

Chemical Separations Schemes for Partitioning and Transmutation Systems, J. J. Laidler (ANL)

In the initial phase of the U.S. Accelerator Transmutation of Waste (ATW) program, a single-tier system was foreseen in which the transuranics and long-lived fission products (specifically, ^{99}Tc and ^{129}I) recovered from spent LWR oxide fuel would be sent directly to an accelerator-driven transmuter reactor [1]. Because the quantity of fuel to be processed annually was so large (almost 1,500 tons per year), an aqueous solvent extraction process was chosen for LWR fuel processing. Without the need to separate transuranics from one another for feed to the transmuter, it became appropriate to develop an advanced aqueous separations method that became known as UREX. The UREX process employs an added reagent (acetohydroxamic acid) that suppresses the extraction of plutonium and promotes the extraction of technetium together with uranium. Technetium can then be efficiently removed from the uranium; the recovered uranium, being highly decontaminated, can be disposed of as a low-level waste or stored in an unshielded facility for future use. Plutonium and the other transuranic elements, plus the remaining fission products, are directed to the liquid waste stream. This stream is calcined, converting the transuranics and fission products to their oxides. The resulting oxide powder, now representing only about four percent of the original mass of the spent fuel, is reduced to metallic form by means of a pyrometallurgical process. Subsequently, the transuranics are separated from the fission products in another pyrometallurgical step involving molten salt electrorefining.

A non-fertile metallic fuel was proposed for use in the transmuter system, to consist of an alloy of transuranic isotopes with an inert diluent such as zirconium. Multi-recycling of the transmuter fuel was required in order to achieve the desired level of destruction of transuranics. Processing of this fuel type by electrorefining should be straightforward; it has been demonstrated at very small scale and appears to be technically feasible. It should behave much like the EBR-II driver fuel, with the exception that the zirconium content of the transmuter fuel is much higher and the fuel is free of uranium. The latter characteristic makes it possible to deposit the transuranics on a simple solid cathode. This eliminates the complex cathode configuration and subsequent distillation step that represents a process complication in the case of transuranic recovery from fertile fuel.

Recently, attention has been directed to a more practical dual-tier system for transuranic transmutation [2]. Oxide fuel discharged from existing commercial LWRs ("Tier 0") would be processed to separate Pu (or Pu and Np) for burning in a Tier 1 thermal spectrum reactor that could be either an advanced LWR or a gas-cooled reactor. Americium and curium extracted from the Tier 0 fuel, together with unburned transuranics remaining after Tier 1 irradiation, would be recovered and sent to a Tier 2 fast spectrum transmutation reactor that could be either a critical reactor or an accelerator-driven subcritical reactor. Chemical separations processing

of the discharged Tier 1 and Tier 2 fuel is effectively limited to pyrochemical methods, because the spent fuel would have a high level of alpha activity, even after extended cooling, that would result in degradation of the fragile organic solvents employed in aqueous separations processes. As shown in Figure 1, the chemical separations processes would be applied to fuels discharged from each tier, with the initial processing of Tier 0 LWR fuel being an aqueous or hybrid aqueous/pyrochemical process. This step would require Pu and Np separation from the other transuranics, with the Am and Cm directed to the Tier 2 transmuter reactor. The long-lived fission products ^{99}Tc and ^{129}I would be recovered in each processing step and sent to the thermal spectrum system for transmutation to stable ^{100}Ru and ^{130}Xe , respectively.

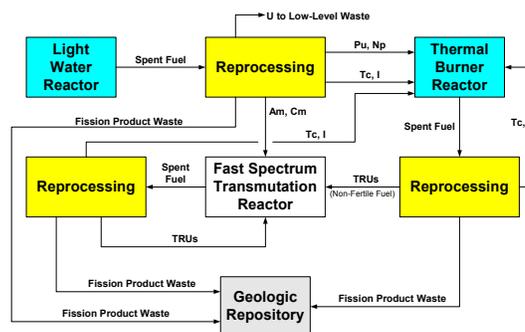


Figure 1. Fuel cycle scheme for a multi-tier partitioning and transmutation system.

An R&D program is in progress for development of the technologies for the various separations processes required in a multi-tier system. Demonstrations of the Tier 0 fuel processing systems with actual spent fuel will be complete by 2004, and the entire system will be demonstrated with spent fuel by the end of the decade.

1. U.S. DEPARTMENT OF ENERGY, "A Roadmap for Developing Accelerator Transmutation of Waste (ATW) Technology – A Report to Congress," DOE/RW-0519, October 1999.
2. G. J. VAN TUYLE AND P.J. FINCK, "Candidate Approaches for an Integrated Waste Management Strategy – Scoping Evaluations," Report No. AAA-PDO-GEN-01-0051, November 2001.

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