

Recycling Nuclear Waste

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American Physical Society Special Session on
Nuclear Reprocessing, Nuclear Proliferation, and Terrorism
15 April 2007

ABSTRACT

In the public mind, the foremost reservation about nuclear power is, "What can we do with the waste?" Fortunately there is an answer: We can use the worrisome, very long-lived components as fuel in the right kind of reactors, and then the rest becomes manageable. Will this lead to proliferation of nuclear weapons or to an increase in the threat of nuclear terrorism? Not necessarily. Prudent recycle of nuclear waste will actually reduce these threats—while also reducing the time that nuclear waste must be sequestered to a few hundred years instead of thousands.

Introduction

Emerging from decades at a plateau, nuclear power is growing globally at an accelerating rate. This has rekindled discussions over what to do with the waste, coming at a time of increasing concern over terrorism and worry about the spread on nuclear weapons to new, potentially unstable countries. It is time to rethink the approach to nuclear power.

When civilian nuclear power was first being developed, some 50 years ago, conditions were very different:

- Uranium was a rare commodity;
- Facilities for processing uranium, in particular for enriching it, were beyond the scope of all but the richest and most advanced nations;
- Plutonium produced in nuclear power plants could only be extracted from used nuclear fuel with great difficulty. With even more complex technology, plutonium from some types of used nuclear fuel could be made into nuclear weapons. Both of these technologies were considered to be beyond the scope of all but the most advanced nations.

To utilize nuclear power to generate electricity effectively, the strategy developed was to use the scarce uranium to produce both power and plutonium. The plutonium produced would be recovered for use in fast reactors, which, if carefully designed, could be net producers (breeders) of plutonium. That way, the energy from the rare isotope of uranium, U-235, could be stretched to keep up with the growth in energy demand.

Because a nuclear power plant can also produce plutonium for weapons, a treaty (the Nuclear Non-Proliferation Treaty—NPT) was developed, and almost all countries ratified it. Under the treaty, the nuclear-weapons states would assist non-weapons states in obtaining the benefits of nuclear power, in exchange

for a commitment to refrain from developing nuclear weapons. This voluntary agreement was widely respected by most countries.

However, by the mid-1970s the plutonium-breeding strategy had begun to be questioned, as several countries developed nuclear weapons outside the scope of the NPT. The United States abandoned plans to recover plutonium from used commercial reactor fuel (by *reprocessing*), and urged others to do the same. Nevertheless, reprocessing continued elsewhere—but only in nuclear-weapons states (both declared and undeclared), within their weapons establishments.

Now most of the basic conditions have changed or are changing:

- Uranium is now available around the world in amounts large enough to satisfy the needs of the current generation of reactors for at least several decades.
- A much more efficient technology for enriching uranium is available.¹
- The downsizing of weapons stockpiles has left a glut of weapons-quality plutonium and enriched uranium.
- Information is widely available on how to construct an unsophisticated nuclear bomb, given access either to weapons-quality plutonium or to highly enriched uranium.
- The inventories of plutonium and higher actinides² that are accumulating in spent fuel around the world are currently viewed as a liability to be disposed of, rather than an asset to be conserved.

¹ The ultracentrifuge. Also, work on laser enrichment is progressing.

² *Actinides* are elements with atomic number 89 (actinium) and above. In this paper, "actinides" means uranium and all transuranic elements, including plutonium.

In addition:

- The United States and other industrialized countries are becoming increasingly dependent on imported oil, leading to calls for significant increases in nuclear power generation.
- The programs for disposal of used nuclear fuel have been stymied.
- The change in the global balance of power following the collapse of the Soviet Union has led to increased concern about the potential acquisition of nuclear weapons by unstable governments or terrorist groups.

The new conditions call for a new strategy. We suggest that one can be constructed that is based largely on available technologies. For clarity in discussing the changes that will be necessary, we use the following terminology:

- We don't say *nuclear waste* or *spent fuel* when we mean *used fuel*, which is a very valuable commodity.
- We use *reprocessing* to refer only to the technology for extracting pure plutonium from used reactor fuel. In Part I, we discuss technologies for nuclear *recycle*, which are fundamentally different.
- In Part II we discuss what we will call *actinide-consuming reactors*—fast reactors that can consume as fuel *any and all actinides*, including plutonium and U-238.

Then, in Part III, we look at how the new strategy might affect the risk of nuclear proliferation and terrorism.

And in Part IV, we discuss briefly concepts such as *restricted deployment*, which could partially address proliferation concerns. The ultimate resolution of those concerns, however, will depend more on diplomatic initiatives than on nuclear power technology.

Part I. Nuclear Recycle: Closing the Fuel Cycle

A nuclear fuel cycle is either *open* (unused energy remains in fuel that is considered "spent") or *closed* (all the fuel's energy had been extracted, which requires recycling). Currently there is no fuel cycle in operation that is fully closed. The traditional reprocessing technology, called PUREX (Plutonium-URanium Extraction), which was developed to obtain plutonium for weapons, is used in France and elsewhere to cycle some of the plutonium back into thermal reactors—but that comes nowhere near to closing the fuel cycle: it results in perhaps a 20-percent improvement in fuel utilization, but still leaves in the used fuel some 95 percent of the energy contained in the original fuel—and utilizes less than one percent of the energy in the uranium that was mined.

Plutonium is always produced in a uranium-fueled reactor, as a consequence of irradiating the U-238 component with neutrons. The plutonium that PUREX extracts from ordinary used fuel has the chemical purity required by bomb designers, although not necessarily the isotopic quality. To get the high proportion of the Pu-239 isotope needed for weapons, the uranium must not be irradiated for more than a very limited time. That way some Pu-239 is produced, but little of it is further transmuted to heavier isotopes.

Clearly, since the world is currently awash in weapons plutonium,³ this process can meet the military requirements very well.

The commercial fuel cycle has different needs. For economic reasons, commercial fuel is kept in the reactor as long as possible. Eventually the combination of fuel depletion and buildup of byproducts (fission products) reaches the point where the chain reaction cannot be maintained. Then the fuel must be removed and either discarded or processed.

PUREX is ill-suited for closing the nuclear fuel cycle. For that task, an appropriate technology would extract the components that poison the nuclear chain reaction, and replace the material that has been consumed. PUREX reprocessing does not work that way. It is designed to extract pure plutonium, leaving everything else as a composite waste. It was adopted for commercial power *only* because both the technology and the (very expensive) facilities were available, and the latter were expected to be largely surplus to weapons programs once a suitable weapons stockpile had been achieved.

For removing the fission products so that the fuel can be recycled, a variety of technologies have been considered. Perhaps the simplest suggestion was to heat the used fuel to boil off various volatile fission products. However, since the negative effect of volatile fission products on the fission process is relatively small, this approach would have limited utility.

All the other approaches that have been considered involve dissolving the used fuel, and then separating the fission products from the usable materials. Two such technologies are of interest here.

UREX+. The first of these recycle technologies, called UREX+, begins with an acid dissolution, followed by traditional chemical extractions to selec-

³ Current estimates are that between the U.S. and Russia, there is some 260 tonnes of weapons-grade plutonium, and another some 3200 tonnes of reactor-grade plutonium, most of the latter incorporated in used reactor fuel. Several countries have stocks of separated reactor-grade plutonium.

tively pull out the major fission products. This process is well suited to high-volume throughput. It is similar in many ways to the initial stages of PUREX processing, but it does not involve separating plutonium from the other actinides. As will be seen in the next section, for consuming the actinides that are in used fuel from light-water reactors (LWRs), there is no need to go beyond separating the transuranic elements from the bulk of the fission products and much of the uranium.

The sequence is as follows: The used fuel is chopped and dissolved in an acid solution, and most of the uranium is extracted in a chemically pure form that can easily be stored for reuse. The cesium and strontium (the fission products that are responsible for most of the radioactivity for the first few hundred years) are recovered, to be stored securely while they decay. Then all the remaining fission products except the lanthanides are removed.⁴ The lanthanides are left mixed with the higher actinides to render the product "self-protecting" during shipment to the fuel-fabrication facility, which would be collocated with the actinide-consuming reactor.

At that final destination, the lanthanides are removed and mixed with the other fission products, notable among which are the long-lived fission products technetium and iodine, and packaged for disposal.

Thus LWR spent fuel is processed ultimately into four output streams:

- the bulk of the uranium
- the cesium and strontium
- the rest of the fission products
- the transuranic actinides mixed with some uranium

This opens the possibility that some of the recovered cesium and strontium could be used for commercial purposes such as sterilization of food and medical supplies, eliminating the need to produce such sources artificially. The excess can safely be stored and allowed to decay for a few hundred years, after which it can be disposed of as low-level waste.

It is likely that the other fission products (the quantities are small) would be prepared for prompt geologic disposal.

As with the PUREX process, the UREX+ technology is well suited to large-volume, high-throughput, continuous operation. The safeguards monitoring and sampling processes would be similar to those used for PUREX type operations, except that there would be no

⁴ *Lanthanides*: elements no. 57–70, one of which is europium (no. 63). Eu-154, has a relatively short half-life (8.6 years), rendering it very radioactive.

separated plutonium to protect. A UREX+ facility is large and expensive. Only a very few such plants will be needed, leading to appropriate concentration of safeguards measures.

Pyroprocessing. A second recycle technology is a high-temperature electrochemical (*pyrometallurgical*) separation process, or *pyroprocess*. It is ideally suited to the fuel that would be used in the actinide-consuming reactor discussed in the next section.

The essence of the pyroprocess is that used, metallic fuel is dissolved in a molten salt. Metallic fission products that do not dissolve can be mechanically filtered out. Volatile fission products boil off, and are collected for packaging and disposal. The actinides are collected electrochemically (electroplating, essentially). The other fission products are collected for disposal as the salt is recharged.

Considerable work has been done on this process, both in the United States and in Russia. To date, it has been implemented as a batch process. The batch size is limited to prevent an accidental self-sustaining nuclear chain reaction. The actinide product contains much of the uranium and all of the other actinides, along with a significant, unavoidable carryover of fission products. After removal of the product, parameters are adjusted to recover the remaining uranium, which is subsequently mixed with the actinide product and the makeup uranium when the new fuel is fabricated. A chemical process is then used to *recharge* the salt—i.e., to strip out the dissolved fission products. As with the UREX+ process, if there is a suitable market for some of the fission products, these can be extracted from the waste streams, or the more active components can be stabilized and stored to allow the bulk of the radioactivity to decay away before disposal.

As an alternative to UREX+ for treating the LWR spent fuel, the General Electric Company is proposing to use pyrometallurgical processing, once the oxide fuel has been reduced to metal. The output streams would be comparable with those from the UREX+ process.

Part II. The Actinide-Consuming Reactor

The original nuclear power plants, built for naval propulsion, were based on fissioning U-235. When an atom of U-235 fissions after capturing a neutron, it releases somewhat more than two neutrons.⁵ In a power reactor, one of these released neutrons is captured by another U-235 atom, keeping the process

⁵ The average number of neutrons released in fission was one of the most highly classified numbers of World War II.

going.⁶ It was recognized that since the neutron yield was more than two, if one of them were used to keep the reaction going, another might be used to produce plutonium to ultimately replace the U-235 consumed, effectively extending the supply of U-235. To do this, the core of the U-235 reactor was surrounded by a "blanket" of natural uranium (essentially, U-238). It was soon found to be more efficient to use a mixture of the core materials (U-235) and the blanket materials (U-238)—i.e., to use uranium only partially enriched in U-235.

Today's reactors are called "thermal" because the neutrons are *moderated* (allowed to slow down to thermal energies) to increase their ability to cause fissions. Most current reactors use water as both moderator and coolant. The water, however, competes for neutrons, so only about half the consumed U-235 gets replaced by plutonium. Several nuclear power plants (fast breeders, which do not moderate the neutrons) have been built, wherein the replacement of U-235 by plutonium can be at or above one-for-one, but these facilities have not been coupled with effective recycle technologies.

Thermal reactors using low-enrichment uranium get about 60% of their energy from burning U-235, the rest coming from fissioning of plutonium that was produced in the reactor from U-238. Overall, the current system uses natural resources very inefficiently. Nature only provides less than one percent of the uranium as U-235, and less than half of that is fissioned in today's LWRs. It would be more than 100 times as efficient if we could consume primarily U-238, rather than relying so heavily on the minor component, U-235.

And indeed, doing that becomes feasible if we abandon the idea of consuming mainly U-235 and some of the small portion of U-238 that today's reactors convert into plutonium, and look to *a system that consumes all the actinides*, including U-238 and all its heavier byproducts. Not only is this possible, the technology to construct such facilities exists.

When the neutron economy of a fast reactor with a metallic fuel containing about 79% uranium, 20% plutonium and 1% higher actinides is examined, it turns out that the energetic neutrons make it possible to consume the uranium resource so efficiently that the waste consists entirely of fission products, all other components remaining in equilibrium. Plutonium and higher actinides act very much like catalysts. The physics of the fission process requires that this system use fast (energetic, unmoderated) neutrons. Fissions

caused by energetic neutrons are more violent, releasing more free neutrons than those caused by moderated neutrons. Further, when the neutrons are energetic, actinides (uranium and transuranics) compete with fission products for neutrons more effectively. With fast neutrons, it is quite feasible to have a system wherein the catalyst is fully and continuously regenerated.

Consider an actinide-consuming system composed of a fast reactor and a pyroprocessor for recycling the fuel. Such a setup can be operated as a net generator (breeder) of plutonium, or in a break-even mode, or as a net *burner* of transuranics. In the first two of those modes, the system can operate indefinitely with only the periodic addition of makeup U-238. The burner mode requires makeup transuranic material to be added along with the U-238.

Currently it is the net-burner mode that is of interest to the policymakers who want to consume the global inventories of transuranic elements (plutonium and higher actinides) now existing in used reactor fuel and in surplus to weapons materials. Therefore, that is the type of operation envisaged in the Department of Energy's GNEP proposal,⁷ which sees plutonium as a waste product to be destroyed (in the GNEP terminology, the acronym "FBR" stands for "fast burner reactor"). For operation as a plutonium burner, perhaps 20 percent of the makeup fuel would be transuranics from used thermal-reactor fuel or from excess weapons stockpiles.

Is the actinide-consuming reactor just the old Fast Breeder under another name? Not really. While it's true that much of the technology is directly carried over from decades of experience with fast-breeder development, the entire fuel cycle is different. At no time in the fuel cycle is there need for separated plutonium. In addition, the new objective (consumption of all actinides) has led to a very different fuel type (metallic rather than oxide), leading to major improvements in safety and operability.

This scheme for consuming U-238 using a plutonium catalyst is not a wild pipe dream. Argonne National Laboratory's fast reactor in Idaho, EBR-II, used fuel similar to this for some 10 years (1984-1994). Concurrently, similar types of fuel were being tested in the Russian BN-600 fast-reactor nuclear power plant. These programs included samples of fuel actually recycled using pyroprocessing. The EBR-II fuel was a metallic composite simulating the equilibrium catalyst-empowered fuel noted above. The experiments not only showed the feasibility of the

⁶ A nuclear weapon must capture a much higher percentage of the neutrons, so as to build up the fission rate very rapidly.

⁷ GNEP: Global Nuclear Energy Partnership; see "Further Reading," below.