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HIGH-LEVEL RADIOACTIVE WASTE DISPOSAL BY TRANSMUTATION*

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A possible approach to the ultimate disposal of high-level radioactive waste is to transmute long-lived radioactive nuclides to stable or short-lived nuclides by exposure to neutrons.

Studies^{1,2} have been made on the possibility of using special high-flux "burner reactors" to reduce the stockpile of the "problem fission products" ^{90}Sr and ^{137}Cs . A reevaluation of the burner reactor design of Ref. 2 showed a cost penalty of possibly 0.8 mill/kWhr(e) when more realistic fission product yields were used. Considering the cost and the extensive development required to solve some of the problems associated with such high flux reactors ($\sim 10^{17}$ n/cm²-sec), transmutation appears unattractive in comparison with storage in geological formations.

Heavy isotopes that are produced in reactors and their daughters have half-lives in the thousands of years and are mostly alpha-emitters. Consequently, a stronger motive exists for complete elimination rather than storage in geological formations. It is obviously more difficult to predict the tectonics of geological formations for 10^6 years compared to the less than 1000 years required for the decay of ^{137}Cs and ^{90}Sr to innocuous levels. In present concepts of power reactors, it is assumed that 99.5 to 99.9% of the uranium and plutonium is reused and that other heavy elements and their daughters are rejected in the waste. The radioactivity of this waste can be reduced by recycling the actinides through the power reactors producing them; elimination occurs by fission at points in the reaction paths.

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To study the effect of actinide recycling, ORIGEN,³ an isotope generation and depletion code, was modified to include the actinides beyond ^{244}Cm up to ^{253}Es , a neutron balance calculation, and a recycling option. Some pertinent results of the calculations with the modified code are shown in Table 1 for a typical PWR and a 3-group neutron spectrum. The results consider 15 recycles of the actinides through the reactor (minus the small fraction that goes to the waste streams after each pass) with its normal loading. By then, k-infinity and the hazard measure, namely, the water required for dilution of the waste to that recommended (RCG) in the Code of Federal Regulations,⁴ attained equilibrium for all practical purposes. The maximum reactivity decrease was 0.8%, which can be counteracted by only a 2% increase in fissile material. The hazard measure of the waste is approximately proportional to the transplutonium actinides present, and the efficiency of recycling was about 20%; e.g., a reduction factor of about 200 in 10^6 years in the hazard measure was obtained when 0.1% of the actinides appeared in the waste in contrast to a maximum possible 1000. For the 0.5% case, which contained only one-fifth the amount of transplutonics as the 0.1% case, the reduction factor was 40. Presumably another factor of 10 reduction can be obtained if 99.99% of the actinides can be recycled. A corollary is that, without recycling, no significant reduction in the hazard measure of the waste is obtained by recovering more than 99.5% of the Pu and U.

Application of recycling of the actinides will increase the cost of the nuclear fuel cycle primarily by requiring additional separations processes in reprocessing plants and remote operation of fuel fabrication plants. A crude estimate indicates the cost increase would be in the order of 0.1 mill/kWhr(e).

Table 1. Effect of Recycle of Actinides on Alpha Activity Waste from Reprocessing PWR Fuel^a

Time After Processing (years)	Water Required for Dilution to RCG for Unrestricted Use (m ³ /ton of fuel)						Nuclide ^d of Predominant Hazard	
	0.1% ^b of Pu + U to Waste			0.5% ^c of Pu + U to Waste			Standard	Recycled
	Standard ^a	Recycled	Stand./Recy. ^e	Standard	Recycled	Stand./Recy.		
10 ³	2.61 x 10 ⁷	1.37 x 10 ⁶	19	2.74 x 10 ⁷	6.72 x 10 ⁶	4.1	²⁴⁵ Cm	²⁴⁶ Cm
10 ⁴	9.95 x 10 ⁶	4.19 x 10 ⁵	24	1.04 x 10 ⁷	2.06 x 10 ⁶	5.1	²⁴⁵ Cm	²⁴⁶ Cm
5 x 10 ⁴	1.41 x 10 ⁷	2.34 x 10 ⁵	60	1.46 x 10 ⁷	1.17 x 10 ⁶	12	²²⁹ Th + daughters	²²⁶ Ra + daughters
10 ⁵	2.64 x 10 ⁷	3.53 x 10 ⁵	75	2.71 x 10 ⁷	1.76 x 10 ⁶	15	²²⁹ Th + daughters	²²⁶ Ra + daughters
10 ⁶	5.97 x 10 ⁷	2.92 x 10 ⁵	204	5.86 x 10 ⁷	1.46 x 10 ⁶	40	²²⁹ Th + daughters	²²⁹ Th + daughters

^a 3.3% enriched uranium irradiated at an average specific power of 30 MW/metric ton to a burnup of 33,000 MWd/metric ton in a typical PWR. Processing 150 days after discharge. All actinides and daughters to waste.

^b 99.9% of remaining actinides recycled but all daughters to waste.

^c 99.5% of remaining actinides recycled but all daughters to waste.

^d Insensitive to Pu + U in waste.

^e These values are essentially an asymptotic minimum and were significantly higher for the waste discharged in the first two or three cycles.

References

1. M. Steinberg, G. Wotzak, and B. Manowitz, Neutron Burning of Long-Lived Fission Products for Waste Disposal, BNL-8558 (1964).
2. M. V. Gregory and M. Steinberg, A Nuclear Transformation System for the Disposal of Long-Lived Fission Product Waste in an Expanding Nuclear Power Economy, BNL-11915 (1967).
3. Chemical Technology Div. Annual Progress Report, May 31, 1969, ORNL-4422, p. 89.
4. Code of Federal Regulations, Title 10, Part 20, Table II, Column 2.