

The Proliferation Resistance of a Nuclear Fuel Cycle Using Fuel Recovered from the Electrolytic Reduction of Pressurized Water Reactor Spent Fuel

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1. Introduction

"Proliferation-resistance" refers to the adoption of reactor and fuel cycle concepts that would make the diversion of civilian nuclear fuel cycles by states or by sub-national groups to weapons purposes more difficult, time-consuming, and transparent [1]. At some points in the fuel cycle, a level of intrinsic or technical proliferation-resistance can be provided by radiation barriers that surround weapons-usable materials. In this report we examine some aspects of intrinsic proliferation resistance of a fuel cycle for a fast neutron reactor that uses fuel recovered from the electrolytic reduction process of pressurized water reactor spent fuel, followed by a melt-refining process. This fuel cycle, proposed by a nuclear engineer at the Korea Advanced Institute of Science and Technology (KAIST) [2], is being examined with respect to its potential merits of higher fuel utilization, lower production of radioactive byproducts, and better economics relative to a pyroprocessing-based fuel cycle. With respect to intrinsic proliferation resistance, however, we show that since europium is separated out during the electrolytic reduction process, this fuel cycle has little merit beyond that of a pyroprocessing-based fuel cycle because of the lower radiation barrier of its recovered materials containing weapons-usable actinides.

2. Converting Oxide Fuel into Metal Fuel

To make metal fuel for a fast neutron reactor, the ceramic form of pressurized water reactor (PWR) spent fuel must be converted into metallic form. The Korean Atomic Energy Research Institute (KAERI) favors pyroprocessing, which consists of a series of spent fuel processes: head-end; decladding and voloxidation; electro-reduction; electrorefining; and electrowinning [3, 4]. The head-end process disassembles PWR spent fuel and chops the fuel into the proper size for the decladding process. The recovered UO₂ is pulverized and converted into U₃O₈ powder by the high-temperature voloxidation process to reduce particle sizes in order to promote fast reactions in the oxide reduction process. During the voloxidation process the following volatile and semi-volatile fission products are removed: Cs, I, Kr, Mo, Rh, Ru, Tc, Te and Xe [4, 5, 6].

The voloxidized oxide fuel is introduced into LiCl-Li₂O molten salt to be reduced to metallic form. The

electrolytic reduction process causes the oxide ions in the spent nuclear fuel to escape as gas and leave the metallic spent fuel in the electrolytic cathode basket, while alkali elements and alkali earth elements are dissolved in the electrolyte. Lanthanides, except europium (Eu) and other metallic fission products, remain in the cathode [4]. The fission products removed by the electro-reduction process which remain in the electrolyte are: Ba, Cs, Eu, I, Rb, Sr and Te [4, 5, 6].

3. Radiation Dose Rates

Table 1 shows calculated dose rates from this work from a 1-kilogram metal sphere of derived from spent PWR fuel with a burnup of 50 MWd/kgHM, containing weapons-usable actinides, at different points in the fuel cycle. The composition of the PWR spent fuel was calculated with ORIGEN 2.1, assuming an initial fuel enrichment of 4.2 percent U-235. The gamma radiation doses were calculated using the American Nuclear Society's gamma-ray fluence-to-dose factors [7]. The self-shielding of the metal sphere was calculated at a density of 16.2 g/cc using the MCNP4C2 Monte Carlo particle-transport code developed at Los Alamos National Laboratory. By comparison, radiation sickness can occur for an acute dose of 1-2 Sv, and death likely occurs for an acute dose of 10 Sv.

Table 1. Dose rates 1 meter from the surface of a 1 kilogram metal sphere derived from spent PWR fuel with a burnup of 50 MWd/kgHM 10 years after discharge (column 2); after removing volatile and gas fission products (column 3); after further electrolytic reduction (column 4); and after further removing separated transuranic mix (column 5).

	PWR spent fuel	Recovered after voloxidation process of the PWR spent fuel	Recovered after voloxidation and electrolytic reduction processes of the PWR spent fuel	Transuranic mixture separated from the PWR spent fuel (similar to product following pyroprocessing)
Dose Rate (Sv/hr)	0.085	0.014	0.00043	0.00003
Dose rate extrap	42.5	7.0	0.215	0.015

olation to 500 kg (mass of fuel assem bly)				
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* Dose rate 1 meter from a PWR spent fuel assembly with a burnup of 50 MWd/kgHM 10 years after discharge is about 14 Sv/hour. The difference between this value in column 2 is assured to be due to geometry consideration – the difference between a sphere and a fuel assembly.

4. Discussion

One of important lanthanide fission product contributions to the calculated dose rate is Eu-154, with a half-life of 9 years. The separation of Eu-154 during the electrolytic reduction process dramatically lowers the radiation barrier of recovered material that includes weapons-usable actinides, i.e. reduction to 0.5% of that of spent fuel and 3% of that of material recovered after the voloxidation process of the spent fuel.

Another important lanthanide fission product, Ce-144 has a half-life of only 0.8 years. Thus, its dose effect becomes negligible for the PWR spent fuel 10 years after discharge [6].

5. Concluding Remarks

Unless europium is not separated following voloxidation, the proposed KAIST fuel cycle is not intrinsically proliferation resistant and in this regard does not represent a significant improvement over pyroprocessing.

We suggest further modification of the proposed KAIST fuel cycle, namely, omitting electrolytic reduction and melt reduction, and producing the fast reactor fuel directly following voloxidation.

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