

## **DEVELOPMENT OF SEPARATIONS TECHNOLOGIES IN THE U.S. PARTITIONING AND TRANSMUTATION PROGRAMME**

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### **Introduction**

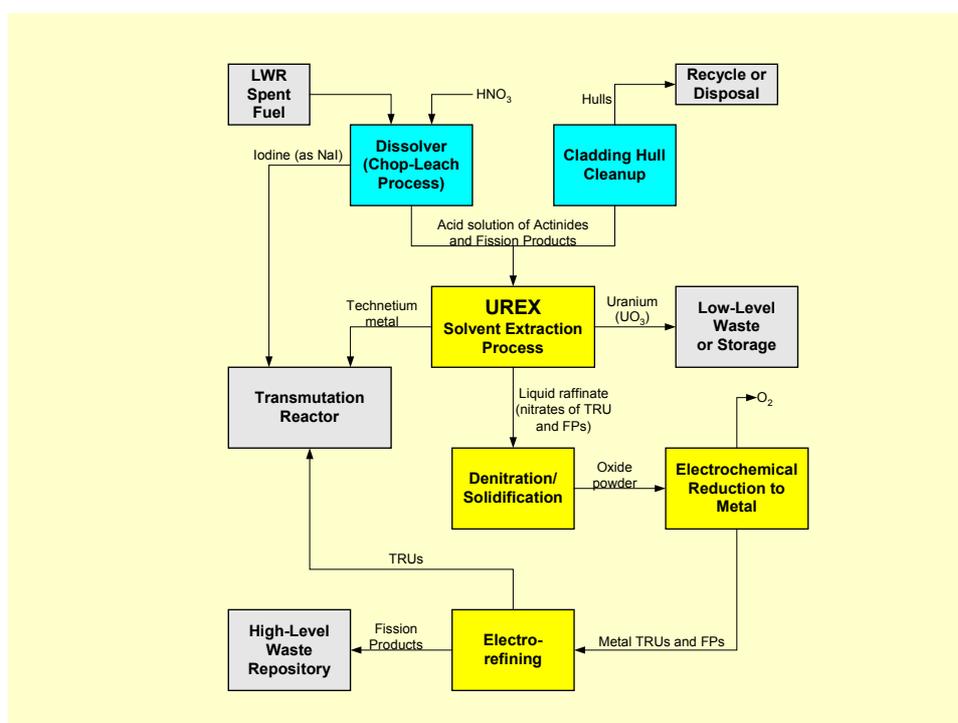
There are 103 commercial nuclear power stations currently operating in the United States, generating a total of about 2 000 t of spent fuel each year. Over 40 000 t of spent fuel have accumulated since these plants began operation, with the fuel being stored first in at-reactor spent fuel storage pools and later in independent spent fuel storage installations and dry cask storage facilities. The Congress of the United States recently authorised the construction of a deep geologic repository for the storage of high-level radioactive waste at the Yucca Mountain site in the state of Nevada. The total cost of this repository, including construction, waste emplacement and closure, is approximately USD 58 billion. The Nuclear Waste Policy Act as amended in 1987 limits the Yucca Mountain repository to the storage of 63 000 t of spent nuclear fuel and 7 000 t of other high-level wastes, although the practical capacity of the site is estimated to be about 120 000 t of spent fuel. At the current rate of spent fuel generation, the U.S. inventory of spent fuel will reach the 63 000 t level at about the same time that the repository is prepared to begin receiving spent fuel. Accordingly, it is necessary to take actions that will limit the rate of accumulation of spent fuel while permitting the expansion of nuclear power, a course of action that may be an essential element of a national strategy for energy supply in the 21<sup>st</sup> century. The alternative is to being the search for a second repository site in the very near future; this option faces significant political obstacles.

### **Evolution of the U.S. programme for partitioning and transmutation**

The evaluation of the partitioning and transmutation alternative to the direct disposal of spent nuclear fuel began in the U.S. in 1999 with the development of a “roadmap” for the transmutation of nuclear wastes in accelerator-driven systems. This evolved into the Accelerator Transmutation of Waste (ATW) programme that was based on the partitioning of spent light water reactor (LWR) spent fuel to produce four separate product streams: uranium, transuranics, long-lived fission products (Tc and I), and other fission products. The chemical separations system, as shown in Figure 1, was a hybrid aqueous-pyrochemical process that used a modified solvent extraction process called UREX to separate the uranium from the transuranics while recovering a separate technetium stream. The pyrochemical process treated the denitrated UREX raffinate, initially with an electrochemical method to reduce the oxides to the metallic state, and then with a molten-salt electrorefining method to separate the transuranics from the fission products. The uranium was to be stored in pure form for future re-use or for disposal as a low-level waste form. The long-lived fission products were destined for transmutation to stable isotopes by neutron absorption, and the transuranics were to be fissioned in

the fast neutron spectrum produced by spallation neutrons generated by the collision of high-energy protons with a liquid lead-bismuth target. Only the fission products other than Tc and I were to be sent to the repository for disposal, because the performance analysis for the repository showed that the principal contributors to the offsite dose were  $^{99}\text{Tc}$ ,  $^{129}\text{I}$  and  $^{237}\text{Np}$ , with Tc and I being dominant contributors over the first 10 000 years. The challenge of developing an accelerator-driven system with sufficient operating reliability to serve as an electricity generator (for the purpose of offsetting the capital and operating costs of the system) led to the consideration of critical fast reactors as the transmutation device. The number of the fast-spectrum devices needed to support the spent fuel generation from 100 GWe LWR operation was considered, however, to be rather prohibitive although a contemporary OECD-NEA working group report [1] showed that the economics of certain systems would be reasonable.

Figure 1. **Hybrid aqueous-pyrochemical process (UREX/PYRO-A) for partitioning of spent LWR oxide fuel**



In 2001, the ATW programme became the Advanced Accelerator Applications (AAA) programme and increasing emphasis was placed on multi-tier systems; i.e., transmutation of certain actinides in thermal-spectrum reactors and complete incineration of the remaining transuranics in fast-spectrum devices (critical or accelerator-driven). This change was made as a practical matter, recognising that a large number of commercial LWRs were already in existence, and that the deployment of the requisite fast-spectrum devices could not be accomplished overnight. The obvious choice for transmutation of transuranics is to recycle the LWR plutonium as mixed-oxide (MOX) fuel in LWRs that are licensed to accept such fuel, sending the minor actinides to the fast-spectrum tier. This, however, raises the difficult political issue of the separation of plutonium in a commercial system, a practice that has been forbidden by official U.S. policy since the mid-1970s because it implies U.S. endorsement of a technology that could lead to the proliferation of nuclear weapons. A number of studies have been made of methods for incorporation of transuranics in LWR fuel without

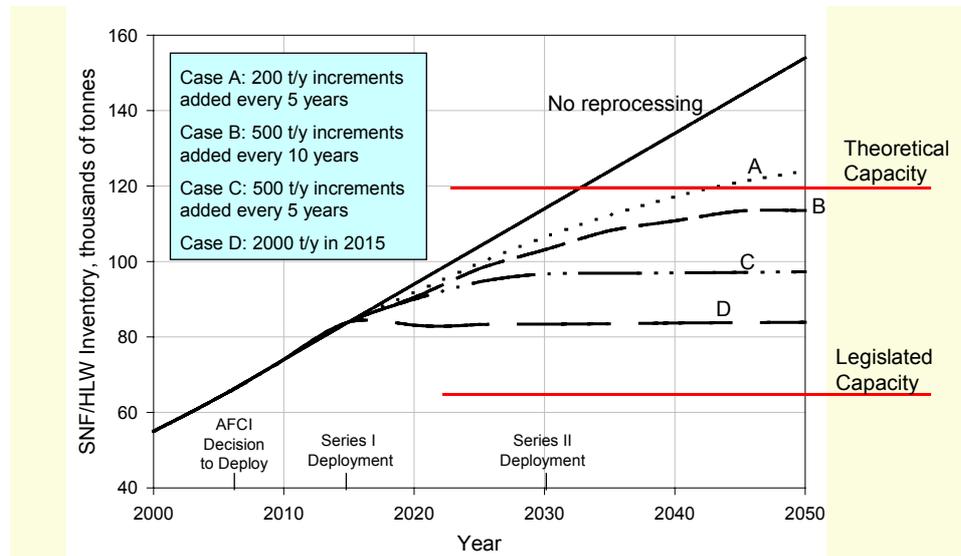
the separation of plutonium, and it is obvious that the presence of the minor actinides severely degrades the neutronic performance of the fuel. Clearly, the search for a technical solution to this dilemma is of high priority, and it will be imperative to consider all aspects of the system to ensure that its deployment is both technically correct and responsive to non-proliferation concerns.

The AAA programme has now evolved into the Advanced Fuel Cycle (AFC) programme, a forward-looking programme that will focus on the appropriate means for management of LWR spent fuel and facilitate the transition to advanced nuclear energy systems, including a strong tie to the Generation IV systems. The AFC programme is set forth in a report to the Congress of the United States that is expected to be issued by the U.S. Department of Energy in the last quarter of 2002.

### **Partitioning and transmutation in support of the advanced fuel cycle**

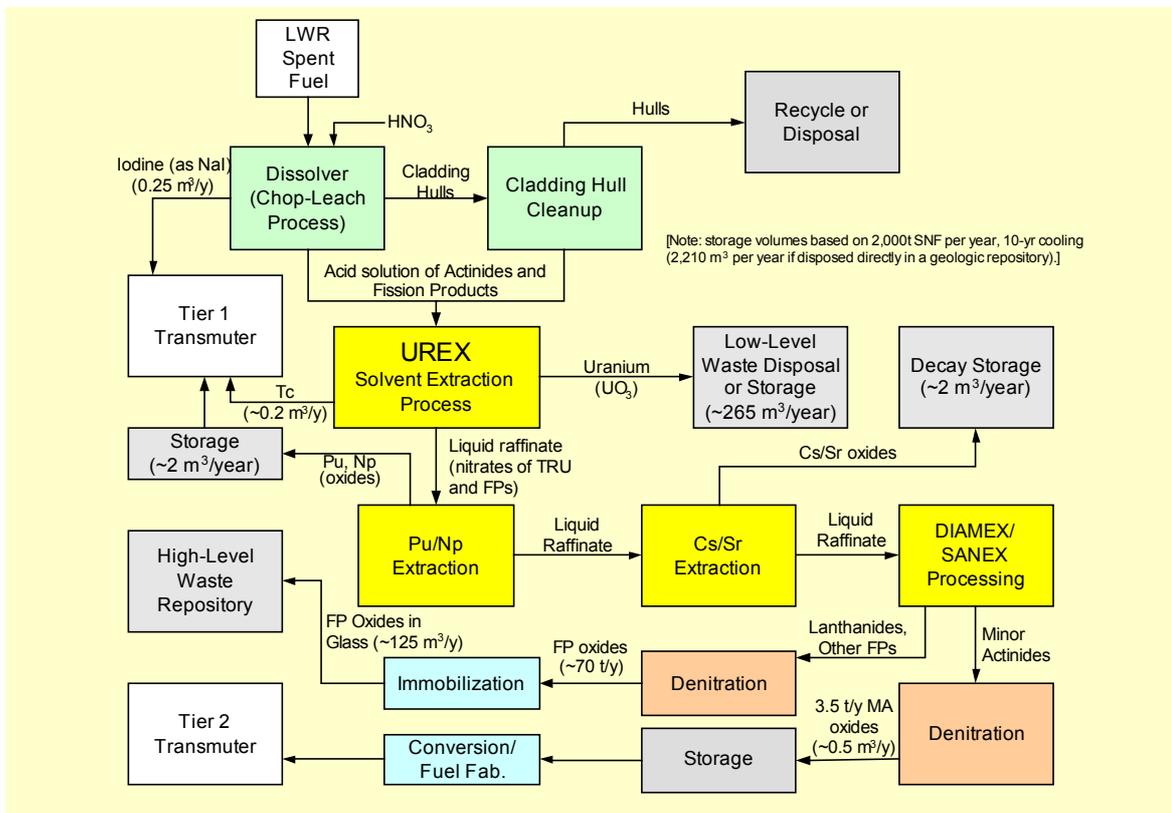
The AFC programme seeks to have a profound impact on the management of spent nuclear fuel, by eliminating the need for a second geologic repository for high-level waste disposal. This will be done by processing the spent fuel being generated by existing commercial nuclear power plants, beginning as soon as 2015. Depending upon the rate of deployment of spent fuel processing plants and their scale, it will certainly be possible to limit the amount of material requiring geologic disposal to a level well within the capacity of the Yucca Mountain repository. As seen in Figure 2, various deployment options can be successful in limiting the total inventory of spent fuel and high-level waste to less than the 120 000 t practical capacity limit for Yucca Mountain. This can be done by means of a staged processing system, one example of which is illustrated in Figure 3. In this system, spent fuel is treated by an aqueous solvent extraction process (UREX+) that recovers the long-lived fission products (Tc and I) and separates pure uranium. The removal of uranium as a pure product that meets current U.S. standards for Class C low-level waste means that this material can be disposed at very low cost or stored in a non-shielded facility for possible future use. It also means that subsequent processing can be done with comparatively small masses of material. The next step of the continuous UREX+ process is the removal of plutonium and neptunium for recycle to the thermal-spectrum reactors (Tier 1), either LWRs or gas-cooled reactors. Temporary storage of the separated (Pu+Np) may be necessary until the reactors are at a position to accept the mixed-oxide fuel; the annual generation of these elements would be equivalent to a 25% core loading of the 60 or so current reactors that could be licensed for MOX fuel. After (Pu+Np) extraction, the next UREX+ step would involve the option for the removal of cesium and strontium. Such removal would, in combination with subsequent extraction of the minor actinides, virtually eliminate heat load as a factor in design of the repository and its components. For example, forced ventilation of the Yucca Mountain repository, which has been planned for the first 75 years or so, would no longer be required. Of course, it would be necessary to store the separated Cs and Sr in a facility with heat removal capability for several decades; the build-up of heat-generating Cs and Sr over 20 years of operation at 2 000 t per year would amount to some 8 megawatts thermal energy. The final step in the UREX+ process, minor actinide extraction, would direct americium and curium to a storage facility, probably in the form of oxides, until fast spectrum reactors are available to fission these elements. Shielded storage, perhaps with active cooling, would be necessary for the Am and Cm, although the volume would be rather small, on the order of 0.25 litre per tonne of LWR spent fuel processed.

Figure 2. Effectiveness of various processing plant deployment options in limiting the total inventory of spent fuel and high-level waste to the practical capacity of the Yucca Mountain geologic repository



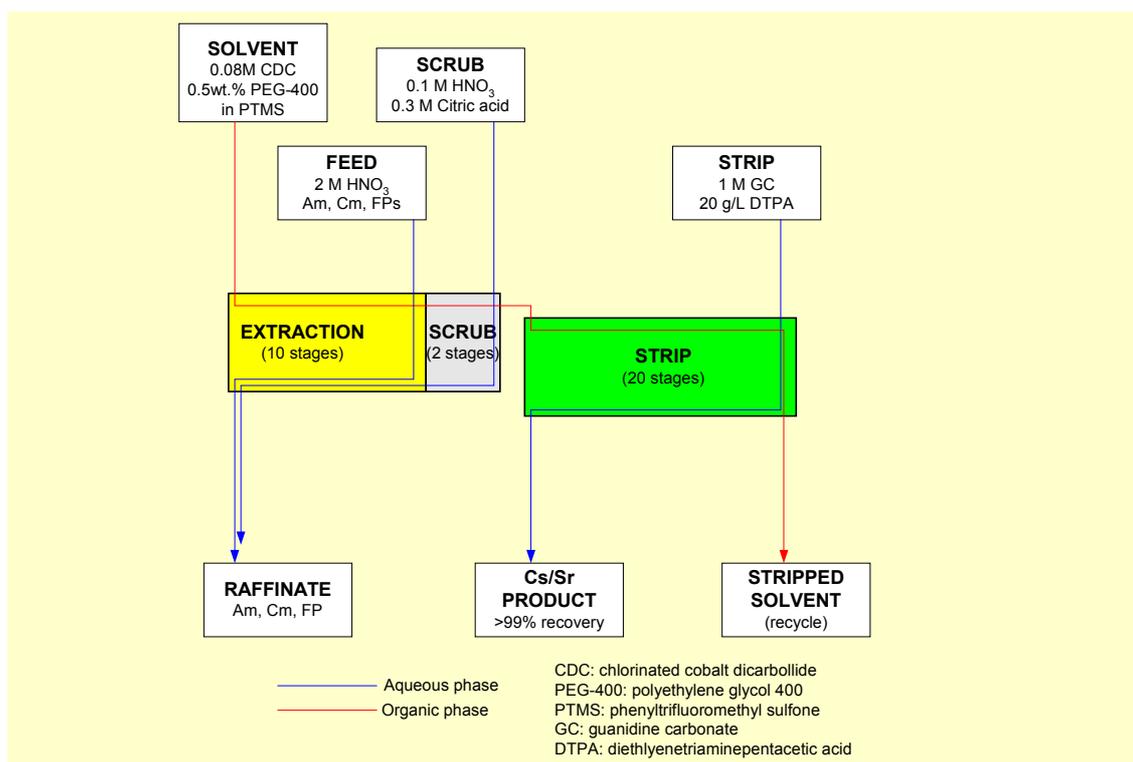
Cases A, B, and C illustrate the serial deployment of smaller-scale plants, while Case D is for a very large plant with 2 000 t per year processing capacity.

Figure 3. An example of a strategy for staged transmutation of the constituents of spent LWR oxide fuel



This process, referred to as the “UREX+” process, can greatly reduce the demands placed on the Yucca Mountain repository, by eliminating (1) those radionuclides that contribute to dose to individuals near the site; (2) those radionuclides that generate significant heat by radioactive decay, both in the near-term and the long-term; and (3) the most radiotoxic of the constituents of spent nuclear fuel. This flowsheet is an all-aqueous process; alternative pyrochemical and hybrid processes are also being considered in the U.S. programme.

Figure 4. **Illustration of a possible flowsheet for the extraction of cesium and strontium in the UREX+ process**

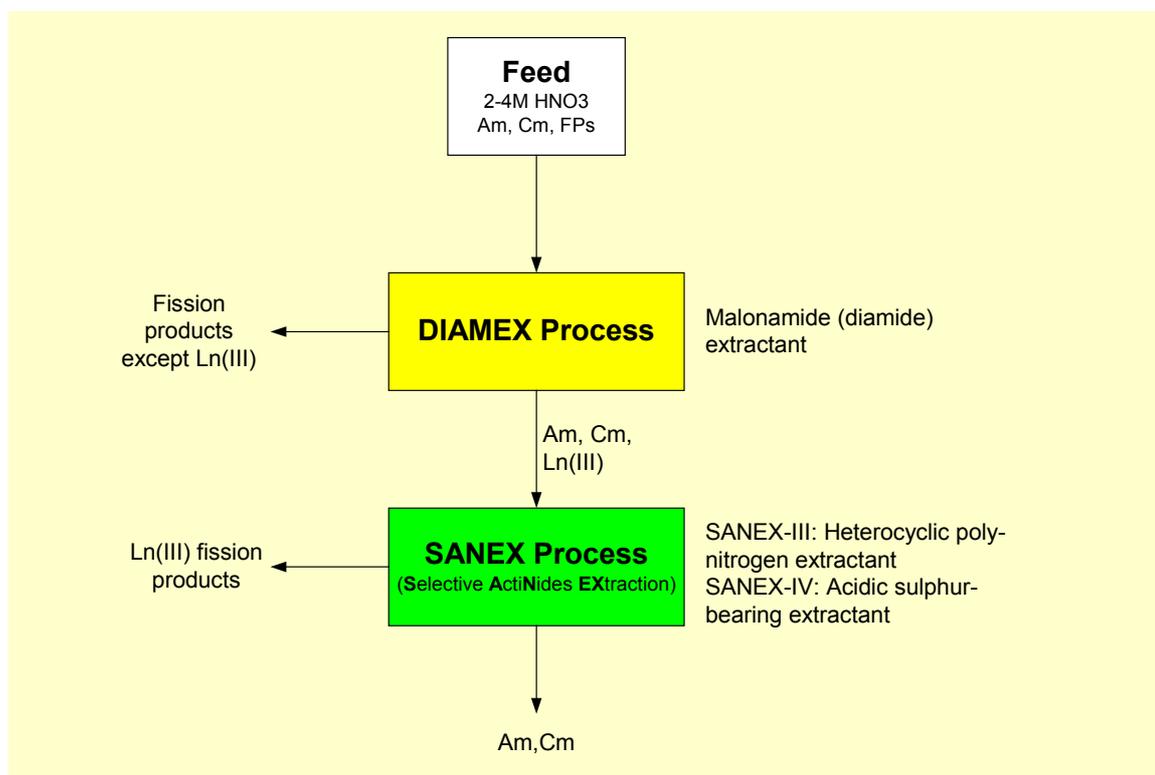


## Processing options

The original UREX process developed in the U.S. programme incorporates the use of a complexant/reductant, acetohydroxamic acid (AHA), to suppress the extraction of plutonium in the first extraction cycle. The AHA also tends to promote the extraction of technetium with uranium. After the uranium and technetium have been extracted, it is quite simple to strip out the technetium, leaving a highly-purified uranium stream. Technetium is to be converted to a metallic form by a process now under development, and can be prepared as a target for transmutation or incorporated into a metallic high-level waste form from which it is released at a rate at least two orders of magnitude lower than in the case of a borosilicate glass waste form. The UREX raffinate containing transuranics and fission products can be denitrated and then sent to a pyrochemical process that involves a reduction of the TRU and fission product oxides to metal and separation of the transuranics by electrorefining. This process, known as PYRO-A, may be useful in providing a compact, simple means for decontamination of the transuranics from lanthanide fission products. Alternatively, the metallic transuranics could be recovered and stored until fast-spectrum reactors are available for fissioning of the un-separated TRU product. The PYRO-A metallic product could also be sent to a downstream aqueous process that could separate plutonium and the minor actinides in accordance with the nuclear energy strategy in effect at the time.

If the UREX+ process is used, the raffinate from the first extraction cycle would be processed by a simple PUREX-like step (Figure 3) to extract plutonium, perhaps together with neptunium, using tributyl phosphate as the extractant. The raffinate from this step, containing americium and curium together with the remaining fission products, could then be treated by a process step much like the UNEX process, with the extractant being changed to (for example) a chlorinated cobalt dicarbollide with polyethylene glycol. As shown in Figure 4, this step will remove the cesium and strontium. The form in which the cesium and strontium are to be placed before storage is under study at this time. After cesium/strontium removal, the minor actinides (Am and Cm) would be extracted by a SANEX-type process (Figure 5).

Figure 5. **The DIAMEX/SANEX process, among the options being considered in the U.S. programme for the extraction of americium and curium**



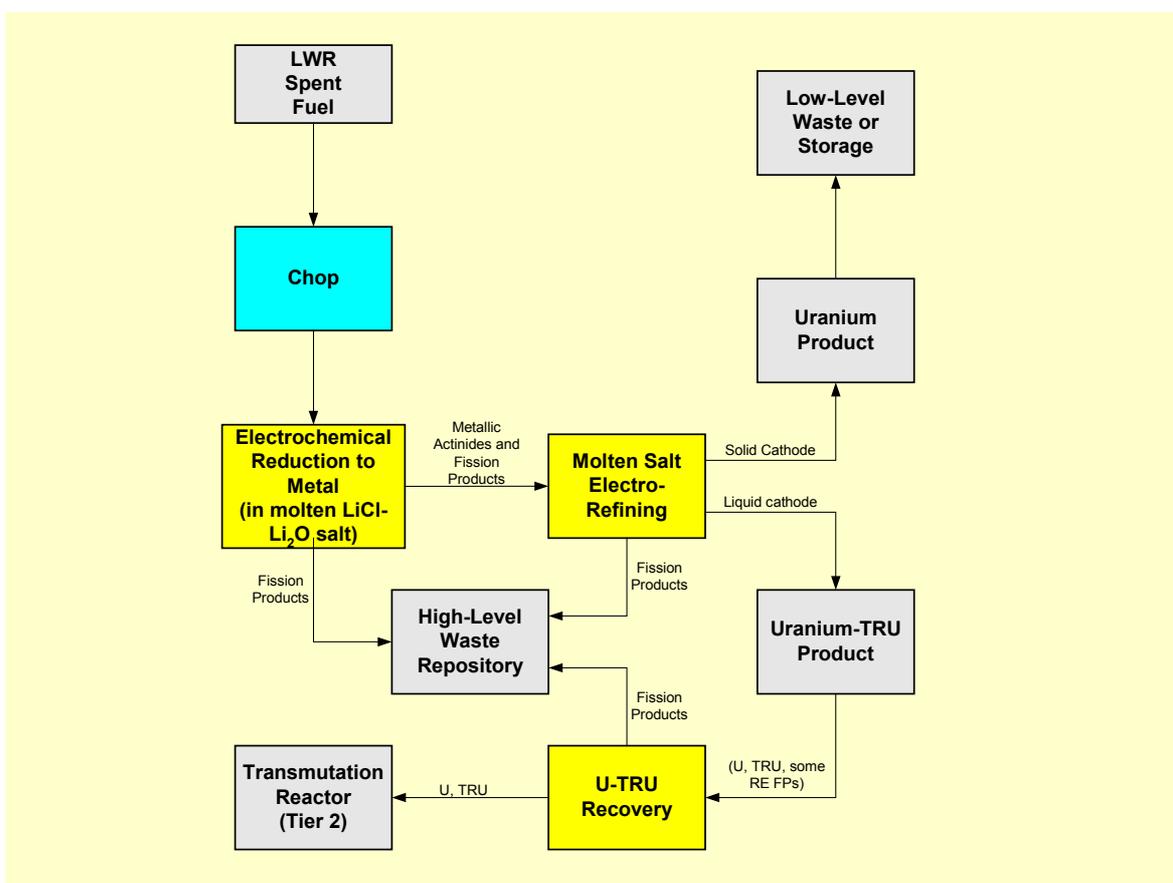
Separation of these +3 actinides from the +3 lanthanide fission products is particularly challenging.

A pyrochemical process known as PYROX is under development for possible application to the treatment of spent LWR oxide fuel. As seen in Figure 6, it is much like the PYRO-A process in using electrochemical reduction of the oxides to provide a feed to the electrorefining step where fission products are separated from the actinides. In the PYROX process, uranium is carried through the electrorefining process, requiring a high throughput capacity for the electrorefiner. The bulk of the uranium is separated as a pure stream, but some uranium carries over with the transuranics during the process of TRU recovery by electrolysis of the actinide-bearing molten-salt. The final product of the process, an alloy of the transuranic elements with some 20-30% uranium, can be sent to a fast-spectrum reactor or it can be processed further by pyrochemical or aqueous methods to achieve the separation of certain desired constituents.

## Development schedule

Separation technology for the AFC programme will be developed over the next five years, leading to a decision in 2007 on deployment of the partitioning and transmutation system. The demonstration of process steps will continue through 2006, at which point a decision on preferred technologies will be made. Engineering-scale demonstrations of the principal separations processes (UREX+ and PYROX) will be carried out at the 20 t scale (UREX+) and the 50 kg scale (PYROX) to provide a basis for future development and deployment decisions. Pre-conceptual design studies for a production-scale facility will be carried out in parallel with the process demonstrations, consistent with a design decision leading to a production-scale plant in operation by 2015.

Figure 6. The PYROX pyrochemical process for treatment of LWR oxide fuel



If required, additional process steps can be added for the separation of individual actinide elements

## REFERENCE

- [1] *Accelerator-driven Systems and Fast Reactors in Advanced Fuel Cycles: A Comparative Study*, OECD-NEA Report, 2002.