plutonium oxide fuel. This reduces the potential for plutonium remobilization and the potential for a criticality event after disposal of spent fuel in a geologic repository.

Cost-effectiveness

Costs of manufacturing annular fuel pellets for zirconia-based fuel might be higher than costs to manufacture pellets of mixed uranium-plutonium oxide fuel for a given amount of fuel. The potential exists for using higher plutonium loadings in the zirconia-based fuels than in the mixed uranium-plutonium oxide fuels, so the potential cost increases in fuel fabrication might be offset by the need for less fuel. There are no intrinsic reasons that reactor operation costs should be greatly different for using either of the fuels in light water reactors. There would be costs associated with testing the zirconia-based fuels prior to use for burning plutonium that would not be as high for mixed uranium-plutonium oxide fuels, since considerable development has already been carried out on these fuels.

Timeliness

The use of zirconia-based fuel would require more development work than the use of mixed uranium-plutonium oxide fuels. The scheduled start of reactor burning for the mixed uranium-plutonium oxide disposal options is 2008. A testing period for the new fuel type of six years should be more than adequate to determine whether the fuel would have the desired performance characteristics in the reactor. Thus, the 12-year period available before start of implementation seems adequate.

Fosters progress and cooperation with Russia and other countries (establish standards for disposition of surplus weapons-usable fissile materials, support negotiations for bilateral and multilateral reductions in these materials, and allow for international inspections at all stages)

The United States has taken the position that because of concerns over the potential for proliferation of plutonium and possible diversion of material, the government does not encourage the civil use of plutonium and does not, therefore, engage in reprocessing of reactor fuel for nuclear power or nuclear explosives purposes (White House Press Office, 1993). This policy was issued in 1993, at which time the only developed options for civil use of plutonium were mixed uranium-plutonium oxide fuels and breeder reactors. It is most likely that the discouragement of civil use of plutonium was intended to prevent the potential for increasing the inventory of separated plutonium associated with civil power generation.

If the intent of the US government policy toward use of plutonium is to decrease the amount of plutonium in existence, development of the zirconia-based fuel would help achieve that goal. As is discussed below, use of the zirconia-based fuel could not only burn weapons grade plutonium, it could also be used to destroy plutonium extracted from commercial reactor fuel. This would remove that plutonium from potential diversion or use in a national weapons program. The use of the zirconia-based fuel would require that reprocessing of spent light water reactor fuel be done; however, the plutonium extraction process could be designed so that international control would be maintained at all stages where the plutonium was available in relatively pure form. A processing technology that could achieve this is described in a later section.

The Russians have stated on many occasions that plutonium is an energy resource and should be used to generate power. Use of the zirconia-based fuel would acknowledge the validity of their position, while eliminating the proliferation risks that could occur with some mixed uranium-plutonium fuel operating conditions.

Establishment of an international program for control of civilian power reactor fuels and the plutonium extracted from them would be a major step forward. The National Academy (1994) noted that commercial reactor fuel should also be considered as weapon-usable. Establishment of a controlled spent fuel processing program with zirconia-based fuel to burn the plutonium would greatly reduce the amount of weapon-usable plutonium in the world.

Public and institutional acceptance

The degree of public acceptance for burning plutonium in zirconia-based fuel should be at least as great as that for using mixed uranium-plutonium oxide fuel once the zirconia-based fuel is demonstrated to be safe and effective under reactor operating conditions. The degree of institutional acceptance should also be the same, with the exception of the case of using an existing commercial power reactor that is still being used to satisfy civilian electrical needs. The amount of down time for the reactor and removal of power from the supply grid when using zirconia-based fuel in the first years of its use might be more than for a uranium-based fuel.

Additional benefits (including the ability to leverage the government investments for disposition of surplus materials to contribute to other national or international initiatives).

The chief additional benefit for using the zirconia-based fuel for burning surplus weapon-grade plutonium would be that the plutonium burning could be used to demonstrate the usefulness of this fuel for commercial applications. The data on performance of the fuel obtained as part of the surplus weapon-grade plutonium disposition task could be used to license the fuel for use in existing power reactors in the United States and other countries. Plutonium that is being separated from commercial reactor fuels in France, Great Britain, and eventually Japan, could than be burned in the zirconia-based fuel instead of being used in mixed uranium-plutonium oxide fuel.

Future application of zirconia-based fuels to reduction of plutonium generated by commercial nuclear reactors

During irradiation of light water reactor fuels some of the ^{238}U atoms capture a neutron and are converted to ^{239}U, which decays within days to ^{239}Pu. By the end of a normal burning period the spent fuel contains about 1% by weight of total plutonium, which is about 60% fissile ^{239}Pu. Worldwide inventories of spent fuel as of 1990 contained about 650 tonnes of plutonium. Estimates of plutonium inventory increases due to spent commercial reactor fuel were 70 tonnes per year from 1990 to 2010 (Albright et al., 1993). While the plutonium isotopic composition in reactor-grade plutonium is lower in ^{239}Pu than in weapon-grade, the National Academy Committee on International Security and Arms Control considered that the plutonium was weapon usable (NAS, 1994). Since the plutonium inventories are growing rapidly and in many states that are not nuclear weapons states, it seems prudent to consider the long-term control of these inventories.

Zirconia-based fuel could be used to burn commercial reactor grade plutonium. Indeed, this was the original purpose for the fuel (Degueldre et al., 1996). The Purex process currently used to reprocess spent uranium dioxide fuels requires large equipment and generates considerable secondary wastes. If processing of light water reactor fuels is to be done in order to destroy the plutonium, it would be advantageous to use a different method to separate the plutonium and reduce ultimate wastes. The electrochemical methods described below have the advantage of requiring smaller equipment that is amenable to modular construction, and the process separates actinides from fission products so that the fission product wastes require only 200 to 300 years storage to decay to low activity, and might not need deep geological disposal.

Electrometallurgical Treatment of Spent Nuclear Fuels

The electrometallurgical treatment process was originally designed for treatment of metallic spent fuels. The heart of the process is the electrorefiner, which dissolves metal fuels anodically while depositing a very pure uranium product at the cathode. The transuranium (TRU) elements dissolve in the molten salt electrolyte as chlorides, and the fission product metals that are more noble than uranium (such as zirconium and the noble metals) remain behind in the anode compartment. The fission products that are less noble than uranium dissolve in the molten salt electrolyte along with the transuranium elements. The TRU elements could be recovered, for use in the zirconia-based fuel, by use of a special liquid-cadmium cathode. The function of the liquid cadmium is to reduce the chemical activity of the TRU metals enough to allow them to be deposited (as $AnCd_6$ in the liquid cadmium, where An is plutonium, the lower abundance higher actinides, and some uranium) in the presence of uranium. The cadmium is later distilled for recycling in the process, and the TRU metal product is melted to form an ingot. The TRU product thus recovered is not suitable for weapon use, because it contains all of the TRU elements (plutonium, neptunium, americium, and curium) as well as some uranium and some of the rare earth fission products. However, the TRU product would be suitable for fabrication of the zirconia-based fuel. A generalized flowsheet for the electrometallurgical treatment process is shown in Fig. 1.

Commercial light water reactor spent fuel is of most interest to this study. These fuels are primarily UO_2 with about 1 wt% TRU elements and about 3 to 5 wt% fission products, depending on burnup. Oxide spent fuels are handled in the process by first chopping the fuel to expose the oxide matrix, then reducing the oxides to metals. The oxide reduction is done using lithium reductant in molten LiCl salt. The Li_2O that is formed in the reduction reaction dissolves in the LiCl. The lithium is later regenerated, to allow recycling of the LiCl, by electrochemical decomposition of Li_2O to form lithium metal and oxygen gas. The oxygen is vented to the atmosphere. The reduced metals are transferred to the electrorefiner for recovery of the uranium as a pure product and the TRU elements for use in the zirconia-based fuel. After the uranium is recovered, the electrorefiner potential can be raised (from 0.45 V to 1.0 V) to recover the zirconium metal from the Zircaloy cladding, if desired. This zirconium could be used to make the matrix material for the zirconia-based fuel.

The wastes from the electrometallurgical treatment process include a metal waste form made from the fuel cladding, fuel assembly hardware, and noble-metal fission products, and a mineral waste form made from zeolite-stabilized salt in a glass matrix. The zirconium-stainless-steel metal waste form matrix is a very corrosion-resistant metal (far more corrosion-resistant than borosilicate glass), and the noble metal fission products placed in this waste form will have decayed (except for ^{99}Tc) after ~100 years. The glass-bonded zeolite waste form is also very stable. Even though it is in the early years of development [Lewis, 1995], it has been shown to have a leach rate, based on the MCC-1 test method, comparable to that of borosilicate glasses.

When the TRU elements are recovered in the electrorefiner for use in the zirconia-based fuel, both the metal and mineral waste forms can be made non-TRU (i.e., <100 nCi of alpha activity from elements with atomic number greater than U per gram of waste). Therefore, the requirements placed on the geological repository for containing these wastes are significantly reduced. In fact, the argument can be made that these wastes may not need to be placed in a geologic repository at all, but can be stored above ground until their specific activity is low enough for contact handling (about 200 years) and disposal as low level radioactive waste.

Proliferation Concerns and Control of the Light Water Reactor Fuel Cycle

Although the electrometallurgical treatment process cannot produce weapon-useable material directly, some will still be concerned about possible proliferation of fissile materials for weapon applications. This could occur if the plutonium product were to be diverted and subjected to further chemical separation using Purex-like methods. The electrometallurgical treatment process is a compact, high-temperature process that would be easy to modularize and to inspect. Use of this technology could be achieved by placing modular units at appropriate sites and developing stringent use-control procedures. The units would be “turned on” for a production campaign in the presence of IAEA inspectors, then “turned off” at the end of the campaign. Sealing and alarming the doors of the facilities would provide remote inspection assurance that the facilities were not used in the absence of IAEA inspectors.

One modular unit of about 150 m^2 area could provide enough TRU and zirconium to keep a 1000-MWe power plant running, with one 100-day fuel processing campaign each year. Such a power plant would consume about 3-kg plutonium (or TRUs) per day, or about 880-kg TRU per year (assuming 80% plant factor). The zirconia-based fuel contains about 40% of the originally loaded mass of plutonium at the end of burning (see Table 1), so about 1500 kg of plutonium would be needed to make the fuel. Light-water reactor spent fuel typically contains about 1 wt% TRU. In a 150-day campaign, 10-kg TRU/d could be produced by processing 1 MTHM of LWR spent fuel per day. Thus, the processing plant capacity would be on the order of one metric tonne of heavy metal per day, operated 150 days per year, under constant IAEA inspection. This plant would process the spent fuel from an equivalent of 5 to 6 years of fuel discharge from uranium dioxide light water reactor fuel. Depending on local conditions, the plant could be run continuously by transporting fuel from several reactors to the processing site, or could be run in campaigns using the fuel from a single reactor and transporting the work force between reactor and processing sites.

Conclusions

Use of electrometallurgical processing of light water reactor fuel under international safeguards and supervision followed by burning of the plutonium in zirconia-based fuel could reduce worldwide plutonium inventories and decrease the danger of diversion of reactor-grade plutonium. The burning of surplus weapon-grade plutonium in zirconia-based fuel could provide the data on fuel performance needed to support the licensing of the zirconia-based fuel for commercial use. The zirconia-based fuel provides for more efficient destruction of the fissile plutonium, reducing concerns about nuclear criticality in the long term after disposal of spent fuel from the plutonium burning cycle. The environmental concerns related to fabrication and use of the zirconia fuel are sufficiently similar to those for MOX fuels that they fall within the range of reactor burning cases examined in the PEIS for plutonium disposition. We recommend adding the zirconia-based fuel to the MOX fuel options in the final PEIS.

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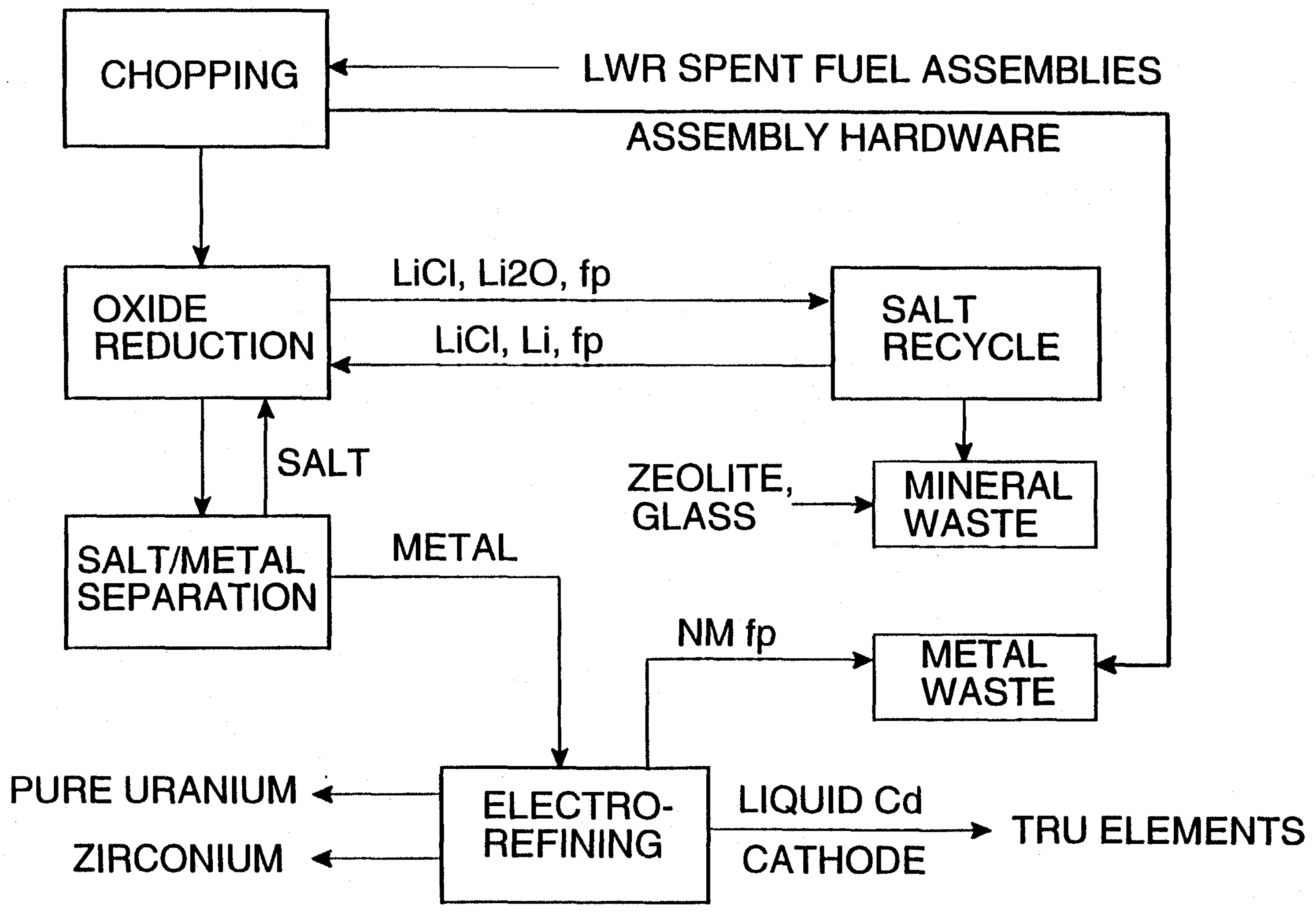

Fig. 1 Electrometallurgical Treatment Process for Light Water Reactor Fuel, with Zirconium Recovery for Use as Fuel Matrix Material.

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