

# CHEMICAL SEPARATIONS TECHNOLOGIES FOR THE US ACCELERATOR TRANSMUTATION OF WASTE PROGRAMME

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## Abstract

Management of the spent nuclear fuel generated by the operating commercial reactors in the United States is entering a new phase because it is clear that the continued rate of accumulation of spent fuel is such that the spent fuel inventory will soon exceed the legislated capacity of the proposed Yucca Mountain repository. An integrated chemical separations system has been conceived for the partitioning of this fuel preparatory to transmutation of transuranic elements and long-lived fission products in an accelerator-driven transmuter reactor. A hybrid aqueous/pyrochemical separations system is being developed, with the initial separation done with an aqueous solvent extraction process called UREX. The UREX process extracts uranium, technetium and iodine and directs the transuranic elements and other fission products to the liquid waste stream. The uranium is sufficiently pure that it can be disposed as a low-level waste, while the technetium and iodine are converted into targets for transmutation to stable isotopes. The liquid waste stream containing the transuranics is converted to solid oxide form and the transuranics are separated from the fission products by electrorefining after having been converted to the metallic state. Demonstrations of the process with actual LWR spent fuel are in progress.

## 1. INTRODUCTION

The nearly 100 operating commercial nuclear power stations in the United States generate about 2000 tonnes of spent fuel each year. Presently, this spent fuel is being stored in water basins or dry cask storage areas at the reactor sites; some spent fuel has been transferred to offsite independent spent fuel storage installations. Because the spent fuel has been accumulating for decades, the total inventory will soon reach a level that exceeds the legislated limit (around 63 000 tonnes of spent nuclear fuel) for the proposed Yucca Mountain deep geologic repository.

The U.S. Accelerator Transmutation of Waste (ATW) program is directed toward the development of an alternative approach to high-level waste management. This program would provide an integrated partitioning and transmutation system for the fissioning of transuranic elements and the transmutation of environmentally-important long-lived fission products. Efficient elimination of these radionuclides would greatly reduce the radiotoxicity of the material to be disposed in the repository and thereby the cumulative risk to the public. By removal of the long-lived radionuclides of concern to public health over periods of several hundred thousand years, the regulatory lifetime of the repository could be reduced to time periods that are well within the realm of recorded human history and hence amenable to credible performance predictions. If properly designed, the chemical separations technologies employed for partitioning might be capable of reducing the volume of high-level waste to be disposed, thus extending the capacity of the repository and possibly precluding the need for a second repository in the near term. All of these advantages of partitioning and transmutation can lead to an optimized nuclear waste management strategy that can be expected to receive favorable public reception and reduce opposition to the construction and operation of a high-level waste repository.

## 2. SEPARATIONS REQUIREMENTS

Conceptual design of the ATW chemical separations system is proceeding on the basis of a requirement to process 1500-2000 tonnes (heavy metal) of commercial spent fuel per year. This is taken to be a virtually homogeneous feed of zircaloy-clad oxide fuel with burnups ranging from 20 to 70 gigawatt-days per tonne and with cooling periods of at least 10 years

and possible more than 25 years. A number of target performance requirements have been established for the processing of this fuel. Principal among these is a requirement to separate uranium as a pure product, sufficiently pure that it will meet present regulations for disposal of low-level waste in greater confinement disposal (GCD) sites. Among other limitations, this requires that the total transuranic content be less than 100 nanocuries per gram of uranium. Other target requirements pertain to the efficiency of recovery of specific nuclides: over 99.9% recovery for the transuranic elements, and over 95% recovery for the long-lived fission products  $^{129}\text{I}$  and  $^{99}\text{Tc}$ . In a more general sense, a guideline for minimization of process waste generation has been adopted for all process design evaluations.

## 2.1. Separations system concept

The overall chemical separations scheme that has been selected for initial study in the ATW program is shown in Fig. 1. An aqueous separations method was chosen for the extraction of uranium; this process has been named the UREX process. Iodine is removed in the dissolution step and technetium is co-extracted with uranium and subsequently separated. The UREX liquid waste stream contains the transuranic elements and all of the remaining fission products. This liquid stream is denitrated and converted to dry oxide powder. The oxide product is sent to the PYRO-A process, a pyrochemical process in which the fission products and transuranics are efficiently separated. The transuranics are recovered as metals that are then sent to the transmuter reactor fuel fabrication step.

After irradiation in the accelerator-driven transmuter reactor to burnups as high as 30 atom percent, the discharged transmuter fuel will be processed to recover the unburned transuranics for recycle and to extract the newly-generated iodine and technetium for transmutation. The process for accomplishing these separations has been given the generic name PYRO-B. The PYRO-B process must deal with fuel that has been cooled for only a short period (about 2 years, to minimize further storage requirements and reduce plant costs) and that contains a significant concentration of fissionable transuranic isotopes. The quantity of fuel that must be processed in one year is much less than in the case of the LWR spent fuel (perhaps 30 tonnes per year vs. 1500-2000 tonnes for LWR spent fuel). These considerations led to the selection of a pyrochemical process for treatment of the transmuter fuel, which has been assumed to have zero uranium content (i.e., a non-fertile fuel). Pyrochemical systems, being free of a water moderator, can accommodate larger masses of fissile isotopes. The molten salts used in these processes operate at elevated temperatures and are stable against alpha and gamma radiation damage, so short fuel cooling periods are feasible.

The only process high-level waste streams arising in the overall separations system come from the pyrochemical processes (PYRO-A and PYRO-B). The similarity of these processes is such that the waste streams are virtually identical. The transition metal fission products accumulate in metallic form and are combined with decontaminated cladding hulls to form a metal alloy waste form. The alkali metal and alkaline earth fission products are initially concentrated in process salts and then absorbed in zeolite that is subsequently transformed to the mineral sodalite; in the process of sodalite synthesis, the sodalite particles are encapsulated in borosilicate glass, yielding a glass-ceramic composite with good leach resistance.



proliferation resistance of the system. The collection of transuranics is sent directly to the transmuter fuel fabrication step.

### **2.3. ATW transmuter fuel processing**

At the present time, a final selection of the fuel type for the transmuter reactor has not been made. It is assumed for now that the fuel is a metallic alloy of transuranics in zirconium, with a composition in the range 40-50 weight percent transuranics and a target burnup of 20-30 atom percent. The initial concept for treatment of this fuel utilized a chloride volatility process whereby the zirconium matrix was digested by formation of volatile  $ZrCl_4$ . The transuranics were then to be extracted by electrowinning from the chloride salt containing transuranics and fission products. While this process may be appropriate for a dispersion of TRU-Zr particles in a zirconium matrix, recent fuel properties evaluations tend to favor a homogeneous TRU-Zr alloy. In this case direct electrorefining of the TRU-Zr fuel appears to be a better option. There is a precedent for this process in the treatment of EBR-II spent fuel, where the fuel composition for a portion of the fuel to be treated is (U,TRU)-10Zr.

### **2.4. Status of chemical separations technology development**

The UREX process flowsheet has been demonstrated with simulated LWR spent fuel. The extraction of transuranics was completely inhibited by the AHA complexant/reductant; consequently, the decontamination of uranium was well below the 100 nanocuries per gram level. The recovery efficiency for uranium and transuranics exceeded 99.999%. Technetium was not extracted in these initial experiments because the acid concentration was not optimized. A complete demonstration of the UREX flowsheet, modified for technetium extraction, will be carried out with a bank of 33 centrifugal contactors in the latter half of 2001. A demonstration of the full process for LWR spent fuel treatment, from UREX separations to denitration and TRU recovery in the PYRO-A process, will be conducted with actual spent fuel beginning in October 2001 and continuing through 2003.

The PYRO-B process for irradiated transmuter fuel is at an earlier stage of development, but will benefit from experience with EBR-II spent fuel treatment. Fuel types other than metal alloy are under consideration, and conceptual flowsheets have been developed for the processing of non-fertile oxide fuel with a ceramic matrix (cercer fuel), TRU nitrides in a ZrN matrix, and TRU oxide and nitride in a metal matrix (cermet fuel). Extensive (and expensive) demonstrations with the transmuter fuel will be deferred until the preferred fuel type is selected.

## **3. CONCLUSIONS**

The chemical separations technology development described in this paper pertains to a single-tier transmutation system in which the transuranics and long-lived fission products from LWR spent fuel are sent to an accelerator-driven fast spectrum system for transmutation. There is increasing interest in the U.S., however, in a multi-tier system. In such a system, the existing infrastructure of commercial light water reactors is supported by new advanced thermal reactors (Tier 1) that can be used as efficient burners of plutonium in a close-coupled system. The unburned minor transuranics in the advanced thermal reactor spent fuel would be directed to a fast spectrum reactor (Tier 2), either critical or accelerator-driven subcritical, where the minor actinides can be destroyed. This multi-tier system would clearly impose different requirements on the associated chemical separations technologies, and the separations processes that would be needed are now being evaluated.