Interim Storage Matrices for Excess Plutonium: Approaching the "Spent Fuel Standard" Without the Use of Reactors

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Introduction

At the present time, both the United States and the former Soviet Union (FSU) are proceeding with the dismantlement of decommissioned nuclear warheads, at a rate of about 2000 each per year, although such a step is not currently required by any international agreement. As a result, large quantities of weapons-grade plutonium, in the form of metallic "pits" (precisely cast, grapefruit-sized, hollow spheres), are accumulating in storage facilities in both nations. It is expected that only a fraction of this material will be designated by the military as an essential strategic reserve; the remainder, believed to be about 50 tonnes on each side, will be declared "excess" (or "surplus") and eventually transferred to civilian control. Determining how best to manage and dispose of the excess plutonium safely and securely has proven to be an extremely complex issue, as a number of recent studies have illustrated, including a highly-publicized one conducted by the National Academy of Sciences (NAS).1,2

The NAS study recommended the following strategy for management of the excess plutonium. Initially, there should be a verified halt to the production of new material for weapons ("fissile cutoff"), accompanied by reciprocal declarations of weapons-usable plutonium stockpiles. Excess plutonium pits should then be placed in internationally monitored interim storage, where they would be subject to stringent security and safeguards. Because the pits are easy to handle and only one step removed from use in weapons, the NAS study emphasized that a security and accounting system as rigorous as the one employed for nuclear warheads will be necessary, which it characterized as meeting the "stored weapons standard" (SWS).

Practical considerations dictate that the surplus plutonium will have to be stored in pit form for a period of at least one decade, and probably longer. However, there are a number of reasons why such storage would not be acceptable in the longer term. First of all, since the plutonium is in a form which can be returned to weapons with little difficulty, the situation provides an unstable technical basis for the disarmament regime. Even with political controls in place, the prolonged storage of a large number of weapons components would be likely to fuel suspicions among other nations. Second, maintaining a SWS safeguards system indefinitely would impose a costly and cumbersome burden on generations to come.

In response to these concerns, a variety of "disposition" options, i.e. alternatives to long-term pit storage, have been proposed. The NAS study suggested that a primary objective of any disposition method should be the conversion of the plutonium from a form requiring SWS-type safeguards to one which meets the "spent fuel standard" (SFS); that
is, to a form in which its accessibility is reduced to a level comparable to that of the plutonium contained in unprocessed spent fuel. Such a step, while obviously not a definitive solution, would nevertheless eliminate the special security risks that distinguish the storage of separated plutonium from spent fuel. Fissile material meeting the SFS may be stored under a security and safeguards regime far less intensive than one adhering to the stored weapons standard.

Conversion of separated plutonium to a form meeting the SFS would diminish its accessibility in a number of ways. The plutonium would be bound in a large, heavy matrix with fission products that generate high levels of penetrating gamma radiation, rendering impossible unshielded handling of the material. Also, it would be diluted to a low concentration with uranium-238 or other non-fissile materials. Those wishing to recover the plutonium would require access to a heavily shielded, remotely operated reprocessing plant.

The value of the conversion of excess plutonium to the spent fuel standard is dependent on the political context in which it is carried out, as well as on the threat being addressed. The most immediate benefit would be to reduce the danger of theft of the material by sub-national groups, by diminishing its attractiveness and supplementing security measures with an inherent physical barrier. Although few regard sub-national theft as a credible scenario with respect to the US inventory, it is a serious issue in the FSU, where the persistence of economic crisis, ethnic conflict and widespread corruption raises doubts concerning the ability of the central government to enforce its authority.

Also, firm commitments to dispose of excess plutonium stockpiles would be an important demonstration to the international community that the former superpowers are sincere in their intention to achieve lasting nuclear arms reductions.

Not everyone agrees that expedited plutonium conversion is a worthwhile objective. For example, some nuclear materials specialists at Los Alamos National Laboratory (LANL) have argued that pit storage, even for an indefinite period, is preferable to options involving processing and dilution. Their argument consists of two parts. First, they claim that there is little point to conversion to the SFS, because any such process is in principle reversible. Second, they contend that plutonium conversion processes themselves entail unacceptable environmental and diversion risks, whereas decades of experience with storage in pit form has demonstrated it to be a safe procedure. They also maintain that the attractiveness of pits to sub-national groups is overemphasized, because they have been designed for use in state-of-the-art warheads and are not directly usable in the crude bombs within the technological reach of terrorists.

These arguments contain some valid points, but are ultimately not very compelling. The fact that SFS conversion processes can be reversed does not mean that it is a simple matter to do so. Even if diversion of the converted material were to take place, recovery of the plutonium in weapons-useable form would require the construction and operation of a clandestine reprocessing plant. Consequently, the time period necessary for "breakout" to occur (the return of a significant fraction of the plutonium to weapons use), as well as the probability of detecting the diversion, would be greater than if the plutonium had remained in pits. The resulting delay could buy time for new political initiatives to resolve the situation peacefully. Therefore, conversion of surplus plutonium to the spent fuel standard would be effective in reducing the threat of covert breakout.

To the potential for risks that may be incurred during processing must be seriously addressed. However, a possible short-term increase in vulnerability should be balanced against the substantial longer-term security benefits of the procedure. With care, the process risks can be minimized. Some conversion methods present greater concerns than others, and this should be an important consideration in the assessment of disposition options.

Finally, it strains credulity to believe that weapons-grade plutonium metal pits would not be an extremely attractive material for prospective bomb-builders, even if some metallurgical processing were necessary. The recently documented theft of military plutonium from Russia, which was highly diluted with other, non-radioactive elements, suggests that the accessibility and ease of handling of the material is far more relevant to its susceptibility to theft than whether it can be immediately used in weapons.

One suspects that, in addition to technical considerations, the strong support of pit storage among certain groups in the weapons complexes of both the US and Russia may also be based on a general reluctance to take further steps that would reduce the flexibility of their respective nuclear arsenals. Counterbalancing this resonatory and dangerous attitude is one of the primary targets of the spent fuel standard approach.

Conversion of pits to the spent fuel standard may also facilitate the application of safeguards to plutonium storage, even though a degree of measurement precision would be lost. This is because mutually acceptable monitoring procedures for plutonium in classified shapes would probably be limited in ways that inhibit their efficacy. For example, international inspectors may be prohibited from direct viewing of the pits, and only allowed to inspect the container in which they are stored, which would introduce a weakness into the verification regime. The destruction and dilution of pits may enable these restrictions to be lifted to a large extent, so that more direct monitoring of the storage forms would be permissible without the concern that sensitive design information would be disclosed.

Because of the physical limitations of the disposition options under consideration, plutonium conversion could not begin for at least a decade, as will be demonstrated in the next section; some processes, such as the use of fast reactors being advocated in Russia, could require considerably more time. In addition, the time necessary for processing a plutonium inventory of 50 tonnes ranges from ten to forty years, depending on the option chosen and the resources available for the project. Thus, implementation of the NAS strategy would mean that some plutonium could remain in pit form for fifty years or longer. Complications arising in the disposition process could lead to further delays.

In view of the instabilities characterizing the Russian political situation, maintaining pit storage for fifty years may seem wise. Nonetheless, the NAS study did not favor approaches in which the pits would be converted for interim storage to a different form, with a diversion resistance intermediate between pits and spent fuel. It was judged that the additional processing required would increase costs and divert attention from the main
goal: preparing the material so that it meets the SFS and is suitable for final disposition.

Present circumstances, however, may require the consideration of a wider range of options for the management of excess plutonium than those recommended by the NAS study. In this paper, the rationale for implementing a strategy including interim conversion of pits is discussed, and several candidate conversion processes, that do not require the use of nuclear reactors, are assessed. One concept, based on a process known as AIROX, appears to be especially promising; not only for interim conversion, but also, with suitable modifications, as a means of final plutonium disposition not considered in the NAS study.

Methods of "spent fuel standard" plutonium conversion

The NAS identified two distinct approaches for converting separated plutonium to a form that meets the spent fuel standard. The first is to irradiate the plutonium by using it as fuel in nuclear reactors. A variety of reactor types and deployment scenarios have been proposed for the task. The second approach is to bypass the use of reactors entirely, and instead combine the plutonium with liquid high-level radioactive wastes (HLW), which have accumulated in large volume from past plutonium production. The resulting mixture would then be blended with glass-forming materials, melted, and cast into large glass logs. This process, known as vitrification, has already been selected in the US and other nations as a means of stabilizing HLW inventories for final geologic disposal.

The reactor options have been advocated by those who believe it is important to extract the energy content of the plutonium, which was produced at enormous financial and environmental cost. Revenue from the sale of the generated electricity could provide partial compensation for the expenses of disposition. However, close examination of the numerous reactor options has shown that most are not likely to rank highly with respect to criteria such as cost, feasibility, timeliness, ease of licensing and public acceptability.

The reactor concept judged by the NAS study as having the best chance for success, within existing constraints, is based on dedicated light-water reactors (LWRs) operating on a once-through cycle with full cores of mixed plutonium-uranium oxide fuel (MOX). Even along this route, the regulatory roadblocks in the United States, where today the licensing of any nuclear facility is very difficult, are expected to be formidable and time-consuming. A number of facilities will require regulatory approval, from the MOX fabrication line to the spent fuel storage pools, which will need modifications to reinforce criticality safety. Although MOX technology for LWRs has been demonstrated at an industrial scale in several nations, in both the United States and Russia the necessary facilities can be regarded as essentially first-of-a-kind with regard to licensing and operating experience. Furthermore, even internationally, there has been little experience with operating full-core MOX LWRs.

The strategy that could probably be initiated in the shortest time would be the recruitment of currently operating reactors that are capable of full-core MOX loading without modification. The three Palo Verde reactors in Arizona, which are of Combustion Engineering System-60 design, are the only ones that fit this description. For MOX fabrication, the NAS proposed adapting an unused facility at Hanford, the Fuels and Materials Examination Facility (FMEF). While it is difficult to estimate the amount of time necessary before the consumption of MOX could begin in this scenario, it is not likely to be less than ten years, and probably more, given the lengths of typical regulatory proceedings today, and a realistic fuel fabrication schedule.

Once the Palo Verde reactors began full-power operation, an excess plutonium inventory of 50 tonnes could be processed in about 60 reactor-yr, for a burnup of 33 GWd/t and fissile enrichment levels similar to current practice. Since it is likely that only two of the reactors would be reliable enough to safely use MOX, the campaign would take 30 years to complete, not ending sooner than 40 years from today.**

** In Russia, formal procedural arrangements for the licensing of nuclear facilities do not exist as they do in the US. However, it is a goal of the international community to foster the development of the recently created nuclear safety agency, GOSATOMNADZOR, to play the role of an independent and authoritative regulatory body. It is uncertain in the current political environment in Russia whether this will soon happen.

As an example of current regulatory timescales, consider the Nuclear Regulatory Commission (NRC) approval process for the design of the General Electric Advanced Boiling Water Reactor (ABWR). The safety review of the design took seven years to complete, and it is estimated that the rulemaking (public comment) period will take an additional eighteen months. The ABWR is an "evolutionary" reactor concept that contains only incremental improvements relative to existing designs. Similarly, the licensing of the Palo Verde reactors to burn MOX would involve reviewing and testing incremental changes in existing reactor designs and may take nearly as long. Also, safety issues already present there, such as crack formation in the steam generators, would have to be resolved. Repairs and replacement of equipment may be necessary.

In addition, prior to the NRC assessment, the DOE must complete a programmatic environmental impact assessment (FEIR) on plutonium disposition. The former is not scheduled for completion until early 1996, and delays of months are not unusual. Conservatively assuming a five year period of safety review and public comment, the licensing would be completed in about 2001. About another year would be necessary between commencement of core loading and full-power operation. However, it is unlikely that a MOX core would be available for loading that soon. The NAS study reports that an estimated five years would be required to install a 50 t/Mo MOX fabrication line in the FMEF, after a permit were obtained (which itself would take a few years). Two Palo Verde reactors fueled with MOX would require 120 cores of about 100 t/MO each. Fabrication of the first core would take at least two years, and probably longer, since the plant would not achieve maximum throughput immediately. Therefore, the first core would not be ready until around 2006.

** One of the reactors (Palo Verde 1) has one of the worst operating records in the world (313/356 with respect to lifetime load factor as of June 1992). (World Nuclear Industry Handbook 1993, Nuclear Engineering International Special Publications, 1989.) The Palo Verde reactors have been operating since the mid-1960's, so assuming that MOX burning does not begin until 2009, they will only have about 20
In analyzing the environmental impacts of the MOX option, it is important to note that, of all the processing stages it requires, the operation of the reactors themselves will be associated with the greatest risks. (See the section entitled "Some Environment, Safety and Health Issues," below.) Therefore, the reactors are likely to elicit the most strenuous public opposition and present the most challenges to regulators.

Also, the potential to recover costs from sale of the electricity produced by utilizing the plutonium as MOX may not be very great. In the extremely competitive electricity market in the US resulting from passage of the 1992 Energy Policy Act, wholesale buyers may purchase electricity from the producer offering the lowest price. It has been estimated that current nuclear plants must lower their operating and maintenance costs by 5%-10% annually over the next decade to remain competitive. It is highly unlikely that a first-of-a-kind plant fueled with full-core MOX would be able to operate at a cost sufficiently low so that its electricity could be competitively priced without substantial government subsidy, especially if decommissioning costs are included.

In the FSU, recycle of excess plutonium in LWRS is not regarded as an attractive approach at all. Instead, the option preferred by officials in the Ministry for Atomic Energy (Minatom) is utilization of the plutonium as startup fuel for a planned series of fast breeder reactors (FBRs) known as BN-800s. Plutonium use in thermal reactors is viewed unfavorably for two reasons. First, it is seen as a waste of the excess neutrons that Pu-239 can supply from fast fission, which could otherwise be used for breeding more fissile material, either Pu-239 or U-233. Second, some argue that because long-lived actinides are fissioned more efficiently in a fast spectrum than a thermal spectrum, the latter approach provides greater long-term environmental benefits. (Similar claims in the US have been met with a great deal of skepticism, however.) Also, the existing expertise and infrastructure in Russia is oriented toward plutonium recycle in FBRs, rather than LWRS.

The vitrification option can achieve similar results to the MOX option, and since it does not involve the operation of nuclear reactors, it may be easier to license in the US. A vitrification line at the Savannah River Site in the US, the Defense Waste Processing Facility (DWPF), is already essentially complete. Preliminary studies have shown that the inclusion of plutonium into the feedstock of the vitrification plant, at concentrations of up to a few percent, is technically feasible in principle. The end product of this process, years left on their operating licenses. Therefore, implementation of a two-reactor strategy will require a license extension permit in addition to the other authorizations.

This point is underscored by the specific circumstances of the plants that are actually being considered for plutonium disposition. The increase in generating capacity that would be provided by completing the WNP-1 and WNP-3 reactors in Washington State is simply not needed, as a study by the Bonneville Power Administration determined. (Status Report: Nuclear Plant Construction in the US: Nuclear Engineering International, March 1994.) The other candidates, the Palo Verde reactors in Arizona, are experiencing steam generator problems, which may be a result of intrinsic flaws in the design ("Palo Verde Back at Full Power, but Six-Month Inspections Continue," Nuclear Week, July 14, 1994.) Prior to MOX use, the steam generators would probably have to be completely replaced, at a cost of $200 million per reactor.

cylindrical canisters of radioactive, plutonium-laced glass, would resemble spent reactor fuel with respect to fissile material dilution and gamma radiation field intensity. The ultimate fate of the glass would be emplacement in a geologic repository. Since geologic repositories will not be available for decades, the glass will have to remain in interim storage for decades. However, such storage is not likely to differ significantly from interim spent fuel storage with regard to cost, safety or security. In the United States, an interim storage facility for vitrified HLW has been constructed adjacent to the DWPF with a capacity to accommodate the product of a campaign of vitrification of at least 50 tonnes of weapons plutonium, at a loading of 1.3 weight-percent.

The vitrification route, also referred to as "anti-reprocessing," has a number of attractive features. It is likely to offer a cost advantage relative to most reactor disposition options, since much of the expense would have already been charged to HLW management. Since it requires fewer facilities, entails less processing, and involves smaller material inventories than the reactor options, the associated security and radiological risks, due either to normal operations or potential accidents, would probably be lower as well. It is also distinct from the reactor options in that it would be much more consistent with the principle of nuclear "waste minimization," since no additional fissile products would be generated.

Despite these advantages, the vitrification option is controversial, since it carries an explicit acknowledgment that separated weapons plutonium has no commercial value and should be treated as waste. Although the plutonium could be recovered from the glass cylinders at a later date, if desired, using conventional reprocessing technology, it is hard to imagine circumstances under which there would be an economic incentive for doing so. Anti-reprocessing is therefore very unpopular in the nuclear communities both in the US, where the plutonium disposition issue is seen as an opportunity to rekindle the ailing nuclear industry, and in Russia, where plutonium is often referred to as a valuable national resource which should not be wasted.

Furthermore, there are several outstanding unresolved safety issues regarding vitrification. In the US, the start-up of the DWPF has been repeatedly delayed due to problems encountered in the pretreatment processes for the HLW feed, which are necessary in order to remove substances that could cause explosions in the melter. Clearly, such hazards must be eliminated with high confidence before the addition of plutonium to the feedstock can be considered. Questions involving the potential for criticality incidents have also been raised, both in the melter and in the final repository.

The pace of plutonium vitrification operations is an important consideration. The
time necessary for the campaign would be on the order of 6-10 years, depending on the plutonium loading in the glass and the plant throughput achieved. This is considerably less than the MOX scenarios described above would require. However, because the primary mission of the DWPF is HLW vitrification, addition of plutonium would be deferred until the last decade of plant operation. According to current estimates, this implies that the conversion of US excess plutonium could commence between 2006 and 2011 and be completed in the period 2012-2021. At least three reactors fueled with full-core MOX would be needed to finish the job on a similar schedule.

At the Hanford site, construction of the planned vitrification plant has been postponed indefinitely, primarily because the plant capacity requirements are still unknown, although recently a proposal to build a commercial facility there based on French technology has been discussed. This may provide an opportunity for designing the plant so that plutonium vitrification can be carried out with good assurances of safety. On the other hand, startup of such a plant is not likely to occur for more than a decade. In Russia, the vitrification process for HLW is based on phosphate glass, which is prone to crystallization (a deterioration of its material properties) and is not considered suitable for the inclusion of actinides. It is unlikely that vitrification could be regarded there as a serious option, even if the political will existed, unless their process were modified to produce the more stable and durable borosilicate glass. Such a modification may not involve more than changing the glass melt in the existing plant and flushing out the process equipment.

Both the reactor and vitrification routes will generate a product meeting the spent fuel standard. However, the isotopic composition of the plutonium each contains is different. Irradiation of "weapons-grade" plutonium (94% Pu-239, 6% Pu-240) in reactors will in general cause an increase in the ratio of Pu-240 to Pu-239, as well as increased concentrations of other plutonium isotopes. The final composition depends on the reactor type and the fuel burnup. "Reactor-grade" plutonium (90% Pu-239, 24% Pu-238) is the composition present in spent LWR fuel of 33 GWd/t burnup. Irradiation of weapons-grade plutonium in fast reactors, in contrast, does not have as great an effect on its isotopic composition, only resulting in a doubling of the Pu-240 concentration after a fuel burnup of 50 GWd/t. In addition, if fertile blankets are present in the core, the plutonium bred into them is weapons-grade. For plutonium vitrified with HLW, on the other hand, the isotopic content is obviously unchanged.

There is a widespread consensus that a nuclear explosive device can be made with reactor-grade plutonium, which is essential to consider when the risks posed by the U.S.-Russian plutonium disposition problem are analyzed. In the context of the international commerce in commercial plutonium are analyzed. In the context of the international commerce in commercial plutonium disposition problem, however, the significance of this point is less clear. In other words, it is impossible to say that under any circumstances, the attractiveness for diversion or theft of a waste form meeting the spent fuel standard will be independent of the isotopic composition of the plutonium it contains. Therefore, it is difficult to judge whether credit should be given to disposition options that are capable of degrading the plutonium isotopic content. The NAS study recommends that little or no significance be assigned to this property, but a deeper analysis is probably warranted. There is no imperative that both the US and FSU choose the same approach for plutonium disposition, but the schedules by which each nation plans to achieve specific goals should closely coincide, if an approximate parity is to be maintained. This could be difficult to accomplish if the US were to select vitrification at the DWPF, which could be completed as soon as 2012, whereas Russia continued to pursue the fast reactor option.

In order for Russia to adhere to a similar schedule, taking into account continued civil plutonium separation at current levels, it would have to have three or four BN-800 fast reactors operational by 2000-2005. Such a plan, although consistent with recent Minatom projections, must be regarded as hopelessly ambitious. The BN-800 program, currently without a clear source of funding, now consists only of one half-complete unit, which also must be redesigned to reduce the sodium void coefficient. In addition, many other safety issues related to fast reactor operation have not been fully resolved and would require careful study. Some have suggested that existing fast reactors in Russia and Kazakhstan could also be utilized for prompt weapons plutonium disposition, but in view of their safety records, this option is unlikely to attract much international support. The 6 May 1994 sodium fire at the BN-600 fast breeder at Beloyarsk does not inspire confidence in the existing generation of Russian fast reactors. For these reasons, the dedication of Minatom to fast reactors as a solution to the plutonium problem could result in extensive and undesirable delays.

The interim matrix compromise

In view of the fundamental (and perhaps irreconcilable) differences in the positions of those who regard the surplus plutonium as waste and those who consider it a resource, it may be fruitful to widen the discussion to include intermediate approaches. An ideal compromise might involve the relatively prompt incorporation of the surplus plutonium into an "interim" matrix that meets or nearly meets the spent fuel standard, in such a manner that either eventual utilization in reactors or direct geologic disposal would remain a viable alternative. This conversion would essentially decouple the fissile material "demilitarization" and final disposition stages of the disarmament process. Once this was accomplished, resolution of the numerous technical, regulatory and political issues relevant to the assessment of long-term disposition options could be conducted in an atmosphere of reduced urgency.

One of the first steps involved in the interim matrix approach is the blending of the separated plutonium with a highly radioactive diluent. Once the blending has taken place, the subsequent process steps, as well as interim storage of the converted material, would no longer have to be subject to security and safeguards conformance to the stored weapons standard. This would impose a far smaller burden on safeguards resources than a strategy such as pit storage and eventual incorporation into MOX, in which case the stored weapons standard would prevail throughout the entire cycle, until the point at which the MOX was loaded into reactors. However, there are other factors that must be taken into account. There may be additional economic and environmental costs incurred as a consequence of processing highly radioactive materials that must be remotely handled. Furthermore, some argu
that plutonium dilution processes result in a loss of accuracy of material accountancy that would increase the risk of diversion. The overall impacts of such tradeoffs must be carefully analyzed.

The interim matrix strategy would be most appropriate in the Russian context. The prospect of an open-ended period of plutonium pit storage in Russia, whose termination would be contingent on the construction of a fleet of breeder reactors, is not a very appealing one. However, Minatom may be willing to consider incorporating their excess plutonium into more diversion-resistant forms, if they can be convinced that the process will not substantially limit their flexibility in making future planning decisions.

Conversion of Russian plutonium pits to a form not readily usable in weapons may also increase the political acceptability of a possible sale of the material to the US, in analogy to the sale of Russian highly-enriched uranium.

Under certain circumstances, the US may find it necessary or desirable to consider an interim conversion strategy as well. These include 1) if Russia decided to proceed with such a plan, and requested strict bilateral reciprocity; 2) if it decided to defer to a later date the decision as to whether the energy content of the plutonium should be recovered; 3) if it proved difficult to resolve questions concerning process safety or geologic disposal raised by the final disposition options currently under consideration.

Clearly, some of the benefits of the interim matrix approach would be lost if the construction, licensing, or operation of the conversion facility itself proved to be difficult, time-consuming or prohibitively expensive. This suggests that acceptable processes would have to be conceptually simple and of demonstrably low risk. In this regard, the benchmark for comparison should be the most straightforward reactor option, full-core MOX in LWRs. It should be noted that the additional flexibility purchased by taking such an approach may be of sufficient value to justify it, even if it cannot be rapidly carried out.

For example, the NAS study considered and rejected an interim option that involved "spiking" the plutonium to a level far below the spent fuel standard by short-term irradiation in nuclear reactors. In view of the above discussion, the merits of such an approach are indeed questionable. Licensing of the "interim" method would be nearly as difficult as for the MOX disposition route, and implementation would probably not pose fewer risks.

Thus, if methods of plutonium conversion are to be useful for producing interim forms, they should not involve the use of reactors, should be capable of providing high levels of diversion resistance, and should be able to achieve processing rates greater than that of the MOX route while at the same requiring fewer facilities.

Furthermore, if a method with these characteristics were identified, it may even be suitable as a final disposition process, depending on the diversion resistance and the chemical stability of the product. Such alternatives may be worth considering if the technical problems with plutonium vitrification, the principal non-reactor disposition option, threaten to disqualify it as an option.

Some of the criteria which are important in judging the merits of interim storage matrices for excess plutonium are discussed below. Then, two blending processes which might be useful in the production of such matrices are described and analyzed with respect to these criteria.

Criteria for evaluating interim options

In order to compare the options for interim storage matrices given in the next section, it is necessary to identify some of the desirable characteristics that such matrices should possess. These include:

- High diversion resistance in interim phase:

One of the chief motivations for interim conversion is to substantially increase the "diversion resistance" of the excess weapons plutonium in storage. While it is impossible to develop a precise correlation between the physical characteristics of an interim matrix and a specified degree of diversion resistance, the spent fuel standard provides a useful benchmark.

Perhaps the most important parameter is the whole-body dose rate due to penetrating radiation to which an individual in the vicinity of the bare (unshielded) storage form would be exposed. The dose rate is related in a qualitative way to the physical harm and discomfort that would be experienced in handling and processing the material.

The gamma radiation dose rates generated by LWR fuel assemblies irradiated to typical commercial burnups have been recently calculated numerically, as a function of cooling time.\(^{19}\) The dose rate at one meter from the edge of the fuel assembly along the perpendicular bisector to the axis (hereafter referred to simply as the "dose rate"), which is the direction of maximum dose, was found to be about 60 Sieverts per hour (Sv/hr = 100 rem/hr), for 5-year cooled fuel of 33 GWd/ton.\(^{19}\) Vitrified HLW canisters would generate a dose rate that would be considerably lower, or about 9 Sv/hr, but the NAS study considers this rate to be high enough to also meet the spent fuel standard.\(^{20}\)

The International Atomic Energy Agency (IAEA) defines a "self-protecting" fissile material container as one which generates a dose rate equal to or greater than 1 Sv/hr. Commercial spent fuel will exceed this criterion for more than a century after discharge from the reactor. In view of the great disparity between the IAEA value and the dose rates characteristic of the majority of the world's spent fuel inventory, it should be regarded as an absolute minimum.

The time period over which a significant degree of self-protection persists is another important characteristic of an interim storage matrix. A dose rate that falls off too rapidly would be a significant disadvantage: It would be hardly worth the trouble to put plutonium into diversion-resistant forms if it were to buy only a few years of protection.

Both the magnitude of the dose rate generated by an interim storage matrix and its rate of decrease are functions of the particular radionuclide composition of the diluent. In Table A1 of the Appendix, the radionuclides in spent fuel which are the major

\(^{19}\) To put this into perspective, an individual exposed to a dose rate of this magnitude would absorb a dose within minutes that would almost surely prove lethal within weeks (about 6 Sv). Acute incapacitation, however, would not occur until 0-50 minutes had elapsed. Also, each second of exposure (short of lethal doses) would increase the probability of contracting a fatal cancer by 0.1%.
contributors to the dose rate are identified, and their contributions are estimated. It is
clear from these results that for spent fuel that has been cooled for at least five years, the
dominant contribution to the gamma dose rate is provided by the cesium isotopes,
followed by the activation product Co-60 and the rare earth fission product Eu-154. After
15 years of cooling, Cs-137, with a half-life of 30 years, is the major contributor. Other
fission products which are intensely gamma-active have relatively short half-lives and
contribute marginally after 5 years of cooling.
The gamma ray flux generated by an object is a function both of its shape and of
the material from which it is made. For a fixed source density, the dose rate from a
cylinder is maximized at a particular value of the radius. Alternatively, wide, flat sheets
will generate the highest dose rates in the direction perpendicular to the sheet (although
the dose rates in other directions may be much smaller). In designing an interim storage
matrix, there may be some leeway to take source geometry into account, depending on
the details of the concept.
Other properties of the interim form that contribute to its diversion resistance are
its size and weight. Clearly, an object that can only be handled by a crane and
transported on a rail car (such as a shielded transportation cask with a loaded weight
greater than 25 tons) presents a major challenge to thieves.
• High diversion resistance subsequent to the interim phase

Apart from the self-protection afforded by the interim matrices themselves, it is also
necessary to evaluate the self-protection to the point of final disposition. If it is to be
used as a source of nuclear fuel, the interim matrix should not require eventually used as a source of nuclear fuel, the interim matrix should not require
additional processing that may release the plutonium it contains to a highly accessible
stream. This would create a “window” of low diversion resistance that may lead to
unacceptable risks in the future.
Also, in order for the option of direct disposal without use in reactors to remain
viable, the level of self-protection should remain acceptably high at least until the
projected time of emplacement in the repository, and probably for a considerable period
time afterwards (i.e., until the time of repository closure, expected to be 50-100 years
after loading commences.)
• Material behavior during storage

The interim matrix should be in a form that is resistant to corrosion processes and
radiation damage, is not easily dispersible in the event of an accident, and will remain in
good condition for a period of prolonged storage. Careful planning should allow DOE to
avoid a repetition of one of its numerous negative experiences with the storage of highly
radioactive nuclear materials.

Other criteria include:
• Flexibility in reactor recycle options

• Suitability for direct disposal
• State of development and complexity of the required technology

Integral Fast Reactor-based plutonium storage matrices

a) The Integral Fast Reactor

One concept for a matrix for interim storage of separated plutonium has been
proposed by Argonne National Laboratory. The idea is based on fuel cycle technology
that has undergone development in support of the Integral Fast Reactor (IFR) program.
The IFR is a liquid-sodium cooled fast reactor fueled by a metallic alloy of uranium,
plutonium and zirconium, and perhaps minor actinides (MA) as well. Central to the IFR
design is a spent fuel reprocessing technique based on pyrochemical methods, which is
called electrorefining. Because the reference IFR electrorefining process does not
separate plutonium from other actinides and some rare earth fission products with a high
degree of efficiency, it has been characterized by its designers as an inherently
proliferation-resistant closed fuel cycle.21

The IFR has also been promoted as a system that, in addition to recycling its own
spent fuel, could also recover and irradiate actinides from spent LWR fuel. In order for
LWR spent fuel to be converted to a form compatible with the IFR metallic fuel cycle, it
would have to undergo pretreatment to reduce the spent fuel oxide to metal. A number
of processes that could be employed to accomplish this have been studied.22 Each
process leads to a somewhat different partitioning of the radionuclide spent fuel
constituents between the different chemical phases (liquid metal, molten salt, and metal
precipitate). The transuranic (TRU) elements are essentially transported as a group in
all of these processes. After the reduction step, the TRU-containing stream would then
require additional pyrochemical processing in order to extract the product from the solvent
metals. Finally, the product would generally require electrorefining in order to further
separate the TRU elements from uranium and fission products, before it could be
incorporated into IFR fuel.

A simplified flowsheet for the IFR-based interim matrix strategy is shown in Fig. 1.

b) Interim plutonium storage

Argonne has proposed adapting the LWR spent fuel conversion process outlined
above to produce metallic ingots for the interim storage of excess plutonium. This would
be accomplished by alloying the plutonium, at a low concentration (around 1 weight-
percent) with the TRU-containing metal stream obtained from the reduction step.23 In
addition to TRU elements, this product would be contaminated with a mixture of uranium,
transition metal, noble metal and some rare earth fission products. The exact
composition depends on the particular reduction process used. Argonne has claimed that
such an approach would be both "cost-effective" and "an effective deterrent against reversion". To verify such claims, however, one needs more detailed information about the process.

Of the LWR spent fuel pyroreduction processes that have been studied at Argonne to date, two have been identified as being especially promising, the salt transport and the lithium processes. The approximate distribution of spent fuel components for the two processes is shown in Table I.

In the salt transport process, the spent oxide fuel is reduced by calcium metal at a temperature of 800-850°C, and the reduced metals are partitioned among two phases: a molten CaCl₂ salt and a liquid Cu-Mg "donor" alloy. The product, which contains plutonium and other transuranic elements (TRU), dissolves in the metal phase. To separate it from the solvent metals requires a fairly complicated treatment: It is extracted by a MgCl₂ salt and then transferred to a Zn-Mg "acceptor" alloy. The resulting mixture is then heated to 950°C, at which temperature the Zn and Mg are volatilized.

In the lithium process, which is conducted at a temperature in the range 500°C to 750°C, a one-phase molten LiCl system is used; some of the metals are not reduced and dissolve in the carrier salt as chlorides. Other metals, including the TRU product and the uranium are reduced and form precipitates, which can be collected by filtration.

As will become apparent below, neither of these processes is suitable as currently described for preparing a diluent for weapons plutonium storage. For such a purpose, they will either have to be modified, or new processes will have to be developed. For example, in the salt transport process the bulk of the uranium is separated from the plutonium stream and would have to be mixed back in, which is a superfluous step that may negatively affect the diversion resistance of the process.

In the Appendix, the information in Table I is used to provide a rough estimate of the self-protection of the IFR storage ingots obtained from spent fuel reduced via the initial stages of the salt transport process. Of the radionuclides in Table A1, only the cerium and about one-third of the cesium charged will be present with the TRU product in the donor alloy after reduction; ruthenium will precipitate with the uranium. This residual level of contamination is significant. However, it is not clear how much cesium would be retained during the subsequent extraction stages. In the final retorting step at 950°C, a substantial fraction of the remaining cesium (boiling point: 680°C) would probably be volatilized. We will conservatively assume that extraction from the donor alloy will be carried out in some (as yet undetermined) manner, so that the radionuclide content of the product of the reduction step remains fixed.

Approximating the metal ingot as an oxide fuel assembly containing Ce-144, Ru-106 and one-third of the initial Cs-134 and Cs-137, Table A1 indicates that it would still clearly meet the spent fuel standard. The dose rate from such an assembly after five years would be approximately 16 Sv/hr; after 15 years, the dose rate would be about 6.0 Sv/hr. (Without process modifications, however, the remaining cesium, and consequently the gamma dose rate, would be much smaller.)

The IFR ingots would actually be made of metal, rather than the less dense metallic oxide. Furthermore, they would be solid blocks, rather than an array of thin rods with large voids between them. These differences imply that the dose rate suppression due to self-shielding within the ingot would be much more pronounced than in an oxide fuel assembly. Correcting for this effect, it is found in the Appendix that the 5-year dose rates generated by a metallic ingot of the same radius would be closer to 6.5 Sv/hr; after 15 years, it would only be 2.1 Sv/hr, or well below the spent fuel standard. Clearly, the cesium contamination of the TRU-containing metal stream that occurs in the salt transport reduction is an advantage in producing a diversion-resistant interim matrix. However, this phenomenon is anomalous and is not well understood. Cesium is a highly reactive alkali metal with a large activity coefficient in the molten salt phase. Such elements would not generally exhibit such high solubility in the metal phase, unless they were stabilized through the formation of intermetallic compounds, which may be the case in this system. It will be necessary to understand the chemistry in more detail, and in particular whether the cesium contamination is a reproducible and controllable effect, before the potential of the process can be fully assessed.

The lithium process does not separate plutonium from uranium. However, Table I indicates that both cesium and europium are completely separated from the metal precipitate product. The transition metal Ru-106 does appear in the precipitate, but as is apparent in Table A1, its contribution to the self-protection after a few years is not large and declines rapidly. Therefore, the lithium process also does not appear to be suitable for the proposed mission.

Because Argonne has not yet identified a spent fuel pyroreduction process which has the properties that would be most desirable for interim matrix production, additional research would be required to find more useful ones. Furthermore, the processes that have been studied are at a very early stage of development, and startup of a demonstration facility would still be several years away, even if the program were fully funded. These facts suggest that the metallic ingot approach would not be ready for industrial-scale deployment for at least a decade.
Table I  Observed distribution of spent fuel components in reduction processes (from Ref. 22).

<table>
<thead>
<tr>
<th>Component</th>
<th>Salt Transport</th>
<th>Lithium</th>
</tr>
</thead>
<tbody>
<tr>
<td>Uranium</td>
<td>Precipitate</td>
<td>Precipitate</td>
</tr>
<tr>
<td>TRU</td>
<td>Cu-Mg</td>
<td>Precipitate</td>
</tr>
<tr>
<td>Transition metals</td>
<td>Cu-Mg and</td>
<td>Precipitate</td>
</tr>
<tr>
<td></td>
<td>Precipitate</td>
<td></td>
</tr>
<tr>
<td>Noble metals</td>
<td>Cu-Mg and</td>
<td>Precipitate</td>
</tr>
<tr>
<td></td>
<td>Precipitate</td>
<td></td>
</tr>
<tr>
<td>Cs</td>
<td>Salt (2/3) and</td>
<td>Salt</td>
</tr>
<tr>
<td></td>
<td>Cu-Mg (1/3)</td>
<td></td>
</tr>
<tr>
<td>Alkaline earths, L,Te</td>
<td>Salt</td>
<td>Salt</td>
</tr>
<tr>
<td>Europlum</td>
<td>Salt</td>
<td>Salt</td>
</tr>
<tr>
<td>Samarium</td>
<td>Salt and Cu-Mg</td>
<td>Salt</td>
</tr>
<tr>
<td>Other rare earths</td>
<td>Cu-Mg</td>
<td>Salt</td>
</tr>
</tbody>
</table>

**c) Final disposition**

The IFR metallic ingot approach also presents problems for the final disposition of the material. First of all, the chief rationale is to convert the separated plutonium into a form compatible with the IFR fuel cycle. Implementation of this process would only be appropriate if there were a high probability that the IFR will be fully developed and deployed. However, this is very unlikely at the present time. In fact, the IFR research program has recently lost a fierce battle in Congress to retain a fraction of the funding level it had enjoyed in the past. In August 1994, Congress voted to terminate the IFR program. The FY 1995 budget contains $38.8 million for the shutdown of the experimental fast reactor EBR-2 at Argonne West in Idaho, where metallic fuel pin irradiation tests were conducted, and the phasing out of the "actinide recycle" program, in which the prospects for using the IFR to fission plutonium and other actinides recovered from commercial LWR spent fuel. Other applications of the technology, such as the continued development of pyroprocessing for the management of Hanford N-Reactor and other metallic fuels, may still survive.

There are a number of reasons, from a general non-proliferation standpoint, why involvement of the IFR fuel cycle in the plutonium disposition mission would be unwise.

First of all, the IFR is a fast-neutron spectrum reactor well-suited for the efficient breeding of plutonium. According to one of its promoters, Charles Till, the IFR is "the best of all possible breeders in the sense of the lowest doubling time and the highest breeding". Breeding ratios in the range of 1.5 to 1.65 were expected. At such high breeding ratios, the IFR could generate extra plutonium at a rate which is more than twice the production rate in LWRs. The continued funding of this project by US DOE has been a concern to non-proliferation advocates who have long opposed the development and deployment of breeder cycles, which not only require reprocessing and the circulation of large inventories of separated plutonium, but which have the potential to swell the already huge world plutonium surplus. The conversion of excess plutonium to a form tailored specifically to the IFR fuel cycle would appear internationally as a US endorsement of fast reactor technology. This would impair US efforts to discourage breeder reactor development in Russia, as well as in France, Japan, and India.

In the past couple of years, Argonne has downplayed the breeding capability of the IFR, choosing instead to emphasize its potential as a plutonium "burner", with a "breeding" (plutonium conversion) ratio of less than one. The implication has been that shifting operation from one mode to the other could only be accomplished with a complete redesign of the core. This argument is disingenuous, however, since the IFR developers themselves have designed the reactor so that cores intended for plutonium burning could be fully interchangeable with cores for breeding, without requiring any adjustments in the operating parameters. In their own words, "...all assembly dimensions, control rods and power levels are maintained" in converting the IFR from burner to breeder mode.

IFR advocates contest the view that dissemination of the technology would pose proliferation risks, by claiming that spent fuel pyroprocessing is a more diversion-resistant technique than aqueous-based PUREX reprocessing. This is a complicated issue that has been discussed at length elsewhere and would be somewhat of a digression if reproduced here. There is one aspect of the discussion, though, which is relevant to the present work and which has not been fully addressed in the literature on the subject.

The plutonium-containing product of the IFR fuel recycle process is contaminated with other actinides and some fission products. As a result, the entire process, including the refabrication of fresh fuel, must be contained within a hot cell and remotely operated. One report states that the product is "about ten thousand times more radioactive than PUREX products,...because of the residual fission product lanthanides." This is the basis for the claims of diversion resistance.

More detailed analysis of the process flow sheet, however, shows that the actual self-protection of the IFR product stream, and in particular the fuel assemblies fabricated from recycled material, is not as significant as the above description may suggest. The above quote refers only to the total activity of the IFR product, whereas only the penetrating gamma (and perhaps neutron) activity is relevant to the dose rate; furthermore, there is no reference to the dependence of the activity on time.

In the Appendix, the dose rate generated by a refabricated IFR fuel assembly due to the contamination by lanthanide fission products and actinides is estimated, assuming a five-year spent fuel cooling and fuel recycle period. The radiological hazard of the
product is dominated by the decay of Pr-144, the short-lived daughter of Ce-144, which with a half-life of 0.265 days does not provide protection for more than a couple of years. After five years, the dose rate from such an assembly, fabricated with plutonium obtained from the intermediate storage ingots described above, would be around 0.18 Sv/hr, which is far below the IAEA criterion and which would provide more than adequate protection. These values are consistent with other analyses of the diversion resistance of pyroprocessing technology and the value of Ce-144 as a fuel “spikant” that were performed over a decade ago.39,40

Because of the low plutonium concentration and an unacceptably high fissile product burden, metallic interim storage ingots would have to undergo electrorefining and secondary processing into fuel assemblies prior to use in the IFR. This requirement is perhaps the most unattractive aspect of the IFR-based interim matrix approach and would appear to be contrary to the original goals of the undertaking unless the product stream met the spent fuel standard throughout the entire process. However, this would not be the case for the plutonium-bearing glass fuels. For instance, if the ingots were produced using 5-year cooled LWR spent fuel, and then stored for a decade before electrorefining, the fresh IFR fuel incorporating the plutonium product so obtained would generate lower dose rates than an ordinary MOX fuel product and would therefore require security and safeguards meeting the stored assembly standard. Under these circumstances, electrorefining of the material would clearly be unacceptable.

Also, if it were later decided to dispose of the ingots rather than to re-vitrify them in IFRs, it is doubtful that the uranium-plutonium-zirconium metal alloy would be suitable for disposal. The alloy would be prone to corrosion in hot, oxidizing environments, similar to that of the prospective US repository site at Yucca Mountain. The ingots would probably have to undergo further processing, such as conversion to oxide and vitrification, to prepare them for disposal.

Furthermore, as discussed above, by the time that a geologic repository would be ready to accept the ingots (not before 2015), their self-protection would probably have fallen to a level below the spent fuel standard, assuming that they were produced between 2000 and 2010 with 5-year cooled LWR spent fuel used as diluent. Because of this, additional irradiation in reactors might be necessary to restore the radiation barrier.

For these reasons, a weapons plutonium dilution process based on IFR technology does not exhibit enough flexibility to be a useful part of an interim matrix strategy.

**AIROX-based interim plutonium storage matrices**

a) Spiked oxide fuel and the AIROX process

The numerous disadvantages of the IFR-based interim matrix approach have led us to examine whether conceptually similar alternatives with fewer problems can be found. An interim matrix that would be preferable on security and economic grounds to the metallic ingot should have qualities enabling it to be used directly as fuel in thermal reactors (as opposed to fast reactors) or emplaced in a geologic repository, without needing significant additional processing in either case. Also, if further processing were necessary for either option, the plutonium-bearing stream should retain substantial self-protection at all points along the process line, even if the matrices were to remain in interim storage for several decades.

For example, it is conceivable that the plutonium-enriched vitrified HLW canisters could themselves be used as fuel in reactors, which would be an ideal realization of this approach. In fact, the irradiation of plutonium-bearing glass fuels was studied on a small scale in the 1950s.42 It was found that the fuel rapidly became brittle and was therefore unsuitable. It is also known that glass corrosion, although generally low at lower temperature, is accelerated at temperatures above 100°C. Glass therefore would not be a very durable fuel. Furthermore, melting points of silicate glasses are generally considerably lower than those of the uranium dioxide-based ceramics which are most commonly employed as nuclear fuels, so that safety margins would be unacceptably small.

One could also reexamine past proposals for increasing the diversion resistance of plutonium fuel cycles by adding highly gamma-active “spikants” to the fresh fuel rods. In general, the radionuclide additives would have to be produced in reactors, which would make little sense in the context being considered here. However, the US DOE has stockpiles of radioactive material which could in principle be employed for this purpose: capsules containing Cs-137, which was at one time separated from HLW at Hanford, and HLW calcine stored at the Idaho National Engineering Laboratory (INEL).40 Although the available quantity of separated Cs-137 is too small to provide a radiation barrier comparable to spent fuel for the entire excess plutonium inventory, smaller dose rates similar to those of vitrified waste glass canisters might be achievable. On the other hand, the calcine, a very fine nitrate-based powder, would probably be too chemically complicated and hazardous to use.41

In Russia, apparently, similar production of radioisotopes including Cs-137 has been carried out, but it is unclear how large the inventories are and if there are competing uses for the material.42 In Russia, there has been some development work on a low-decontamination, pyrochemical recycling process for spent oxide fuel.43,44 This process involves the dissolution of spent oxide fuel in a molten fluoride-chloride salt, and the electrotransport of a uranium-plutonium-minor actinide oxide product to graphite cathodes. From this description, it is apparent that there is a close resemblance to the Salt Cycle Process that was developed at the Hanford Laboratories (precursor to Battelle Pacific Northwest Laboratory) over thirty years ago.45 It is essentially the oxide fuel analogue of the IFR metallic fuel electrorefining process. If the cathode product contained a sufficient amount of fission product contamination, the Russian technique could be used to produce a radioactive diluent for blending with weapons plutonium. However, it has been reported that in the Salt Cycle Process only about one-fourth of the rare earths follow the plutonium.46 Without further information, it is reasonable to assume that cesium will remain completely dissolved in the molten salt, as in IFR electrorefining. Therefore, it is likely that the salt cycle product would not provide a significantly greater degree of self-protection than the IFR product.

A different approach to production of an interim storage matrix, which could
potentially be much more diversion-resistant, is based on the AIFROX (Atoms
International Reduction Oxidation) process, a dry, low decontamination spent oxide fuel reprocessing technique. The goal of AIFROX is to treat spent fuel rods in a manner so
that they can be recycled in reactors without separating plutonium either from uranium or from a major fraction of the fission products.

In this process, a spent oxide fuel assembly is punctured and then subjected to a series of oxidation-reduction reactions at temperatures of 400°C–600°C. The initial oxidation step causes the fuel matrix to expand. As a result of this expansion, the fuel becomes separated from the cladding and is comminuted (pulverized into granules), with an average size of greater than 10 microns. The average particle size can be reduced with subsequent repetitions of the cycle; it was found that three cycles were sufficient to produce a suitably fine powder for fuel reprocessing. During the oxidation-reduction step, all of the volatile fission products, including krypton and iodine isotopes, tritium, and carbon-14, are evolved from the fuel. The resulting powder can then be sintered into pellets and recast into fuel rods for use directly in CANDU reactors (in which case the process is referred to as OREOX rather than AIFROX). If recycle in LWRs is desired, the powder must be re-entrained by ball-milling the granules and blending with additional fissile material, either PuO2 or 17%-

enriched UO2. During the sintering step, which takes place at 1700°C, significant quantities of semi-volatile fission products would be released. Experimental trials found releases ranging from 80% to 95% of the Cs-137 inventory, and from 20% to 50% of the Ru-106 inventory.

The products of this process are essentially “dirty” MOX fuel rods that cannot be contact-handled. Preliminary paper studies have shown that the presence of the residual
fission products in the fuel should not have a detrimental effect on its neutronics performance in PWRs. Some changes would occur in the reactor operating parameters, such as the control rod worth, reactivity coefficients, and neutron energy spectrum, which would require adjustments. However, calculations indicate that with respect to all the criteria examined, the “dirty” MOX fuel would perform as well or better than “clean” MOX fuel in thermal reactors, for similar deployment fractions in the core. Experimental validation of these calculations would be essential for firmly establishing the viability of AIFROX fuel.

As a consequence of the extremely high radiation levels generated by the AIFROX process streams, all stages of the cycle must be carried out remotely. Although not a problem for the relatively simple AIFROX processing steps, this aspect may present a serious technical challenge to the industrial-scale development of the fuel refabrication stages of the process. Conventional MOX fuel fabrication technology relies primarily on glovebox operations, which are possible when working with fuel materials that emit relatively weak penetrating radiation fields, such as LWR plutonium that has not aged more than a few years after reprocessing. On the other hand, in state-of-the-art MOX
fabrication plants (such as the soon-to-be-commissioned MELOX facility at Marcoule), which have been designed to handle high-burnup or aged plutonium, much of the
equipment is automated or remotely controlled. Novel processes have been developed to perform tasks that were previously considered difficult to handle remotely, such as rod decontamination and rod size inspection.

However, the radiation fields that would be encountered in an AIFROX fuel refabrication line would be of considerably greater intensity than those in a high-burnup plutonium MOX facility. Robotics equipment that is capable of the intricate maneuvers required for fuel pellet fabrication contains delicate electronic circuitry, which is not able to function reliably in such a high radiation environment. This problem is the weakest link in the AIFROX cycle, and much research and development in this area would be necessary.

In spite of these difficulties, the OREOX process has recently been identified by
an international study as the most promising technique for the recycling of LWR spent fuel directly in CANDU reactors. This is of interest to the nuclear programs of both Canada and South Korea, who have jointly undertaken a project to further develop it, with the participation of the US State Department. Preliminary safeguards analyses of the cycle have concluded that the diversion risks it may pose, although not zero, are “relatively minimal.”

b) Interim plutonium storage

A modified version of the AIFROX process could be employed to combine spent fuel and weapons plutonium into an interim storage product that would clearly meet the spent fuel standard. The interim matrices, which would resemble spent oxide fuel, could then be safely stored using well-demonstrated technology, such as dry casks, for a period of several decades or longer.

The appropriate plutonium loading in this matrix would be determined by the probable means of final disposition of the material. For utilization in LWRs, a total fissile enrichment of around 5.5% is necessary, which is higher than the minimum enrichment for ordinary MOX fuel because of the presence of fission product absorbers; this requires an addition of about 4% weapons-grade plutonium. In contrast, in CANDU reactors, AIFROX-processed fuel can be utilized without fissile re-enrichment; thus beginning-of-cycle core reactivity does not impose restrictions on the minimum weapons-grade plutonium loading. The additional fissile enrichment would enable the burnup of the fuel to be extended.

An AIFROX processing plant could be used to convert separated plutonium to a SFS storage product at a far greater rate than could be achieved in a single reactor of

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1 Automation of the fuel fabrication process has not yet been perfected, however, as is evidenced by a recent mishap at an automated continuous-production sintering furnace in Japan that caused a six-month delay in the startup of the Monju fast breeder reactor. (More Fuel Fabrication Problems Push Monju Start to Spring 1994, Nuclear Fuel, July 1993, p.14.)
comparable capital cost.* The conversion rate would depend on the facility throughput, desired fissile concentration and other factors. Not including construction time (probably 5-10 years), a medium-sized plant (150 tHM/yr throughput), could incorporate 50 tonnes of separated plutonium into 5.5% fissile-enriched AIROX fuel in about ten years. In contrast, around six Palo Verde-type PWRs fueled with full cores of MOX would be required to attain an equivalent conversion rate.

The isotopic content of the plutonium in the fuel rods fabricated from this method depends on the weapons plutonium enrichment and thus on the planned mode of utilization of the fuel. For use in LWRs, the ratio of the residual plutonium content of the spent fuel feedstock to the added weapons plutonium would be about 1:4. Assuming that the isotopic composition of the residual reactor-grade plutonium is 60% Pu-239 and 24% Pu-240, then the resultant composition would be 47% Pu-239 and 8% Pu-240, which is not a significant shift from weapons-grade material. It is interesting to note, however, that this composition is close to that of the spent fuel from a fast reactor fueled with weapons-grade plutonium, which would only have a Pu-240 content of around 8%.

For use in CANDUs, the magnitude of the weapons plutonium loading is flexible, and can be adjusted to produce fuel with a plutonium isotopic content quite close to reactor-grade, if so desired. For instance, if the loading were only 0.5%, then the ratio of reactor-grade to weapons-grade would be 2:1 and the overall isotopic composition would be 71% Pu-239 and 18% Pu-240. Of course, a much larger throughput than 150 tHM/yr would be required to process the plutonium at a reasonable rate at this loading. The point here is that the AIROX process can in principle be used to “degrade” the isotopic content of the weapons plutonium without burning it in reactors, should it prove worthwhile to do so.

The first question to be addressed is whether the technology is sufficiently well-developed so that it could be implemented in a timely manner. The initial AIROX processing steps are fairly simple and have been demonstrated on a laboratory scale in both hot and cold tests. However, the fuel refabrication step may pose difficulties, in addition to those associated with conducting remote operations in highly radioactive environments, which were discussed previously.

In particular, the sintering step will have an impact on the radiation barrier generated by the product. During sintering, which is conducted at 1700°C (for uranium-based fuels), a significant fraction of the cesium and ruthenium contained in the fuel is volatilized (the boiling points of metallic cesium and cesium pertechnetate are 676°C and 650°C, respectively; ruthenium can form the highly volatile tetraoxide). As was seen in

*One reference estimates the capital cost of an AIROX plant with a throughput of 500 tHM/yr to be $850 million. (D. Majumder, et al. 1992.) As is usually the case, this estimate is probably highly optimistic. However, there is no reason to expect the facility (excluding the remote MOX fabrication line) would be more costly than a conceptually similar operation such as a vitrification plant. Current cost estimates for a new vitrification plant with a throughput of 500 tonnes of glass per year are around $1 billion. (E. Hinou, "DOE Multi-Commercial Proposal to Use French Vitrification Process at Hanford," Nuclear Fuel, March 14, 1984.) Also, in order to reduce costs and delays, it may also be possible to install an AIROX line in the FMEF at Hanford, which is a heavily shielded, canyon-like building containing 17 hot cells. The section on metallic ingots, the loss of cesium has a negative impact on the diversion resistance of the final product, with respect to both the magnitude and persistence of the gamma radiation field. Release of 80%–95% of the Ce(134+137) and 20%–50% of the Ru-106 from the fuel would reduce the dose rate from an AIROX fuel assembly recycled from five-year-old spent LWR fuel (33 GWd/ft³ burnup) from about 60 Sv/hr to the range of 9.2 Sv/hr - 16 Sv/hr (see Appendix). These are still very high dose rates, and are greater than those typical of five-year-old vitrified waste canisters. However, the AIROX radiation barrier would weaken more rapidly with time, so that after fifteen years of storage the AIROX fuel would deliver a dose rate in the range of 3.4 Sv/hr - 6 Sv/hr, which would be below that of the vitrified waste. Thus the high-temperature sintered AIROX fuel would fail to meet the spent fuel standard if it remained in storage for more than about fifteen years. Note that this problem would also be encountered in the production of Ce-137- spiked MOX fuel rods, and would be more severe, as there would be no contributions to the dose rate from non-volatile spent fuel constituents such as Eu-154.

In order to maintain a greater level of self-protection for the interim matrix, a number of different approaches can be considered. One option would be to defer the actual fuel refabrication step until prior to reactor use. Another would be to replace the fuel fabrication process based on pellet sintering at 1700°C with one that could be performed at a lower temperature, at which the cesium volatilization would be suppressed.

Deferral of the fuel fabrication step would simplify the manufacture of AIROX-based interim matrices, enable them to unambiguously meet the spent fuel standard, and probably render the procedure significantly more viable in the near term. It would also allow time for research and development of needed technologies such as radiation-hardened robotics components. On the other hand, it would for some scenarios increase the total number of processing steps required to prepare the material for reactor use.

The strategy requiring the least processing would be preparation of the AIROX fuel feedstock granules as outlined above, and then storage of the mixture in cans. This is not a satisfactory approach from a safety perspective, however. Many of the undesirable features of storage of plutonium dioxide (or MOX) powder, such as radiolysis effects and risk of dispersal, would be present in this case as well, and the additional presence of fissile products would tend to worsen the consequences of an accident.

A safer option would be to store the fuel powder in stainless steel cans and then densify it at 1200°C and a pressure of nearly 1000 MPa with a pneumatic compaction press to produce large ceramic slugs. Slugs produced in this fashion from a mixture of UO2 and PuO2 powders were found to have fine scale homogeneity and a density close to 100% of theoretical.
Alternatively, a process similar to that chosen for the demonstration commercial-scale SYNROC (a ceramic nuclear waste form) production plant in Australia could be employed to produce a similarly dense solid product. This involves cold-pressing of the fuel powder at 40 MPa, followed by bellows hot-pressing at about 1150°C and 14.21 MPa. Since SYNROC is a ceramic based on titanium oxide, which has a melting point lower than uranium oxide, the temperatures or overpressures may have to be greater in the AIROX case in order to achieve the desired material properties using this process. Both of these processes involve simpler maneuvers than fuel fabrication and would be compatible with automation and remote control.

The relatively low temperature (comparable to the operating temperature of a HLW glass melter), the strongly reducing conditions, and the lower surface-to-volume ratios encountered in these processes all tend to inhibit the volatilization of cesium and ruthenium relative to high-temperature pellet sintering. For example, it was observed during the hot-pressing of simulated HLW-containing SYNROC precursor powders at 1150°C and 14.21 MPa that only 0.14% of the cesium, and a negligible fraction of ruthenium, was volatilized. Also, the fabrication of SYNROC at around 1200°C using conventional pellet sintering only volatilized about 5% of the cesium inventory. As in the case of IFR-based metallic ingots, the dose rate from a solid ceramic slug would generally be lower than that from an oxide fuel assembly of similar dimensions, because of the greater average density of the slug. However, the dose rate suppression would not be as pronounced as for the ingots. Assuming also that the loss of cesium and ruthenium through volatilization can indeed be reduced to, say, 5%, the ceramic slugs would generate dose rates around 30 Sv/h at 5 years, and 12 Sv/h at 15 years.

If it were later determined that the material should be converted to reactor fuel, it would not be difficult to crush the blocks and use them as feed for the fabrication process, provided equipment for that purpose were installed initially in the hot cell facility. However, the additional handling and processing resulting from the introduction of the intermediate stage would probably lead to an increase in overall costs and occupational risks.

A few possibilities for fuel fabrication processes that do not require temperatures as high as 1700°C merit further examination. The sintering temperature cannot simply be reduced without having a deleterious effect on fuel properties essential for good performance, such as density, degree of solid solution, and grain size. However, by sintering under high pressure (hot-pressing), good material properties can be achieved at a lower temperature. This procedure requires a temperature of about half the melting point of the material (mostly uranium dioxide) or about 1400°C. Although lower than the (ambient pressure) sintering temperature, the hot-pressing temperature is still high enough to cause appreciable volatilization of cesium and ruthenium. It is also not clear if this process, which is expensive and requires frequent equipment replacement at such high temperatures, would be feasible for industrial-scale, remotely-operated fuel fabrication.

Sol-gel processing methods for fuel fabrication are carried out at low temperatures. However, the processes generate large amounts of residual wastes, which would be highly radioactive in the case under consideration. Such an approach is probably unsuitable.

Another alternative, which has been demonstrated on a small scale, is the "Vi-Pac" process, which uses high-energy vibration to induce the dense compaction of fuel granules. High process temperatures are not required. The feed material for this technique must consist of particles of fuel which have a nearly theoretical density and a high degree of PuO₂–UO₂ solid solution. Therefore, to incorporate this process into the AIROX cycle the recycled and blended fuel powder would first have to be densified and then crushed. Thus the Vi-Pac process is a natural choice for fabricating fuel from the ceramic slugs described above. Fuel assemblies refabricated with Vi-Pac would generate a radiation barrier nearly as intense as that of the original spent fuel. Because of its apparent simplicity, this method was originally envisioned as ideal for remote refabrication applications. However, in practice it has proven to yield irregular results, so that frequent sampling is necessary to guarantee that the product has the desired characteristics. Substantial development would be needed to refine this process for remotely-controlled production of acceptable fuel on an industrial scale. Nevertheless, the Vi-Pac process shows promise for integration into a maximally self-protecting AIROX cycle and warrants further investigation. A flowsheet for this low-decontamination strategy is presented in Fig. 2.

c) Final disposition

It is clear from the above discussion that strategies do exist for producing an AIROX-based interim form that would be compatible with subsequent reuse in reactors or direct geologic disposal. However, demonstrating the feasibility of either option would require research and development. In Russia, redirection of their research program along these lines may alleviate some of the international concerns regarding their current program while preserving employment opportunities within the nuclear complex.

If a lower temperature fuel fabrication method were employed, the greater retention of cesium, ruthenium and other semi-volatiles (tellurium, cadmium and indium) would increase the fission product burden of the AIROX fuel. This would have a detrimental effect on the neutron quality of the fuel. In fact, recent assessments of using AIROX-recycled fuel have been concerned with decreasing, rather than increasing, the level of contamination. Assuming zero release of semi-volatiles, the beginning-of-cycle total neutron absorption cross-section of the residual fission products would increase by about 15% relative to the high-temperature sintered fuel, mostly due to the increased concentration of stable Cs-133. Further analysis would be necessary to evaluate whether this is acceptable.

One of the advantages cited above of the AIROX over the IFR approach is that it is potentially compatible with reuse in thermal reactors. However, the Russian nuclear industry has not expressed much interest in thermal recycling. Furthermore, the apparent determination of Minatom to retain a large plutonium inventory for eventual use in fast reactors has not yet wavered under U.S. pressure, and may prove to be an obstacle to future negotiations. In view of the probable delays in advancing their program, which will
would contain 1.6% U-233. The number of recycles possible will be determined by the total fission product buildup.

It must be kept in mind, however, that AIROX fuel assemblies fabricated for fast reactors would be more attractive targets for diversion than ones fabricated for thermal reactors, because of their greater fissile concentration. Also, because it is generally acknowledged that there is no "technical fix" for the proliferation risks posed by breeder technology, the larger implications of any compromise encouraging the continued development of fast reactors in Russia must be carefully assessed.

If it were eventually decided not to use the plutonium in reactors, the interim storage matrices produced by one of the methods described in the previous section could be sent directly to geologic disposal. With a durable overpack similar to the Zircaloy cladding of LWR fuel, it is likely that the performance of the waste blocks in a repository environment would be as good or better than the original spent fuel assemblies. The waste blocks may have a greater density, and would be relatively free of cracks and other defects in spent fuel caused by reactor irradiation.

One potential problem with the direct disposal of blocks that had been re-enriched for possible LWR use is that the fissile concentration, 5.5%, would be high compared to spent fuels, which may increase the risk of a criticality event occurring in the repository. This would be less of an issue if the blocks had been intended for re-use in CANDUs, since they would have much smaller enrichments in that case.

Summary

In Table II, characteristics of the interim storage forms discussed above are summarized.

Some environment, safety and health (ES&H) issues

At first glance, the objective of minimizing the diversion risks posed by weapons plutonium by dramatically increasing the hazard of handling it appears to conflict with the goal of keeping near-term environmental, safety and health (ES&H) risks to a minimum. However, with careful attention paid to potential environmental impacts and danger to personnel, there need not be a major tradeoff in risks. These considerations should also figure prominently in the choice of conversion process.

Generally speaking, SFS conversion processes that do not involve the use of nuclear reactors, such as vitrification or low-decontamination AIROX, will have much smaller ES&H impacts than the ones that do, for equivalent plutonium throughputs.

To see this, first consider a comparison of the risks to the public resulting from the operation of two full-core MOX reactors to those from the MOX fabrication facility that supports them, which would have a throughput of 60 tHM/yr at an enrichment of 3% weapons-grade plutonium. Both the probability of a catastrophic release of radioactive material, and the greatest potential consequence of such an event, would be far greater.
### Table II: Summary of dose rates of plutonium storage matrices

<table>
<thead>
<tr>
<th>Matrix</th>
<th>Average density (g/cm³)</th>
<th>Principal radionuclides*</th>
<th>Dose rate at 1 meter (Sv/hr)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>5-year</td>
</tr>
<tr>
<td>IFR: storage ingot</td>
<td>15</td>
<td>Ru, Cs(0.33), Ce</td>
<td>6.5</td>
</tr>
<tr>
<td>IFR: fuel assembly</td>
<td>3.7</td>
<td>Ce(1.6)</td>
<td>0.18</td>
</tr>
<tr>
<td>AIROX: storage slug</td>
<td>10²</td>
<td>Ru(0.95), Ce(0.95), Cu</td>
<td>30</td>
</tr>
<tr>
<td>AIROX: fuel assembly</td>
<td>3.2</td>
<td>Ru(0.5-0.8), Cs(0.2-0.05), Ce, Eu</td>
<td>9.2-16</td>
</tr>
<tr>
<td>(pellet sintering)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>AIROX: fuel assembly</td>
<td>3.2²</td>
<td>Ru (0.95), Cs (0.95), Ce, Eu</td>
<td>48</td>
</tr>
<tr>
<td>(VI-Pac process)</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

(a) Figures in parentheses indicate the ratio of radionuclide concentrations to that of the spent fuel assembly described in the Appendix.
(b) Assumes 90% of theoretical UO₂ density is achieved.
(c) Assumes process is capable of achieving similar densities to pellet sintering.

The AIROX-based interim conversion flowsheet is roughly comparable to MOX fabrication with respect to the quantities of plutonium in-process. In addition, the AIROX process involves fission products. However, the fission product inventories at points along the AIROX production line will generally be far smaller than the amount in the reactor core after a few weeks of operation.

The risks of breach of containment during AIROX processing would also be similar to those encountered in MOX fabrication, since a similar range of process temperatures would be involved in both.

For these reasons, it is possible to conclude in a preliminary way that the AIROX process is capable of incorporating plutonium into a product meeting the spent fuel standard in a much safer manner than any of the reactor options. It may even be safer than vitrification at the DWPF, since it would avoid the risks due to contamination of the HLW feedstream with potentially unstable organic substances. There are risks associated with AIROX: for instance, there is the possibility of an explosion should contact take place between the oxygen and hydrogen used in the oxidation-reduction cycles. While serious in itself, an event of this type need not lead to the dispersal of plutonium into the environment, since plutonium is not introduced into the process until the oxidation-reduction stages are complete. Therefore, it should be possible to design the facility so that the risk of plutonium dispersal is minimized: for instance, the cell where AIROX processing would place should be well-separated by a robust barrier from the cell where blending and compaction would take place.

Routine emissions generated by the conversion processes should also be taken into account. The AIROX process causes essentially complete evolution of volatile fission products H-3, Kr-85, I-129 and C-14 from the spent fuel matrix, as is the case in aqueous reprocessing or pyroprocessing.²⁹ Emissions of these gases during large-scale reprocessing operations disperse globally and contribute to the committed radiation dose burden of the world population. However, the overall impact of a limited AIROX campaign for disarmament purposes would not be substantial, especially if currently available systems that control emissions of C-14 and iodine are employed.³⁰

To provide the material for encapsulating 50 tonnes of separated plutonium at a total fissile enrichment of 5.5% would require the AIROX processing of about 1250 tonnes of spent PWR fuel. This amount is around two times the annual throughput of a large commercial reprocessing plant. Assuming standard characteristics for the spent fuel (33 GWd/t burnup, 5 years' cooling period), the Kr-85 inventory contained in 1250 tonnes is about 3×10¹⁰ TBq. Standard dispersion models predict that this would result in a 500-year population dose commitment of around 120 person-Sv.³¹ This is less than 5% of the 500-year dose commitment delivered annually to the world population due to (routine) civilian nuclear power operations.³²

The applicable EPA standard for Kr-85 emissions from the entire fuel cycle is 1900 TBq per GWe-yr. This rule would effectively prohibit the operation of reprocessing facilities on a large scale in the US unless expensive krypton retention equipment was installed. However, since there is not likely to be commercial reprocessing in the US in the foreseeable future, the US fuel cycle, with a generating capacity of 99 GWe in 1993, has a Kr-85 credit of nearly 2×10⁷ TBq/yr.³³ The AIROX processing of 1250 tonnes of
spent fuel over a ten-year period would only release about 1/6 of the Kr-85 allotment per year.

Occupational radiation exposure occurring during AIROX processing is another important consideration. One might think that AIROX operations would lead to higher individual exposure levels than ordinary MOX fuel fabrication, because of the higher radioactivity of the process streams. However, this is not necessarily the case. Since conventional MOX fabrication takes place in gloveboxes, there are more opportunities and pathways for both internal and external exposure of employees to radioactive materials than there would be in a well-designed hot-cell facility where all operations were remotely conducted. This trend can be seen by comparing recent French individual (external) occupational exposure data for employees reprocessing plants to those in MOX fabrication facilities. Individual doses in 1991 at La Hague averaged 0.5 mSv per year, more than ten times less than the 1988 average at the MOX facility at Cedarsache, which appears to be a typical value.47 Although the meaning of such comparisons is unclear without more details, they are quite suggestive in this case.

Additional ESSH issues would be raised in considering the relative risks of the utilization of AIROX-processed fuel and MOX fuel in reactors. Perhaps the greatest difference is the impact on the maximal consequences of an accident due to the increased inventories of some fission products in the core. The levels of I-131 and Xe-133 would be similar, whereas Sr-90 releases could increase by almost 1/3 (assuming that only 1/3 of the core was loaded with AIROX fuel). The Cs-137 and Cs-134 inventories depend on the fraction evolved during fuel fabrication. These cesium isotopes dominate the population dose commitments in most accident scenarios. If nearly the entire cesium content were retained in the AIROX fuel, a core melt with a breach of containment could increase the total cancer death toll by thousands. This will have to be taken into account when determining the optimal amount of cesium retention in the AIROX fuel. It should be noted that a similar issue arises when the extended burnup of fuels is considered.

Conclusions

The encapsulation of excess separated plutonium into interim storage matrices that are highly diversion-resistant, yet in principle usable in reactors, should be investigated. This strategy may be appropriate for the extended storage of Russian plutonium, should it appear likely that there will be delays in implementing the final disposition of the material.

Two types of storage matrices were considered in this report. One of them, based on InteReal Fast Reactor metallic fuel cycle technology, exhibits a number of significant disadvantages. It is impractical and does not provide a very high level of diversion resistance overall.

An approach based on AIROX oxide fuel recycling technology shows more promise. With process adjustments, it may also be possible to employ the method as a means of converting separated plutonium to a waste form that meets the spent fuel standard and is suitable for final disposition. Such a technique would provide another reactor-free option in addition to vitrification, the prospects of which are currently uncertain, despite its many attractive features.

Appendix: Comparison of the Gamma-Ray Dose Rates from Spent PWR Fuel, IFR Storage Ingots, Refabricated IFR Fuel and Refabricated AIROX Fuel

In order to estimate the dose rates from interim storage matrices fabricated from spent LWR fuel recycled via the IFR pyroreduction or AIROX processes, it is necessary first to identify the radionuclides in spent fuel which are primarily responsible for the emissions of penetrating gamma radiation, and their relative contributions. Then, the type and concentrations of the various radionuclides that would be present in the final product must be calculated. Because it is likely that for radiation protection, spent fuel employed for this purpose will be cooled for several years prior to processing, the computation assumes a five-year cooling period. Since interim forms should be expected to retain high levels of self-protection for a minimum of ten years after fabrication (and perhaps much longer), the 15-year dose rates are also provided.

Spent PWR fuel

Five years after it is discharged from the core of a pressurized-water reactor, a spent fuel assembly of 33 GWd/MTM burnup will generate a gamma-ray dose rate at one meter from the fuel edge, measured along a perpendicular bisector, of approximately 60 Sieverts (Sv) per hour, according to the results of recent computer simulations.48 Fifteen years after removal, this will have decreased by about 60%, to about 24 Sv/hr. After fifteen years has passed, the dose rate will be determined primarily by the decay of Cs-137, and it will thereafter decrease exponentially, with a half-life of 30 years.

The gamma-ray emitting radionuclides which dominate the generation of the radiation field, and their approximate relative contributions to the dose rate, are listed in Table A1.49
Table AI Contributions of radionuclides to gamma dose rates at one meter from the center of a LWR spent fuel assembly, for 5- and 15-year aging times.

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half-life</th>
<th>5-year cooled activity density (TBq/cm)</th>
<th>Dose rate at 1 meter (Sv/hr)(a)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>5-year</td>
</tr>
<tr>
<td>Co-60</td>
<td>5.27 y</td>
<td>0.19</td>
<td>9.7</td>
</tr>
<tr>
<td>Ru-106</td>
<td>368 d</td>
<td>0.89</td>
<td>1.3</td>
</tr>
<tr>
<td>Sb-125</td>
<td>2.73 y</td>
<td>0.19</td>
<td>0.5</td>
</tr>
<tr>
<td>Ce-134</td>
<td>2.05 y</td>
<td>1.34</td>
<td>21</td>
</tr>
<tr>
<td>Ce-137</td>
<td>30.0 y</td>
<td>4.41</td>
<td>21</td>
</tr>
<tr>
<td>Ce-144</td>
<td>285 d</td>
<td>0.61</td>
<td>0.3</td>
</tr>
<tr>
<td>Eu-154</td>
<td>8.6 y</td>
<td>0.33</td>
<td>5.6</td>
</tr>
<tr>
<td>Am-241(b)</td>
<td>430 y</td>
<td>0.05</td>
<td>0.0001</td>
</tr>
<tr>
<td>Cm-243</td>
<td>28.5 y</td>
<td>0.0001</td>
<td>&lt;0.0001</td>
</tr>
<tr>
<td>Total</td>
<td></td>
<td></td>
<td>59</td>
</tr>
</tbody>
</table>

a) Distance measured from edge of fuel assembly. Results approximated using analytical method. Totals are in good agreement with those obtained by numerical simulation (Ref. 19).
b) Results scaled from the dose rate from a fresh MOX fuel assembly fabricated with reactor-grade Pu, as given in the NAS study (Ref. 1).

The values in Table AI were obtained by approximating the rectangular spent fuel assembly as a cylinder and using an analytic method. The spent fuel characteristics were obtained from Ref. 19. The fuel assembly was modeled in Ref. 19 as having uniform density equal to the average density, or the total mass of heavy metal divided by the fuel assembly volume. Since approximately two-thirds of the fuel assembly cross-section consists of space between fuel rods, the average fuel density is about one-third of that of uranium dioxide. This simplification is also followed here. The absolute dose rate values depend sensitively on the fuel assembly geometry. An effective cylindrical radius of 12 cm was selected by equating the cross-sectional area of the cylinder with that of the rectangular fuel assembly. This procedure gives total dose rates that are in good agreement with the numerical results in Ref. 19. Nevertheless, these values should be regarded as highly approximate. It has been found that dose rate calculations can differ by as much as a factor of 2 from measured values.

exercise is not intended to present accurate values of absolute dose rates, but merely to provide a sense of the relative importance of the various radionuclides to the overall dose rate.

IFR storage ingots

The dose rate generated by IFR-based metallic storage ingots is a function of the concentrations of radionuclides that are retained with the product of the LWR spent fuel pyroreduction process. This in turn depends on the particular process employed, as is apparent from a comparison of Tables I and AI. The size and shape of the ingot are also important factors.

In the reduction stage of the salt transport process, Table I indicates that about 1/3 of the cesium inventory is retained with the TRU-containing product. If the uranium precipitate is collected and re-blended with the TRU product, then 100% of the transition metals (i.e., Ru) charged will be retained in the final mixture, since they are partitioned between the liquid metal and precipitate phases. Corium, which is a rare earth, will also be entirely retained. Europium is completely lost to the salt, and probably so are semimetals (Sb). Also, activation products in fuel structural materials (Co-60) will have been removed and discarded separately. Therefore, the gamma dose will be generated by Cs-134, Ru-106, and Ce-144 only. We will further assume, conservatively, that extraction of the product from the Cu-Mg solvent metal will be carried out so that none of these radionuclides are lost. Such an extraction has not yet been identified.

As a first step, consider an oxide fuel assembly with the same dimensions as the one modeled for Table AI, but with these radionuclide concentrations. The dose rate after 5 years would then be about 16 Sv/hr. After 15 years, the rate would decline to around 6.8 Sv/hr and would be due almost entirely to Cs-137 emissions. The 5-year dose rates are greater than those of vitrified waste canisters and certainly meet the spent fuel standard.

To correct this result, the fact that the average density of the ingot is greater than that of the fuel assembly must be taken into account. There are two reasons why the ingot is more dense: first, uranium metal has a higher density than uranium oxide; and second, whereas the ingot would be composed of solid metal, the assembly contains voids between the fuel rods. A greater density implies that the gamma radiation source density will also be greater. However, for fixed outer dimensions, a solid metallic ingot will experience greater gamma radiation self-shielding than an oxide LWR fuel assembly would. Also, the attenuation coefficients of metallic uranium are slightly greater than those of uranium oxide. The overall effect of the greater density of the ingot will be smaller dose rates.

The density of a metal alloy of composition U-Pu-10 wt-% Zr is 15.5 g/cm³ at STP. With the addition of a few weight-percent fission product metals, this may decrease the density by a few percent, to about 15 g/cm³. This is 4.6 times the average density of 3.24 g/cm³ that was used in the oxide fuel assembly calculations. Assuming that the ingot has the same outer dimensions as a PWR fuel assembly, it can be shown that the
contributions of radionuclides to the dose rate at one meter from the ingot will be smaller than those in the fuel assembly by between 30%-65%, depending on the gamma-ray energy. The dose rate from a metallic ingot of the same size and shape as a LWR fuel assembly would be around 6.5 Sv/hr after 5 years of cooling, decreasing to 2.1 Sv/hr after 15 years. Changing the radius of the ingot would not help, because it can be shown that the dose rate as a function of ingot radius is near its maximum at the above value. These dose rates are somewhat smaller than those of vitrified HLW canisters.

The lithium process would provide even less self-protection. According to Table I, the only gamma-emitting radionuclides of consequence in the product would be Ru-106 and Am-241. A glance at Table AI demonstrates that the corresponding dose rate at five years would be around 1 Sv/hr and would be unacceptably small. Therefore, it does not appear to be suitable for the fabrication of interim storage ingots.

Refabricated IFR fuel

The metallic ingots proposed as storage matrices for separated plutonium would have to be pyroprocessed and recast as fuel elements before they could be used as IFR fuel. The IFR pyroprocess, also referred to as electrorefining, is based on the partitioning of metals between a liquid metallic anode, metallic cathodes, and a molten chloride salt electrolyte. The process efficiently separates many of the highly radioactive radionuclides from the product stream, with cesium and other alkali metals dissolving entirely in the salt phase, and ruthenium and other noble metals remaining in the anode. However, the plutonium-containing product, which is collected at a liquid cadmium cathode, will be contaminated with a fraction of some of the lanthanide fission products, as well as minor actinides. Not all lanthanides are transported with the product to the cathode: for example, experiments at Argonne have shown that europium is entirely transferred to the molten salt phase.64

Of all the fission product radionuclides in Table AI, only Ce-144 is transported in non-negligible amounts with the product stream. However, since CeCLi is much more stable than PuCLi under process conditions, only about 13% of the cerium fed into the electrorefiner is transported to the cadmium cathode, assuming that the correspondence of its activity with the product is 80% for plutonium.65

The amount of cerium per unit mass of plutonium that will be present in the product depends on the characteristics of the spent fuel fed to the electrorefiner. The concentration of Ce-144 in spent fuel is roughly the same for LWR fuel and for fast reactor fuel (averaged over core and blankets), for equivalent energy production, about 4.8 x 10⁻⁴ Tban/mole. The plutonium concentration of the IFR spent fuel is about 12% (again averaged over core and blankets), while it is about 1% for LWR fuel. Therefore, the amount of cerium per tonne of plutonium is about twelve times less for FBR spent fuel than for LWR spent fuel. A fresh IFR core fuel element will contain about 25% recycle plutonium. If this plutonium were obtained from the pyroprocessing of IFR spent fuel, then the cerium concentration in the fresh fuel would be 0.13 x (25/12) = 0.3 times that of the spent fuel (either LWR or IFR). On the other hand, if the plutonium were obtained via the salt transport process from spent LWR fuel, then only about 70% of the cerium charged would be present in the salt transport product.66 Then the cerium concentration in the fresh IFR fuel would be 0.13 x (25/12) x 0.7 = 2.3 times greater than it is in LWR spent fuel. A third possibility is that the plutonium would be recovered from interim storage ingots derived from LWR spent fuel and weapons plutonium. If the ingots contained 1% additional weapons plutonium, the total plutonium concentration would be about 2%. Then the cerium concentration would be 0.13 x (25/12) x 1.6 times greater than in LWR spent fuel.

Modelling the IFR fuel assembly at first as an oxide assembly, the dose rate can be estimated from Table AI. Using the information in the previous paragraphs, the dose rate after 5 years' cooling due to the residual Ce-144 contamination would be 0.09 Sv/hr, 0.7 Sv/hr and 0.5 Sv/hr for plutonium recovered from spent IFR fuel, spent LWR fuel or interim storage ingots (at 1% additional plutonium loading), respectively.

Some of the minor actinides in the IFR product, Am-241 in particular, provide contributions to the dose rate that increase with time. However, as is apparent from Table AI, even at their maximum values these contributions are far smaller than those of the fission products. In the case of IFR fuel fabricated from LWR recycle plutonium, in which the actinide concentrations would be about 25 times larger than those in Table AI, the dose rate due to Am-241 would still be below 0.01 Sv/hr. The IFR fuel is actually made of the denser metallic uranium, and the reference design fuel assemblies are shorter and narrower than LWR fuel assemblies. The self-shielding effects are not as large as they were in the solid metallic ingot, because the IFR fuel assembly contains voids and has an average density not much greater than that of the PWR assembly; on the other hand, the linear source density is much smaller for the IFR assembly. Also, the shorter IFR fuel assembly would generate lower dose rates at points on the perpendicular bisector of the assembly axis. The net effect is that the IFR dose rate would be about 65% less than the dose rates for the three cases computed above, or 0.03 Sv/hr, 0.24 Sv/hr and 0.18 Sv/hr.67 These values are below the IAEA minimum self-protection criterion. In addition, since the Ce-144 half-life is only 285 days, the dose rate of the IFR product would decline sharply with time. It is clear that the refabricated IFR fuel assemblies would possess negligible diversion resistance, if the time for cooling, reprocessing, refabrication and subsequent storage were to exceed five years.

This would create a highly undesirable window of low diversion resistance in the fuel cycle, at which point "stored weapons standard" security and safeguards would be necessary.

It has been suggested that modifications to the reference IFR pyroprocess could lead to a product with enhanced self-protection. Even if 100% of the Ce-144 inventory were retained (an increase by a factor of 8), it would only delay the decline of the dose rate to very small values by less than three half-lives, or under two years. Modifications to the process to retain the more useful Ce-137 or Eu-154 in the product would appear to be extremely difficult without major changes, because of the very different chemistries of these highly active metals compared to the transuranic elements in the electrorefiner environment.

Therefore, in the time interval between pyroprocessing and loading into the core,
the diversion resistance of IFR fuel assemblies must be regarded as minimal, if the original spent fuel is cooled for more than a few years.

Ceramic slugs, refabricated AIROX fuel

From Table A1 and the information provided in the text, the dose rates generated by AIROX products at various stages of the process can be estimated. In particular, it will be necessary to evaluate the effect of high-temperature sintering on the radiation barrier of a fresh fuel assembly fabricated from AIROX-processed spent LWR fuel and weapons plutonium. After sintering at 1700°C, the AIROX fuel would retain from 5%-20% of the cesium and 50%-80% of the ruthenium inventories of the original LWR spent fuel. Other radionuclides listed in Table A1 are non-volatile and would entirely remain in the fuel, except for Co-60, which is generated by activation in the structural materials, which would be removed prior to AIROX processing, and Sb-125, which is semi-volatile but was not listed in the literature as having been liberated during tests. The residual radionuclides would generate a dose rate of around 9.2-16 Sv/hr after cooling for 5 years, and 3.4-6.1 Sv/hr after 15 years. These values are 75%-85% smaller than those generated by spent LWR fuel of the same age. In absolute terms, the 5-year dose rates are still significant and meet the spent fuel standard. However, the loss of Cs-137 results in a much steeper decline of dose rate with time than occurs in spent fuel.

The dose rates generated by ceramic slugs would be somewhat smaller than these values for fuel assemblies, for the same radionuclide concentration. The difference in densities, though, is not as great as in the case of the metal ingots. The slug density would be about 10 g/cm3, or 3.1 times the fuel assembly density. This results in a dose rate of about 60% of that of the fuel assembly. However, the ceramic slugs, which are fabricated at lower temperatures than the fuel assemblies, would have much greater cesium retention, which would more than offset this effect. Similarly, fuel assemblies fabricated with the low temperature ViPac process would have dose rates within 20% of spent LWR fuel.

Acknowledgements

The author would like to thank Oleg Bukharin and Harold Feveson of the Center for Energy and Environmental Studies at Princeton University for helpful conversations, and Marvin Miller of the Department of Nuclear Engineering at the Massachusetts Institute of Technology for the review of an early draft.

Notes and References

3. In November 1993, U.S. Secretary of Energy O'Leary disclosed a figure for the quantity of weapons-grade plutonium produced by the United States, 89 tons (which independent researchers have claimed is 1.4 tonnes too low). However, the US has not revealed the fraction of the plutonium inventory which is still in weapons. No disclosures of this type have been made by the FSU.
8. The FMEF was originally built to house facilities supporting a fast breeder reactor, including a MOX fabrication line with a throughput of 6 tHM/yr and a reprocessing plant with a 15 tHM/yr throughput. Most of the equipment was never installed. In addition to processing areas where MOX fabrication could take place, the building contains 17 hot cells of various sizes. Therefore, it is suitable to support a wide range of nuclear materials processing activities.
9. F. Berkhoot, et al., op. cit. The Palo Verde reactors are rated at 3817 MWe. At a burnup of 33 GWd/yr and a capacity factor of 70%, the annual fuel loading per reactor would be about 29.5 tHM, containing about 0.84 t of weapons-grade plutonium.


17. V. Mikhailov et al., op. cit.


25. C. McPheters, et al., op. cit.


27. C. McPheters, op. cit.


32. A 1000 MW(thermal) fast breeder reactor with breeding ratio of 1.25, operating at a capacity factor of 0.8, produces about 250 kg of extra plutonium per year, assuming a 75% fissile content. At a breeding ratio of 1.65, about (0.65/0.25)*(250 kg = 700 kg would be produced per year. This is comparable to the production rate of a natural uranium-fueled reactor optimized for plutonium production (assuming 1 gram of plutonium produced per MWd-thermal) although the latter would have much greater natural uranium requirements and generate far more spent fuel in the process. In contrast, a typical LWR would produce about 300 kg of plutonium per year. Data is taken from Table 1-3-2 in A. Nero, Jr., A


35. R. Wymer, et al., op. cit.

36. In R. Wymer et al., op.cit., the cooling and recycle period is assumed to be 4.5 years. 5-year cooling is standard for contemporary reprocessing plant operations. The time for repackaging and refabrication is apparently short compared to the cooling time.


41. The total Cs-137 activity contained in the capsules at Hanford is about 50 MCI (1.9 million TBq). The Cs-137 concentration necessary for a "dirty" MOX fuel rod to meet the 10-year cooled spent fuel standard is about 0.1 MCI/MM. Furthermore, initial Cs-137 loadings would have to be even greater to compensate for losses due to volatilization that might occur during the sintering of the fuel rods. The total Cs-137 requirement for 1800 tonnes of fuel would therefore be of order 180 MCI, more than three times the available inventory. However, the dose rates that would be achievable if cesium volatilization were suppressed would be on the order of 15 Sv/hr, which is still adequate to meet the spent fuel standard.

42. Oleg Bukharin, Princeton University, personal communication, April 1984.

43. V. Mikhailov, et al., op. cit.

44. W. Hannum, Argonne National Laboratory, personal communication, June 1994.


51. D. Majumdar, et al., op. cit.


55. This assumes a residual fission enrichment in the spent fuel of about 1.5% (U-235 and Pu-239 combined), so that 4% weapons-grade plutonium must be added.


58. A. Ringwood, et al., op. cit.


64. V. Mikhailov et al., op. cit.


70. The plutonium inventory at a MOX fabrication plant is greatest at the head-end, where pure PuO2 feed is stored, and is considerably smaller at later process stages. Scaling from a flowsheet for a reference 200 THM/yr plant (Ref. 14), a plant with a throughput of 60 THM/yr would store about 210 kg of plutonium (in the form of PuO2) in the head-end, about 600 kg/M of MOX powder (containing 18 kg plutonium) in silos awaiting pelletization, and about 5.3 t-M of finished MOX fuel rods (containing 160 kg plutonium) awaiting transport. In-process batch sizes, such as the charges to the sintering furnace and to the scrap recovery reactors, are on the order of 45 kg/M of MOX (1.4 kg of plutonium).

71. EPRI, 1979, op. cit.

72. Carbon-14 generated in the cladding would not be released during AIROX treatment. Whether it would be released later depends on the processing method chosen for the hulls.

73. For example, the newly commissioned Thermal Oxide Reprocessing Plant (THORP) at Sellafield has an installed waste treatment system that will scrub C-14 from the off-gas. It is anticipated that this will reduce stack emissions of C-14 by a factor of about 80.


76. Most of the Kr-85 generated in spent fuel is trapped within the fuel cladding and is only released during reprocessing. Less than 1% of the total production, or about 100 TBq/Gwe-yr, is released by failed fuel rods during reactor operation. Therefore, the Kr-85 credit is (1800 TBq/Gwe-yr)(95 Gwe-yr/yr)/6.8x10^5 TBq/yr = 1.3x10^5 Bq/yr.


78. W. Lloyd, et al., op. cit.


81. The dimensions of the Westinghouse 15x15 PWR fuel assembly are: an active fuel length of 5.66 m and a flat-to-flat dimension of 0.21 m. The assembly contains a mass of 0.47 t-M. This corresponds to an average density of 3.24 g/cm^3. From W. Lloyd et al., op. cit.

83. This estimate can be derived from information provided in Table I of Bauer et al., "Behavior of Metallic Fuel in Treat Tests," Nuclear Technology 92 (1990) 325.


85. J. Ackerman and T. Johnson, "New High-Level Waste Management Technology for IFR Pyroprocessing Wastes," Argonne National Laboratory, CONF-930913–31, September 1993. The separation factor, which characterizes the partitioning of an element between the salt and metal phases relative to that of uranium, is 1.66 for plutonium and 49 for cerium under process conditions, implying that cerium favors the salt phase to a greater extent than plutonium does. Apparently about 20% of the transuranics fed to the electrorefiner end up in the salt phase. This implies that the ratio of cerium concentrations in the salt and cadmium cathodes is approximately (49/1.66) x (1/4) = 6.5, or the fraction of the cerium inventory transported to the cathode is 1/7.5 = 0.13.


87. A reference IFR driver fuel assembly (PRISM Mod B/92) has an active fuel length of 107 cm (42 in), a flat-to-flat dimension of 14.3 cm (5.63 in), and a mass of 80 kgf/m. It is composed of 217 cylindrical fuel pins with an outer cladding diameter of 7.4 mm (0.293 in), a cladding wall thickness of 0.46 mm (0.018 in), and a fuel smear density of 75% (which means that when the fuel metal slug expands during irradiation to fill the available space, the final density will be 75% of theoretical). Each fuel pin has a mass of 0.97 g/kgf/m. This corresponds to an average density of 3.65 grams per cubic centimeter in the fuel assembly. This difference in density and radius relative to the PWR fuel assembly results in a 35% decrease in the dose rate. Also, because the IFR fuel assembly is less than one-third the length of an LWR fuel assembly, there is an additional 50% reduction in the dose rate at one meter, so that the overall dose rate is about 65% smaller than from a PWR assembly.

**Figure 1: IFR-based interim conversion (Cu-Mg solvent)**

- **Reduction**
  - Pu+MA+RE+1/3 Cs+Cu-Mg
  - U+NM metal

- **Extraction**
  - Co-Mg
  - Pu+MA+RE+1/3 Cs

- **Alloying**
  - W-Pu metal
  - Ingot 2% Pu

- **Interim Storage**
  - Ingot

- **Pyroprocessing and Refabrication**
  - Fresh fuel 25% Pu (0.15 Cs-144)
  - LLW, HLW (Cs, 0.85 Cs-144)

- **IFR**
  - Spent fuel 12% Pu

**Definitions:**
- NM = noble metals
- MA = minor actinides
- RE = rare earths
- --- = alternative pathways
- ■ = undetermined process
Figure 2: AIROX-based interim conversion (minimum decontamination flowsheet)

**Legend:**
- FS = fission products
- MA = minor actinides (Am, Cm, ...)
- "alternative pathways"

**Diagram:**
- LWR spent fuel → AIROX → off-gas treatment (Hg, C, K, N)
- oxidation → PuO₂ → blending → compaction (U, FS, Pu, MAO₂) (1% Pu)
- interim storage
- slugs → 5% Cs, Ru → Vi-Pac fuel fabrication → "dirty" MOX assemblies
- LWR spent MOX fuel → repository
- LWR spent MOX fuel → repository