quick study

Reprocessing spent nuclear fuel

David Bodansky

New methods being developed for future extraction of plutonium and uranium from nuclear waste incorporate safeguards against weapons proliferation.

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The world's first large nuclear reactors were built in the US during World War II to obtain plutonium-239 for nuclear weapons. Fueled with natural uranium, they produced Pu by neutron capture in ^{238}U followed by two beta decays. The steps taken to recover the Pu and other selected components from the spent fuel constitute reprocessing.

Although reprocessing is intrinsic to a weapons program, it is optional for the commercial nuclear-power fuel cycle. The US decided in the mid-1970s against reprocessing in the commercial nuclear program, largely due to concerns about weapons proliferation. France, Japan, Russia, and others did not follow that example, and recently Congress and the US Department of Energy have shown a revived interest in reprocessing. That shift, embodied in DOE's Advanced Fuel Cycle Initiative and the more recently promulgated Global Nuclear Energy Partnership (GNEP), and the weapons ambitions of North Korea and Iran have thrust the topic more into the spotlight.

Without reprocessing, the spent fuel becomes nuclear waste. Spent-fuel nuclides fall into two main categories: actinides and fission products. The great majority of the fission products have half-lives of less than 10 years. Notable exceptions are cesium-137 and strontium-90, both with half-lives of roughly 30 years. Cesium-137 is a strong gamma-ray emitter; the resulting high radiation levels help to protect the spent fuel against theft.

The transuranic actinides present the main long-term challenge in waste handling because many have moderately long half-lives. They are produced primarily through neutron capture and radioactive decay. Chief among them is Pu, but reactors also produce significant amounts of neptunium, americium, and curium, the so-called minor actinides. The Pu—mainly ^{239}Pu, but with a large admixture of ^{240}Pu and heavier isotopes—is termed reactor-grade to distinguish it from weapons-grade, which is more than 90% ^{239}Pu. Given sufficient expertise, a bomb can be made with reactor-grade Pu although the yield may be "only" about 1 kiloton. That danger argues against a reprocessing method that creates a pure Pu stream vulnerable to theft or diversion.

The renewed US interest in reprocessing is associated with an anticipated increase in nuclear power use. With a tripling of nuclear capacity, for example, and no changes in waste handling practices, a repository with Yucca Mountain's original 70 000-tonne statutory capacity might be required every decade or so. Although constructing those repositories appears technologically and economically possible, the option is politically unattractive. Reprocessing can reduce the demands on a repository by removing U and transuranics for recycling as fuel in reactors. Such a step both greatly reduces the long-term radioactivity of the remaining waste and extends U supplies. If the Cs and Sr are also extracted, the initial heat load is reduced, which allows a potentially denser packing of the waste.

On the negative side, the spread of reprocessing could lower the barriers against the theft or diversion of Pu, and the need to handle large amounts of highly radioactive materials creates some risks. Further, reprocessing probably will add slightly to the cost of nuclear power. Building a reprocessing plant, moreover, is likely to face its own political problems.

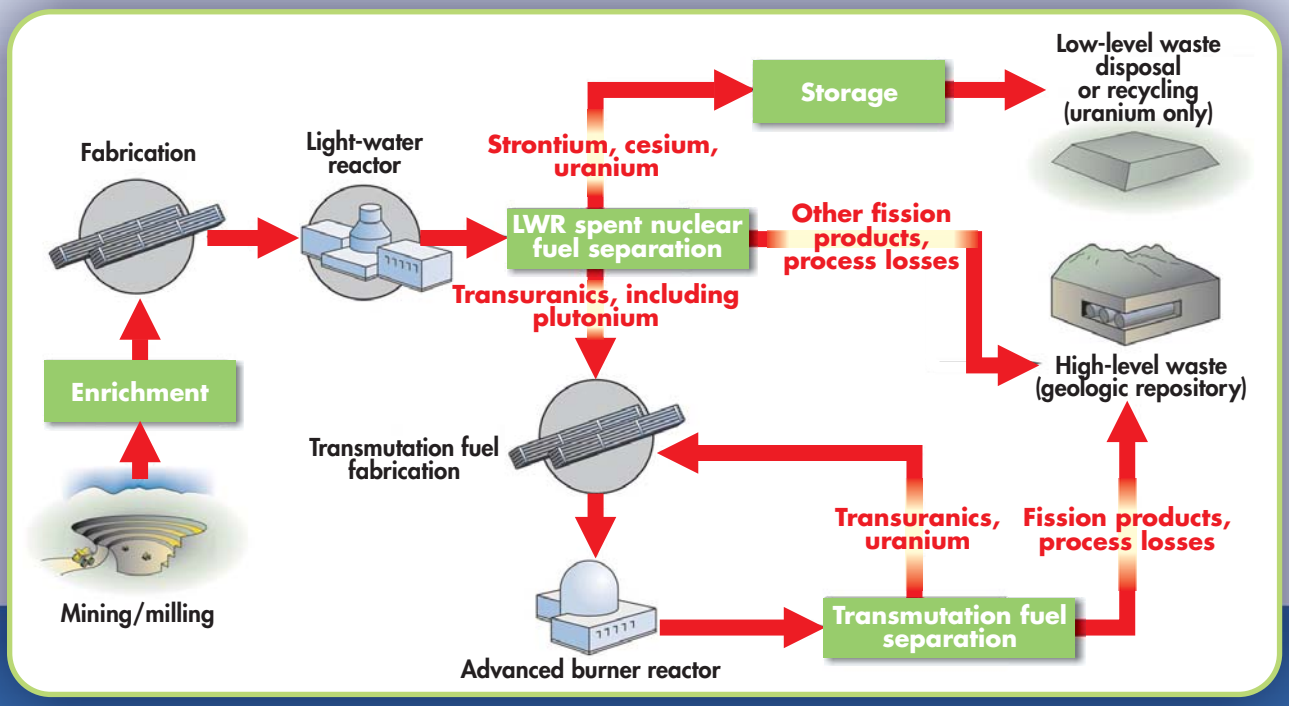
The once-through (also called open) fuel cycle, which does not include reprocessing, is currently used for US commercial nuclear power. Spent fuel is initially kept in water-filled cooling pools at the reactor site, pending eventual transfer to a central repository for interim storage or long-term disposal. No central repositories have been developed yet, and some of the cooling pools have reached their capacity. A common solution has been dry storage—moving the fuel to on-site, heavy, protective casks where convective air cooling suffices.

An American Physical Society study published in 2005 judged that dry cask storage, either at the reactor sites or at central facilities, would be "safe and affordable" for at least 50 years. Two years earlier an MIT study recommended continuing the US reliance on the once-through fuel cycle "for the next decades." The possibilities of interim storage and of long-term disposal in the Yucca Mountain repository do remove the urgency of selecting a reprocessing option, but the once-through cycle will not suffice for the long-term, large-scale use of nuclear energy.

Aqueous reprocessing

To date, the dominant reprocessing method has been the PUREX (plutonium and uranium recovery by extraction) process, which has been used in the US and other weapons programs and in commercial programs abroad. It is an aqueous process in which nitric acid dissolves the spent fuel. The U and Pu are extracted and the remaining constituents—the fission products and minor actinides—are left as wastes. The separated Pu, which goes into new fuel elements, is in its own product stream.

A different aqueous separation process under development in the US, the UREX+ (uranium extraction) process, is intended as a proliferation-resistant alternative to PUREX. The GNEP, which seeks to further nuclear power in the US and worldwide, has adopted UREX+ development as one of its major goals. Again, acid dissolves the fuel. The U is extracted for disposal as low-level waste or use in fuel, and Cs



Candidate fuel cycle system for the US Department of Energy's Global Nuclear Energy Partnership program. Transuranics from reprocessed light-water reactor spent fuel are fabricated into fuel elements—possibly using UREX+ (uranium extraction) reprocessing—for use in a fast burner reactor. Transuranics and uranium are separated—possibly using pyrochemical processing—from the burner reactor's spent fuel and are fabricated into new fuel elements for repeated burner cycles. (Adapted from DOE's *Report to Congress: Spent Nuclear Fuel Recycling Program Plan* [May 2006], fig. 3.)

and Sr may also be removed. The remainder is separated into the fission products, which become the high-level wastes, and the Pu and other transuranics, which can be incorporated in new reactor fuel. The minor actinides are kept with the Pu, raising the heat output and radioactive emission rates of the Pu product stream.

In its January 2003 report to Congress on the Advanced Fuel Cycle Initiative, DOE suggests that the effects of the minor actinides make the Pu "unusable for weapons applications" and, in weaker words, "relatively unattractive to potential proliferaters." Other assessments vary. Some analysts say that a 1-kiloton weapon could be made, but others conclude that a bomb is not feasible. Of course, as the difficulty of making a Pu bomb increases, bomb aspirants may turn to an easier route using enriched U.

The pyrochemical process

Another proliferation-resistant method, pyrochemical reprocessing, was a key part of the Integral Fast Reactor program. Originally developed at Argonne National Laboratory, the IFR program was terminated in the mid-1990s. In recent years DOE has revived support for aspects of the program, including the development of the pyrochemical process and new fast reactors. As Pu breeders, fast reactors offer the prospect of an almost unlimited supply of energy. With a different arrangement of the fuel, they can take advantage of the relatively high fast-neutron cross sections for actinide fission and serve effectively as actinide burners.

The pyrochemical process is particularly suited for use with the metallic fuel that the Argonne fast reactor program has favored, although it could be adapted for use with other fuels. The spent fuel is fed into a bath of chloride salts at a temperature high enough to melt the fuel. Electrorefining separates the spent fuel into three streams as in the UREX+ process: U, transuranics, and fission products. Proliferation resistance is achieved by having the fuel-cycle facilities in close proximity and by keeping the minor actinides with the Pu.

The immediate future

The US is probably at least a few decades away from the large-scale implementation of either UREX+ or pyrochemical processing. The figure shows a possible fuel cycle that uses both methods. In early 2006 DOE anticipated that within 20–25 years a commercial-scale demonstration of a fuel cycle with reprocessing would be under way. Later in the year, it began exploring a faster UREX+ schedule, a move that elicited warnings against premature decisions.

A prolonged interval before reprocessing begins need not inhibit the revival of US reactor construction if there is a consensus that satisfactory near- and long-term waste disposal solutions exist. The long-term solutions may include the permanent disposal of light-water-reactor spent fuel without recycling and rely on, for example, Yucca Mountain with an expanded capacity; other excavated geological sites; deep boreholes; or sub-seabed disposal, a now taboo option. Still, the development of recycling technologies remains important so that the US will be prepared should it opt for a major sustained increase in its reliance on nuclear power.

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Additional resources

▶ D. Bodansky, *Nuclear Energy: Principles, Practices, and Prospects*, 2nd ed., Springer, New York (2004).
▶ W. H. Hannum, G. E. Marsh, G. S. Stanford, *Physics and Society* **33**(3), 8 (July 2004); see http://www.aps.org/units/fps.
▶ R. Vandenbosch, S. E. Vandenbosch, *Physics and Society* **35**(3), 7 (July 2006); see http://www.aps.org/units/fps. ■

The online version of this Quick Study has further resources, substantiating references, and explanatory material.