Domestic and International Experience With Spent Fuel Recycling

Domestic Experience

The DOE and its predecessor agencies operated several facilities that reprocessed spent fuel for the recovery of materials for defense, nuclear energy, and space programs. Plutonium was the main element recovered. Neptunium, americium, tritium, cesium, and strontium were also recovered on a significant scale. In excess of 100,000 metric tons of heavy metal (MTHM) were reprocessed at Hanford, Savannah River, and Idaho over a period of more than 40 years. Most of the spent fuel consisted of relatively small, metallic elements with a low burnup (usually less than 2,000 megawatt-days per metric ton of heavy metal (MWD/MTHM). The principal technology used was a solvent extraction technique known as the plutonium and uranium extraction (PUREX) process (and its variants). Pilot-scale facilities have used pyrochemical, metal, and eletrometallurgical technologies for reprocessing and recycling. DOE's high-level waste (HLW) is a result of these reprocessing activities.

There is limited domestic experience with commercial reprocessing and recycling. The Atomic Energy Commission (AEC) encouraged private organizations to become involved in reprocessing in the 1960s. The West Valley facility operated in the late 1960s and early 1970s, using the PUREX process. The facility nominally had a 300 MTHM/yr capacity and reprocessed about 650 MTHM. Approximately 60% of this material was metal fuel from the Hanford N-Reactor with a relatively low burnup. The remainder was oxide fuel—the highest burnup was around 20,000 MWD/MTHM. The facility also performed a demonstration on thorium spent fuel. West Valley operations generally met regulatory requirements, although exposures were not as low as reasonably achievable (ALARA) and radiation protection was a significant problem. The operator planned an expansion of West Valley to quadruple its capacity. Seismic issues were raised as part of the regulatory review and these issues increased the estimated costs by over an order of magnitude. Based on the increased costs and the potential for significant competition from other companies, the operator decided to cease operations.

GE designed and built a reprocessing facility in Morris, IL, utilizing a dry process for the main separations. The process relied on the volatility of uranium hexafluoride and was successfully demonstrated in the laboratory. Pre-operational testing at the constructed facility was not as successful and would have required major renovations. Given the projected costs and competitive reprocessing market, and increasing regulatory scrutiny (from the West Valley seismic reviews and the required safeguards), the operator decided not to pursue reprocessing at the facility. It is currently used as an independent spent fuel storage installation (ISFSI) for wet storage of commercial spent fuel.

The AGNS consortium constructed a third facility adjacent to the Savannah River Site in Barnwell, South Carolina. This facility utilized advanced PUREX technology for a planned capacity of 1,500 MTHM per year. The facility conducted uranium testing but never operated due to President Carter's decision to indefinitely defer commercial spent fuel reprocessing. The facility is currently undergoing decommissioning. Altogether, private industry invested approximately \$2 billion in the Morris and AGNS facilities, however, neither facility began reprocessing operations. Other companies were planning for reprocessing and recycle facilities. Exxon planned a 1,500 MTHM/yr facility at Oak Ridge, TN, and Westinghouse planned a Recycle Fuels Plant for approximately 600 MTHM/yr of mixed-oxide (MOX) fuel fabrication. These plans were shelved in the late 1970s and early 1980s.

All of these facilities were based upon a burnup of 30,000 MWD/MTHM typically used for spent fuel in the early 1970s. Utilities in the United States currently have about 45,000 MTHM in spent fuel, with an average burnup around 45,000 MWD/MTHM. Current spent nuclear fuel (SNF) discharges are around 55,000-60,000 MWD/MTHM burnup. Some pressurized water reactor (PWR) fuels are currently licensed for a 62,000 MWD/MTHM burnup at some sites. The maximum burnup currently licensed for a dry storage cask is 65,000 MWD/MTHM.

International Experience

Reprocessing is conducted on a significant scale in France, the United Kingdom, Japan, Russia, India, and China. Other countries (e.g., Belgium and Germany) have conducted pilot activities. As in the United States, reprocessing started in support of defense and nuclear energy programs, primarily using low-burnup metallic fuels. Subsequently, several large facilities have evolved providing commercial reprocessing and recycling services across national boundaries. The commercial reprocessing facilities are based on a nominal design capacity of 800 MTHM/yr and medium burnup (circa 40,000 MWD/MTHM) of oxide fuels, using optimized PUREX solvent extraction. To date, commercial operations have generally been economic and within regulatory requirements. Doses and discharges have decreased considerably from the late 1970s/early 1980s and now appear to have plateaued. Current trends indicate a decrease in reprocessing across national boundaries due to the startup of a new reprocessing plant in Japan, higher fuel burnups, more spent fuel storage (particularly dry storage), planned nuclear phaseouts in Germany and Sweden, limited new orders for reactors, and uncertain future plans. Separated materials (plutonium) are returned to the country of origin as MOX fuel. Commercial reprocessing facilities have indicated plans to return an amount of vitrified HLW equivalent to all the wastes generated from reprocessing a specific country's spent fuel back to the country of origin. Some vitrified HLW shipments have already been made to Belgium, Germany, and Japan.

France has two large reprocessing plants at the La Hague site, on its northern coast. The facilities are very large, occupying a space approximately 1.5 miles long by 0.75 mile wide, as shown in Figure 1. The UP-2 facility reprocesses domestic fuel for the French PWR fleet. Typical throughputs are 600-800 MTHM/yr. The French utility is intending to increase discharge burnups to approximately 50,000 MWD/MTHM. The UP-3 (sometimes called UP-3A) facility reprocesses spent fuel from PWRs and boiling-water reactors [BWRs]) for overseas customers, including Japan, Germany, and Belgium. The facility cost between \$3 and 4 billion (1990 dollars) and was financed by international contracts. An additional, UP-3B facility was planned but not pursued due to the cancellation of many reactor orders in the 1980s and early 1990s.

UP-2 and UP-3A recover uranium and plutonium. Both are recycled—the plutonium in MOX fuel. Currently, the transuranics are sent with the fission products to onsite HLW vitrification facilities. Approximately 3 gigacuries of vitrified HLW canisters are in dry storage at the site (for comparison, the Hanford HLW tanks currently contain about 250 megacuries). Current French operations reduce the volume of material requiring a repository by approximately a factor of 6 compared to the estimated volume for direct disposal of the fuel.



Figure 1. La Hague Plants - Two 800 MTHM/yr plants

France operates a separate facility (MELOX, in southern France) for the manufacture of MOX fuel. MELOX has a nominal capacity of 200 MTHM/yr. In France, MOX fuel is irradiated to a burnup of 42,000 MWD/MTHM; the plan is to increase this burnup to approximately 50,000 (i.e., comparable to UO_2 fuel). MELOX also produces MOX fuel for overseas customers. France has reprocessed commercial spent MOX fuel through the UP-2 plant (primarily once through but there have been several tests with twice irradiated MOX fuel). France has conducted laboratory tests on americium and curium recycle, and has irradiated several assemblies.

French authorities and organizations have found reprocessing and MOX to be economic as waste management strategies but not as fuel management alternatives. French analyses have shown americium recycle to reduce repository dose impacts by a factor of about 40 and curium recycle to reduce dose impacts by two orders of magnitude. However, the need for a repository is not eliminated.

The United Kingdom (UK) operates several reprocessing facilities at Sellafield (Windscale) on the Northwest coast. These facilities reprocess low-burnup metallic fuels (approximately 6,000 MWD/MTHM from Magnox reactors) and medium-burnup oxide fuels (from advanced gas reactors [AGR] and light-water reactors [LWRs]). The B205 facility has a relatively large capacity and is used for the metallic spent fuel from Magnox reactors; Magnox reactors are approaching decommissioning, so the operations at B205 may cease in the next 10 years. The THORP facility reprocesses commercial oxide spent fuel. The facility has a nominal design capacity of 800 MTHM/yr and has been entirely financed by overseas sales contracts. The

THORP facility is the main facility of interest in the UK (the UK currently only has one LWR, and AGR fuel is now frequently stored dry). THORP has a vitrification plant that also processes HLW from Magnox spent fuel activities. Operationally, vitrification has experienced melter problems but the facility currently has some 1.5 gigacuries of vitrified HLW in dry storage. The overseas contracts at THORP expire around 2010 and there are no current plans to extend operation beyond that time.

The UK has approximately 100 tons of separated plutonium in storage from the reprocessing operations. The country is evaluating options for this material and is also reevaluating its energy options, including nuclear energy. Sellafield includes a separate MOX plant (SMP) for returning plutonium as MOX fuel to the country of origin. UK analyses of transuranic recycling revealed similar results to the French work. Only limited testing has been conducted to date.

Commercial reprocessing will begin soon at the Rokkasho-mura plant in Japan. The Rokkasho plant has a nominal design capacity of 800 MTHM/yr and was constructed at a cost exceeding \$6 Billion (in 2005 dollars). The plant is undergoing uranium testing in 2006. It is designed for the production of a mixed uranium-plutonium product that can be used to produce MOX fuel for recycle in Japanese LWRs. Japan's intention is to recycle materials as much as possible and, ultimately, to use fast neutron reactors both for energy and to manage HLW.





Detailed Description of Limited Recycle and Full Recycle

The staff expects DOE to consider two approaches for its spent fuel recycling program that would achieve the goal of effectively increasing the utilization of repository space in a proliferation-resistant manner. The first approach would involve (1) an advanced separations step (based on the UREX+ technology, which mitigates the disadvantages of the more common PUREX process¹) and (2) a limited recycle of mixed-oxide (MOX) fuel in conventional LWRs and/or Advanced Burner Reactors (ABRs) such as a liquid metal fast flux reactor. The second approach would involve full recycle beyond these two primary steps, using ABRs to close the fuel cycle and transmute the transuranic waste streams. A process flow diagram for the stages included in limited and full recycling and the fate of the products separated from the spent nuclear fuel is included in Enclosure 2. A summary of the processes involved is discussed below.

Limited Recycle

The primary stages of limited recycling include spent fuel reprocessing and limited recycle in existing LWRs. The UREX+ technology is a refined solvent extraction technology that allows the separation and subsequent handling of several highly pure product streams: (1) uranium, which can be stored for future use or disposed of as low-level waste, (2) a mixture of plutonium and neptunium, which can be reused as MOX fuel, (3) separated fission products that would eventually require long-term storage and disposal, and (4) the transuranic elements americium and curium. Several of the processes associated with the UREX+ technology are discussed further in the next section (on full recycle).

The performance goals of the UREX+ process are to achieve the following:

- Purity levels of uranium, plutonium, and neptunium sufficient to meet the MOX fuel specifications in ASTM C833-01
- Recovery of the fission products technetium and iodine to levels sufficient to achieve up to a 20-fold decrease in offsite dose, with sufficient separation of fissile actinides to allow future transmutation
- Recovery of the fission products cesium and strontium to a level sufficient to reduce their contribution to the heat load in the repository equal to the heat load of all other fission products and to remove sufficient transuranic content to allow decay storage and ultimate disposal as low-level waste
- The separation of americium and curium to levels that result in a 100-fold reduction of the heat load to the repository
- Produce final raffinate streams containing the rare earths and all soluble fission

¹PUREX stands for <u>p</u>lutonium <u>u</u>ranium <u>r</u>eduction and <u>ex</u>traction, and UREX+ stands for <u>u</u>ranium <u>r</u>eduction and <u>ex</u>traction. Both processes use liquid-liquid solvent extraction techniques. The PUREX process separates a stream of plutonium and a stream of uranium from the waste stream containing both transuranics and fission products, while the UREX+ process separates a mixed uranium-plutonium stream from a transuranic stream and fission product streams.

products (except cesium, strontium, technetium and iodine), which can then be converted into a solid form for final disposal in the repository

The advantages of the UREX+ process over the PUREX process are the potential for significant cost reductions, the elimination of the need for waste tank farms, and the ability to separate and manage very pure product streams of key elemental and isotopic constituents.

The fuel stream, comprising uranium, plutonium, and neptunium, accounts for about 94% of the total spent fuel mass and 15% of the initial and 70% of the long-term repository heat load. The transuranic elements account for less than 1% of the total mass and 20% of the initial and 30% of the long-term heat loads. The fission products make up 5% of the mass and 65% of the initial and negligible long-term heat loads.

As shown on the process flowchart in Enclosure 2, the resulting streams from the UREX+ process follow several different paths. In addition to the UREX+ facility, a new fuel fabrication facility would need to be built to handle the uranium and the mixed plutonium/neptunium UREX+ output streams. The output from the new fuel fabrication facility would be recycled as MOX fuel, resulting in the limited recycle of the spent fuel.

Some of the benefits of limited recycling would depend on when the material is recycled. For example, recycling spent fuel within the first several years of removal from the reactor would significantly limit the buildup of americium-241 from plutonium-241 decay. Limiting the buildup of americium-241 is desirable because the isotope's energetic alpha decay and relatively short half-life result in a high heat output. The presence of plutonium-241 in the spent fuel, however, would require greater shielding in the limited recycling facilities than americium-241. As shown on the process flowchart in Enclosure 2, recycling the uranium, neptunium, and plutonium stream has the potential to reduce the long-term repository heat load by 70 percent.

Interim storage may be needed for several of the UREX+ output streams, especially the transuranic elements americium and curium. The length of time the interim storage phase would need to last before the final processing of the transuranic actinides and fission products is unclear. While it is likely that the storage facility would be co-located with the UREX+ facility, some process streams might be stored at another location. The remaining fission products could be separated into a stream for short-term storage (to reduce the heat load) and a stream for long-term storage in specialized waste forms.

Full Recycle

Full recycling would include the primary stages of limited recycling but would close the fuel cycle loop by using ABRs (and/or possibly linear accelerators) to transmute the fuel constituents into much less hazardous elements and by using pyroprocessing technologies to recycle the fast flux reactor fuel. Full recycle has the potential to significantly reduce proliferation risk by eliminating the buildup of all isotopes.

In addition to the reprocessing and limited recycling of spent fuel, full recycling would involve transmutation of the transuranic elements. Transmutation occurs in the high flux field typically associated with a liquid metal cooled fast flux reactor. DOE currently has two potential technologies from which to choose: the Integral Fast Reactor (IFR), which uses metal fuel, and the Advance Liquid Metal Reactor (ALMR, also known as the GE Power Reactor, Innovative, Small Module [PRISM]), which can use either ceramic fuel or metal fuel.

The IFR system developed by Argonnne National Laboratory would use a new type of metal

alloy fuel. This fuel would be recycled using a pyroprocess whereby uranium, plutonium, and other transuranic elements could be separated from the other radioactive waste and reused in new fuel assemblies. The IFR design also has the potential to burn actinides from LWRs. This would require a separate aqueous reprocessing facility to be built (i.e., the UREX+ facility). The remaining waste would have a significantly reduced heat load and volume and would remain a radioactive hazard for only hundreds of years, not for hundreds of thousands of years. As shown on the process flowchart in Enclosure 2, the transuranic elements contribute approximately 30 percent of the long-term repository heat load (integrated over time). With full recycling, most of the transuranic elements would be transmuted and two-thirds of their heat load could be reduced. Therefore, full recycling has the potential to achieve an overall 90 percent reduction in the total long-term repository heat load.

The ALMR would involve much the same fast flux reactor technology as the IFR, however, the primary difference is its ability to use ceramic or metal fuel. DOE may prefer this technology since ceramic fuel is currently more commonly used than metal fuel.

The remaining fission products from the reprocessing of the spent LWR fuel would be separated into streams for short-term storage (heat load reduction), possible transmutation, and long-term storage in specialized waste forms.