LWR Recycle: Necessity or Impediment?

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Abstract – The nuclear fuel cycle can be truly closed by supplementing today's thermal reactors with fast reactors, which can use as fuel the heavy, fissionable isotopes that accumulate in thermal-reactor fuel. In a fully closed cycle, the waste for disposal consists only of fission products with trace amounts of actinides. Eliminating the transuranics reduces the heat load in the repository, increasing its capacity by about a factor of five. This permits the expanded use of LWRs to produce pollution-free electricity and reduce dependence on foreign oil.

Cycling back to LWRs amounts to an expensive storage option that puts off for maybe a decade or two the need to deal effectively with the transuranics. That delay is bought at the cost of implementing and operating the thermal-recycle infrastructure, with extra expense later on because the resulting spent fuel would significantly complicate fast-reactor processing.

Before DOE undertakes development of technologies for thermal recycle, the viability of forgoing it altogether should be carefully assessed— it might turn out to be an impediment rather than a necessity. Since there is no need in the long term for such an infrastructure, the energy and dollars needed to implement it might be better spent on wrapping up the development fast reactors and their fuel cycle. It is not too early to embark seriously on a program to deploy pyroprocessing and fast reactors, for ultimate closure of the fuel cycle and optimal long-term utilization of the Yucca Mountain repository.

I. INTRODUCTION

At present the nuclear fuel cycle is "open"—that is, the spent fuel that is now considered waste still contains most of the energy it started with. Current U.S. policy is to use the fuel once and then throw it away, along with more than 95% of its original energy. In addition, a huge amount of energy is latent in the depleted-uranium residue from military and civilian enrichment activities. In a fully closed cycle, essentially all of the energy in the mined uranium would be exploited, with only the real waste—the fission products left over for disposal.

The fuel cycle cannot be closed with today's thermal reactors by themselves, even with recycling.^{*} It can be done, however, by supplementing them with fast reactors, which can use as fuel the heavy, fissionable isotopes that accumulate in thermal-reactor fuel. More about that later.

There are sound reasons for U.S. policymakers to expedite the development of a closed nuclear fuel cycle. The most compelling goal is the ability to expand the use of environmentally friendly nuclear energy without having to begin planning immediately for a second geologic repository. Eventually other motivations, such as managing resources responsibly and limiting the net production of plutonium, come into play, but the criteria for deciding whether and how to close the fuel cycle have to be consistent with efficient waste management. The overriding reason for closing the fuel cycle sooner rather than later is to make the most efficient use of the Yucca Mountain repository—to maximize the amount of nuclear electricity that can be produced before the repository's capacity to accommodate high-level waste is exceeded.

Other reasons include controlling costs, minimizing risk to future generations, using resources efficiently, and implementing a safe operating strategy for the repository.

The purpose of this paper is to provide a rational framework for evaluating the potential role of mixed uraniumplutonium oxide fuel (MOX) in a U.S. waste-management strategy that is based on a closed cycle.

II. REPOSITORY LIMITATIONS

Nuclear waste, in whatever form, is quite compact, so its physical volume has only a minor influence on how efficiently the repository is utilized. The important factor is heat generation: the maximum waste loading in a repository like Yucca Mountain is determined by various temperature limits

Numerous analyses have been performed to determine the maximum acceptable temperatures in the Yucca Mountain design. There are several temperature limits, including the centerline temperature of the waste package, the temperature of the container, and the temperature of the wall of the drift (the emplacement tunnel), but the controlling limit in the present design is the temperature between drifts.

For direct disposal of used fuel, the inter-drift temperature reaches its peak some thousand years after the waste is emplaced. That is determined by the long-term heat source

^{*} In a thermal reactor the neutrons are thermalized (slowed down, or "moderated"). Reactors moderated by ordinary water are called light-water reactors (LWRs). Other commonly used moderators are heavy water and graphite. In the United States, virtually all the power reactors in operation today are LWRs.

—mainly the transuranic elements^{*} in the spent fuel. Although plutonium is the most abundant of the transuranics, long-term heat production is dominated by americium and neptunium—which, along with curium, are known as the "minor actinides." The minor actinides also dominate the long-term radiotoxicity of the waste, so managing them appropriately is also key to reducing the potential for harm to individuals many generations from now.

Substantial improvement in the utilization of the repository, therefore, requires removing the transuranics from the waste—a conclusion reached by every study that has addressed the question. The best-known study, performed by the National Academy of Sciences,[1] concludes that eliminating most of the transuranics improves the utilization of a repository by about a factor of five.

Most of the various schemes for separating and managing fission products are of second-order importance. In principle, however, a further order-of-magnitude improvement could be achieved by removing from the waste stream the two elements supplying the highest heat load cesium and strontium. Further development work would be needed to confirm that the additional step is economically justified.[2]

With a fully closed fuel cycle, the waste for disposal consists of fission products with only trace amounts of actinides. As a result, the radiotoxicity of the contents of a repository will be below that of the original ore in well under a thousand years—which should allay any perception of hazard from long-term leakage or geologic instability.

III. REMOVING THE TRANSURANICS

The transuranics can be kept out of the waste that goes into the repository by processing the spent fuel chemically, to separate it into the following three constituent parts, which can be managed separately: (a) most of the uranium; (b) the transuranics, together with a roughly equal amount of uranium; and (c) the real waste—the fission products—for disposal in the repository.

Today's commercial spent fuel is approximately 94.5% unused uranium, 3.6% solid fission products, and 1.2% transuranics. Of the transuranics, about 84% is plutonium, with minor actinides comprising the balance—roughly 0.2% of the total mass of the initial unirradiated uranium. About 5 kg of plutonium and 1 kg of minor actinides are contained in a spent fuel assembly (initial uranium mass 500 kg) from a pressurized-water reactor (the most common type of LWR).

Following separation, closing the fuel cycle requires preparing the 18 kg of fission products for permanent disposal, storing most of the 473 kg of unused uranium (0.8% enrichment) until it is needed as fuel,^{**} and recycling

the 6 kg of transuranics (along with a similar amount of the uranium), to consume them while producing energy. There are a number of ways to do this. The Advanced Fuel Cycle Initiative (AFCI) and Generation-IV (Gen-IV) programs envision exhaustive investigation of a variety of alternative technologies that are not yet fully developed.

However, a logical start to the process of closing the U.S. fuel cycle would be to consider the technologies that have already been established.

IV. MOX TECHNOLOGY

There are no international examples of fully closed systems, although reprocessing, using mixed plutoniumuranium oxide, is performed by several countries. Some of them have taken sporadic steps toward closure, most prominently France, but none has yet completed the job.

Mixed-oxide technology is well developed. The UK and Russia have been reprocessing reactor fuel for half a century. The United States operated several reprocessing facilities until President Carter banned the practice in 1977. France has established a large reprocessing capacity (~2000 metric tons of heavy metal per year) to service both domestic and international customers. Japan is embarking on a similar program.

The technology is based on the aqueous Purex^{***} technique. France has made large strides in producing compact waste forms (an achievement not duplicated by the U.S. Department of Energy in producing its weapons plutonium). With this capacity up and running, France has been accumulating separated plutonium at a fairly brisk rate. Some of the plutonium is used in MOX by the nation's large fleet of LWR power plants, but France can separate more plutonium than can currently be absorbed by facilities that can utilize it. Because reprocessing is an international business in France, some plutonium is returned to the countries of origin.

Worldwide as well, the capacity to produce MOX does not yet match the rate at which plutonium is separated—for a number of reasons, the most basic being that there are not enough power plants that can use the fuel. Because the accumulation of separated plutonium is considered a serious national problem, France is developing ambitious schemes for multiple recycle. If implemented, those schemes would eventually lead to an equilibrium amount of plutonium in commerce within the country. However, France does not yet have even a design for a geologic repository, so that the specific requirements for form and composition of the highlevel waste are not confirmed. Thus the minor actinides are now being treated as waste—they are stored temporarily, along with the fission products.

^{*} Transuranic elements have atomic number greater than 92 (uranium). The first four transuranics are neptunium, plutonium, americium, and curium.

^{**} Burnup in LWRs being about 3.5%, the enrichment of the uranium in the used fuel has been reduced from its initial 4.4% to about 0.8%—somewhat greater than natural uranium (which is 0.7% U-235). It can be re-enriched for re-use in LWRs. As-is, it can be used as fuel for CANDU reactors, and is a useful fuel

component for fast reactors.

^{****} Purex is a chemical reprocessing method that was developed in the weapons program to produce chemically pure plutonium. Since it uses a water-based solvent, the process is called "aqueous." It is the reprocessing method of choice for thermalreactor fuel. Fast-reactor fuel can be processed by a "dry" pyrometallurgical method, which uses molten salt as the solvent and does not produce pure plutonium.

V. FAST-REACTOR TECHNOLOGY

A technology that fully closes the fuel cycle must consume the plutonium and minor actinides almost completely. Currently, at least, that can only be done in a fastneutron spectrum.

Under the present schedule, the United States is putting off the decision as to whether to close its fuel cycle until the year 2030.[3] That decision could be made much sooner, however. Technologies that can do the job have already been established or are close to being demonstrated.

Of potential fast-neutron systems, the one that is closest to commercial viability is the Advanced Liquid Metal Reactor (ALMR; PRISM), developed by General Electric with support from Argonne National Laboratory, [3] and converted by GE to a larger design called Super-PRISM (S-PRISM).[4] The reactor uses metallic fuel and a liquidmetal coolant (sodium), and is passively safe. It operates in conjunction with a pyrometallurgical reprocessing facility that is part of the reactor complex,^{*} thereby minimizing the need to transport plutonium and spent fuel.

The pyroprocess is non-aqueous and exceptionally proliferation resistant—its plutonium is sequestered in an inert atmosphere in very radioactive surroundings, never has the chemical purity needed nor the isotopic purity desirable for weapons, and never leaves the complex during the plant's lifetime (except for possible shipment of startup fuel for a new plant, when spent fuel from thermal reactors is no longer available).

The details of a feasible system for integrating the thermal- and fast-reactor cycles have been presented by Dubberly et al.[5]

Ehrman et al. have shown that LWR spent fuel can be processed to supply LMRs at no cost to the government the cost being covered by the (competitive) busbar cost of power from the LMRs.[6]

In 1994 a consortium headed by General Electric proposed to design, construct, and test a functioning prototype ALMR in less than fifteen years. Such a project could be initiated immediately, while optimization studies for future systems proceed in parallel under Gen-IV.

VI. U.S. POLICY

Like France, the United States has a large fleet of LWRs that generate spent fuel, and therefore using MOX fuel in tandem with a large reprocessing capacity and an expanding fleet of LWRs might seem to be a logical next step in closing the fuel cycle. After all, as a part of the disarmament agreement with Russia, the United States has already committed to dispose of some 35 metric tons of weapons-grade plutonium by irradiating it as MOX in commercial nuclear reactors.

On the accumulation of separated civilian plutonium, however, the United States is more restrictive than most

other heavily industrialized nations, and its policy will need to be modified. All potential ways to increase the utilization of the U.S. geologic repository involve processing the spent fuel to separate the fission products, stabilizing them in waste forms engineered for disposal, and emplacing those wastes in the repository. The uranium can be stored in a surface facility for future use, rather than extravagantly disposing of it as low-level waste. The more difficult constituents, the transuranics, must be stored in safe, secure facilities until they can be consumed as fuel in fast reactors.

In the LWR MOX approach, then, plutonium with a low-enough fraction of higher isotopes can be recycled, but most of the minor actinides and some of the key plutonium isotopes do not fission in the thermal spectrum. Thus the minor actinides and the more highly degraded plutonium would be accumulated and stored until fast reactors were available to finish the job.

Once the MOX had been irradiated, it would still contain a substantial amount of plutonium, along with a fresh batch of minor actinides. If this fuel is reprocessed, the recovered plutonium is less suitable for LWR use, because the fraction of fissile isotopes is seriously degraded. This problem can be mitigated, but only somewhat, by blending that plutonium with fresher material obtained by reprocessing normal commercial uranium-oxide fuel.

There are various other, complex schemes for multiple thermal recycling. One suggestion is that some neptunium, a minor actinide, should remain with the plutonium throughout the separation and fuel-production steps. That would have a nonproliferation purpose: since pure plutonium is not separated, a policy conflict would be avoided, and the product would be more difficult to handle—although, with suitable shielding, chemical separation for illicit weapons production could be accomplished. DOE has referred this issue to a blue-ribbon committee for resolution.

VII. BENEFITS OF MOX

The benefits of using MOX fuel in the existing fleet of thermal reactors are obvious. First, the reactors are already deployed, and enough of them are suitable for conversion to MOX (up to one-third of the core can be used) that the plutonium output from a large reprocessing plant could be accommodated. Presumably, as newer reactor plants started up, their updated designs would be more compatible with MOX.

There is sufficient experience worldwide to permit the cost of reactor conversion, efficiency reductions, and fuel fabrication to be predicted with enough confidence to permit an informed decision as to whether to proceed. Because the base technologies involved—Purex, LWRs, and MOX fabrication—have 50-year-old roots, the risk of technical failure is minimal.

Although the benefits are not inconsequential, the MOX alternative needs to be examined in terms of a number of objective measures, to see whether it is really the right way for the United States to go.

^{*} The plant for processing LWR spent fuel would supply the initial loading for more than one ALMR, and therefore would not necessarily be part of a reactor complex.

VIII. DOWNSIDE OF MOX

The obvious downside of the MOX alternative is that it only delays facing up to the real repository issues. Fundamentally, MOX recycle to LWRs amounts to a storage option that puts off for a decade or two the need to deal effectively with the transuranics. In the end, fast-fission systems will have to be deployed. They must constitute at least 20% of the total nuclear capacity if they are to eliminate enough of the plutonium and minor actinides to have a substantial impact on the repository.

The delay would be purchased at considerable cost, some of it incurred early on, and the rest when the time arrived to use the residue in fast reactors:

- If the delay is not needed, the entire MOX-handling infrastructure is unnecessary.
- An aggressive MOX scheme means multiple recycle of the plutonium. After each cycle the buildup of evennumbered plutonium isotopes and other non-fissile, alpha-emitting actinides renders the recycled product both harder to work with and less useful as thermalreactor fuel. France is planning on not more than two recycles, because of the expense of modifying their MOX-handling facilities to meet the enhanced safety requirements.
- Even for the eventual fast system, the increased fraction of heavy isotopes makes the mixture significantly harder to handle. Because of the energetic alpha decay and the relatively short half life of curium and the higher plutonium isotopes, small actinide-bearing particles will be quite mobile. That makes it a challenge to confine the radioactivity and decontaminate the finished fuel assemblies, to prevent worker uptake of actinides. Thus the product of actinide recycle will have to be handled in heavily shielded facilities that are advanced relative to most of today's hot cells.**
- Although fuel samples containing neptunium and americium have been successfully fabricated, the same cannot be said for curium.
- The thermal-reactor option reduces the fissile-isotope component of the actinides, leaving an enhanced inventory of minor actinides and even-numbered plutonium isotopes. Design of fast-fission systems that can safely handle a core based on a varying isotopic distribution dominated by minor actinides will necessarily be conservative. That increases the cost relative to fast-reactor facilities of more conventional design that would be loaded with actinides discharged from typical LWRs.

IX. DEPLOYMENT OF FAST REACTORS

Much of today's fuel cycle program in the United States is inherited from the Advanced Accelerator Application (AAA) program. When this program started, the fundamental premise was an expected phase-out of nuclear energy, with accelerator-driven fast-fission systems consuming the transuranics left from the nuclear era, thereby eliminating most of the radiological risk to future generations. Now, however, that objective has been abandoned, and the current Advanced Fuel Cycle Initiative (AFCI) recognizes that if there is to be affordable electricity and plentiful hydrogen for transportation, increasing use of nuclear power is inevitable.

The capacity of Yucca Mountain has been shown to be adequate for directly disposing of the spent fuel from the present fleet of nuclear power plants. Therefore, if nuclear electricity is to be phased out, the fuel cycle does not need to be closed—the ultimate reason to close it is to permit substantial growth of nuclear energy.

In a long-term growth scenario, fast reactors are both desirable and necessary. At some point, they will be needed for responsible management of the uranium resources. With only thermal reactors, if nuclear power grows globally at even 2% per year, all currently known and speculative uranium reserves will be depleted by mid-century.[7] If a fast reactor infrastructure is established as part of an economical waste-management system with a closed fuel cycle, the transition to widespread deployment of fast reactors, and therefore sustained nuclear development, will be much more feasible.

In comparison with the present once-through fuel cycle, fast-spectrum actinide burners will extract more than 100 times as much energy from each pound of original uranium.

X. NONPROLIFERATION POLICY

Closing the nuclear fuel cycle in the United States will require some modification of the national nonproliferation policy, regardless of the option that is ultimately chosen. The Purex/MOX route, no matter how it is approached, will require separation and some storage of civilian plutonium, as well as open-ended storage of minor actinides. This reality cannot be changed by blending a token amount of neptunium into the fuel. Neptunium would add no measurable proliferation resistance, nor would it reduce the cost of safeguarding the fresh fuel. It would, however, greatly increase the cost of fabrication and fuel handling, because there is no experience with manufacturing MOX when operation, maintenance, and quality assurance have to be accomplished completely by remote control.

Also, power-generation companies might not be willing to accept the fuel, because handling it in the plant would be more difficult. If the LWR MOX route is selected, the overriding concern should be producing the fuel that would be most acceptable to the power producers. Nonproliferation issues would then have to be resolved through appropriate policy governing safeguards, security, and export control.

^{*} The small particles are impelled by the recoil when an alpha particle is emitted.

^{***} The regular fuel from ALMRs also has to be handled remotely, but the logistics are much simpler because collocation permits the radioactive actinides to be transferred and processed without ever leaving the shielded facility.

XI. DISPOSITION PROGRAM FOR WEAPONS PLUTONIUM

The U.S. government has decided to implement the MOX strategy for disposing of weapons plutonium, under a Fissile Materials Disposition Program run by the Department of Energy. The program pays commercial reactor owners to accept and use the MOX fuel. On the surface, that would seem to lay the foundation for following up with a similar strategy for civilian plutonium (as in the weapons-plutonium program, the transuranics will be owned by the federal government).

However, the weapons program only has to handle some 35 tons of weapons plutonium over a couple of decades. Operations at that scale will have no impact on management of the civilian plutonium, which is currently accumulating in spent fuel at a rate of about 20 tons per year in the United States (worldwide, the plutonium inventory is headed toward 2,000 tons by 2010 or so). Further, because it lacks the intense external radiation of the plutonium discharged from LWRs, weapons-grade plutonium is much easier to handle. There will be interesting lessons from the current disposition program, but as a model for closing the fuel cycle it has little obvious relevance.

XII. ECONOMICS

The real issue is the relative economics of the alternatives. Because of the sunk cost in the already-deployed reactors, an alternative that would use them would seem to have an advantage over options that don't. However, since the LWR MOX approach does not do the whole job, one has to consider not only the cost of implementing that reprocessing system, but also the cost of dealing with the discharged MOX and the more complex fast-reactor infrastructure eventually needed to handle the actinides. The greater the emphasis on full utilization of the MOX potential, the more difficult and expensive will be management of the spent MOX at the end of the cycle.

In France, for example, with its already-in-place reprocessing facilities, fabrication facilities, and LWRs, but no repository, the MOX option is an easier choice as a delaying strategy. But the United States is starting with a much different infrastructure—we do have a designated repository and we do not have reprocessing or fuel-fabrication facilities.

There will also be costs for using existing commercial reactors: modifications, licensing, and loss of efficiency due to likely incompatible refueling intervals for the uranium and MOX fuels. These costs can probably be estimated with at least some confidence.

A recent OECD-NEA report on transmutation provides a cost comparison based on the price of electricity.[8] The report concludes that the cost of burning plutonium in LWRs, coupled with burning the actinides in fast reactors, would be slightly less than doing the whole job with fast reactors. The principal reason for predicting that using MOX for part of the job would be cheaper is the anticipated higher capital cost of fast reactors. However, the issue requires more study in the U.S. context, as the economics of fast reactors have not been well established—although at least one rather detailed and well-founded study indicates that they will be competitive.[9]

XIII. INTERNATIONAL LEADERSHIP

Giving top priority to implementing the aqueous reprocessing technology that has been established in Europe, and has been at the center of policy debates in the United States, would be likely to galvanize antinuclear activists. While the technology proposed for the MOX program is really advanced aqueous and not traditional Purex, that will be a fine distinction when the battle lines are drawn. With plutonium separation, which occurs when MOX is recycled for LWRs, it will be hard to distinguish that fuel cycling program from what the country turned away from twentyfive years ago.

MOX is a "me-too" technology, and choosing it will send a message abroad that will give other countries more latitude to pursue plutonium separation (unless, perhaps, we package it with an ambitious proposal for managing the international fuel supply). International leadership and longterm considerations should be influential in making technology decisions: what seems technically expedient today might be counterproductive in the long run. In the end, the decision may be that MOX for civilian plutonium would be desirable, but that case is far from made.

XIV. CONCLUSIONS

The MOX option for disposing of weapons plutonium has been decided upon, but the way to deal with civilian plutonium has not. Although doing it with MOX would be a technically interesting and ambitious undertaking, there are many reasons for delaying that decision until a comprehensive systems analysis has been done.

Quite possibly the entire proposed MOX-handling infrastructure is unnecessary. The only reason for the United States to proceed with large-scale MOX would be to try to manage waste in a nuclear-growth scenario so as to delay both additional repository construction and the deployment of fast reactors. However, LWR MOX is an incomplete solution that would actually complicate the transmutation that is needed for efficient use of the geologic repository.

Only an objective analysis can answer the question of whether short-term implementation of LWR MOX is right for the United States. That analysis should start immediately. The study should cover the complete fuel cycle system in all scenarios, and should try to detail the impact of an existing fast reactor infrastructure on future growth in the face of declining resources, rising prices, demands for electricity by an expanding population, and the need to reduce the environmental impact of energy production.

Thermal reactors are needed now for electricity production; fast reactors are needed very soon for waste management; and fast reactors will be needed for long-term nuclear growth and sustainability. Whether MOX-fueled LWRs are an essential part of a U.S. nuclear energy complex can only be answered by an objective analysis of the complete, evolving system

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That said, long-term there is no need for an LWR MOX infrastructure. The energy and dollars needed to implement it might be better spent on wrapping up the development of fast reactors and their fuel. Because it will take perhaps two decades to get significant deployment under way, it is not too early to embark seriously on a focused program of fastreactor implementation.

With a closed fuel cycle, the waste for disposal consists of fission products with only trace amounts of actinides. As a result, the radiotoxicity of the contents of a repository will be at or below that of the original ore in less than five hundred years—which should allay any perception of hazard from long-term leakage or geologic instability.

With sufficient focus, it should be possible to have a combined pyroprocessing plant and fuel-fabrication facility operating at Yucca Mountain by the time the first shipments of spent fuel arrive.

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