ACTINIDE DEPLETION STUDIES

J. W. Lewellen
U. S. Department of Energy
Washington, D.C., 20545

It is well known that plutonium, neptunium, curium and americium isotopes form in considerable quantities as byproducts of power reactor operations.

To the degree that the heavy actinides fission, they contribute to overall nuclear fuel utilization. To the degree that they remain in spent fuel or high level waste destined for disposal, they add to the long-lived radioisotope inventory to be controlled. The best means of managing the actinides is not yet determined; however it is expected to depend on detailed aspects of reactor irradiation conditions, outcomes of fuel cycle development work, choices of reactor types and fuel management strategies, and upon the success of developmental procedures for radioactive waste storage.

The technical aspects of actinide formation and burnup are being studied in the U.S. with two major objectives. The first is to characterize neutronics which describe heavy actinide production, burnup by conversion to fission products, and evolution of the actinide isotope ensembles during operational exposure of power reactor fuel. The second objective is to understand elements of actinide separations from each other and from fission products.

Neutronic transmutation studies in fast reactors have been carried out at General Electric and Oregon State University. Savannah River Laboratory studies have also focused on actinide burnup in plutonium-recycle PWRs. These studies typically begin with calculations of actinide depletion corresponding to representative models of actinide loads in a "host" reactor such as an LMFBR or PWR. The results show that substantial actinide depletion, permitting equilibria between production and fission utilization, is feasible in principle.[1,3] Generally, the fission to capture ratios progressively improve with increasing neutron energy, and utilization and control of actinide inventories in LMFBRs is therefore more efficient than in PWRs.

If such equilibria can be reached, then the inventory of heavy actinides could be small, stable and entirely contained in operating reactors and fuel support facilities. Their production would have little long-term consequence other than being a secondary part of useful energy production from nuclear fuel.

Beyond this, however, practicalities complicate the picture greatly. Important issues still being considered include:

- o Mode of reprocessing consistent with actinide burnup, including the magnitude of lanthanide fission product carryover with the actinide stream.
- o Achievability of equilibria and size of equilibrium inventories may be sensitive to timing of initial actinide recycle in fast and/or thermal reactors.[2]
- o Sensitivity of production and burnup ratios to actinide cross sections.
- o Modes of actinide loading in reactors (e.g., separate "target" pins versus blends with major fuel isotopes; target pin distribution can also be design variable).
- o Interest in special fast reactors optimized for actinide utilization.[2,3] These would feature hard spectra to improve the fission/capture efficiency and high depletion fractions per residence to minimize recycle volume. Possible actinide diluents include tungsten, molybdenum and thorium.

Preliminary results indicate sufficient promise connected with the last of the above to warrant further consideration. A brief survey of infinite actinide driver lattices showed maximum case americium 241 and 243 reductions of 60% and 40% after a three year burn cycle. Although the amounts fissioned are less because of parallel transmutation to curium 242 and 244, these numbers represent attractive rates of consumption. In this part of the work, the average neutron energy is about 0.2 MeV, the power density is 600 KW/l and highest flux and fractional burnup are obtained with 50 w/o

moly diluent. Also, target actinides constitute a major component of the reactivity driving fuel inventory. Therefore, as actinide depletion occurs, these target materials are exposed to increasing neutron fluxes when constant power densities are maintained.

As an alternative, passive actinide targets in an EBR-II like driver showed less effective americium consumptions of about 35% and 20%. In this case, the neutron flux irradiating the target materials would be lower and about constant during a burn cycle; therefore, depletion is retarded despite a slightly higher energy neutron spectrum. Some results of these calculations are illustrated in Figure 1, which is from Reference 3.

Assessment of actinide management by burnup also requires fuel recycle studies and research on chemical partitioning (recovery) of long-lived actinides from radioactive waste. The work of this type is carried out at ORNL and is broadly based, consisting of both experimental and computational activities. Several DOE installations, such as Allied Chemical, ANL and Rocky Flats which have specialized experience and experimental facilities are also participating in this program.

The various studies are expected to produce (1) reprocessing and mixed oxide fuel refabrication plant partitioning flowsheets which have been at least partly verified by experimental benchscale work; (2) a meaningful cost risk/benefit analysis; and (3) an estimate of the scope and magnitude of the development program that would be needed to implement actinide partitioning for recycle. Chemical flowsheets for partitioning actinides from fuel reprocessing and fabrication plant wastes are being developed for use in feasibility and incentive studies. The processes used in these flowsheets are those being experimentally investigated at ORNL and other DOE installations. Some parts of this work promise to be useful for reducing fuel wastage whether or not americium and curium are recovered. For example, the ANL salt waste approach facilitates recovery of that fuel otherwise lost to carbonate scrub wastes by extracting the solvent degradation products from the acidified salt wastes. After the degradation

products are removed, the salt wastes containing actinides can be recycled through the chemical separations. On completion of these studies, it should be possible to judge whether actinide management alternatives are worthy of development through the demonstration stage.

In brief summary, several aspects of actinide recycle are being explored. These include studies of the capacity of FBR or LWR systems to reduce a previously accumulated actinide inventory as well as research on partitioning and actinide recovery processes. Results of risk and economic analysis tasks will also help define research and development required to achieve actinide management objectives.

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1. S. L. Beaman and H. S. Bailey, "Plutonium Burnout in Fast Reactors as a Waste Management Alternative," Trans. Am. Nucl., 806 23, 263, 1976.

2. S. L. Beaman, "Plutonium and Actinide Burnout as a Waste Management Alternative," Trans. Am. Nucl. Society, 23, 351, 1978.

3. A. H. Robinson, "Studies of Actinide Waste Burnup and Use in Fast Spectra," Report RCO/2227/T32-6, July 1978.

Fraction of Initial Americium Consumed in
3 Years vs. Initial Flux Level

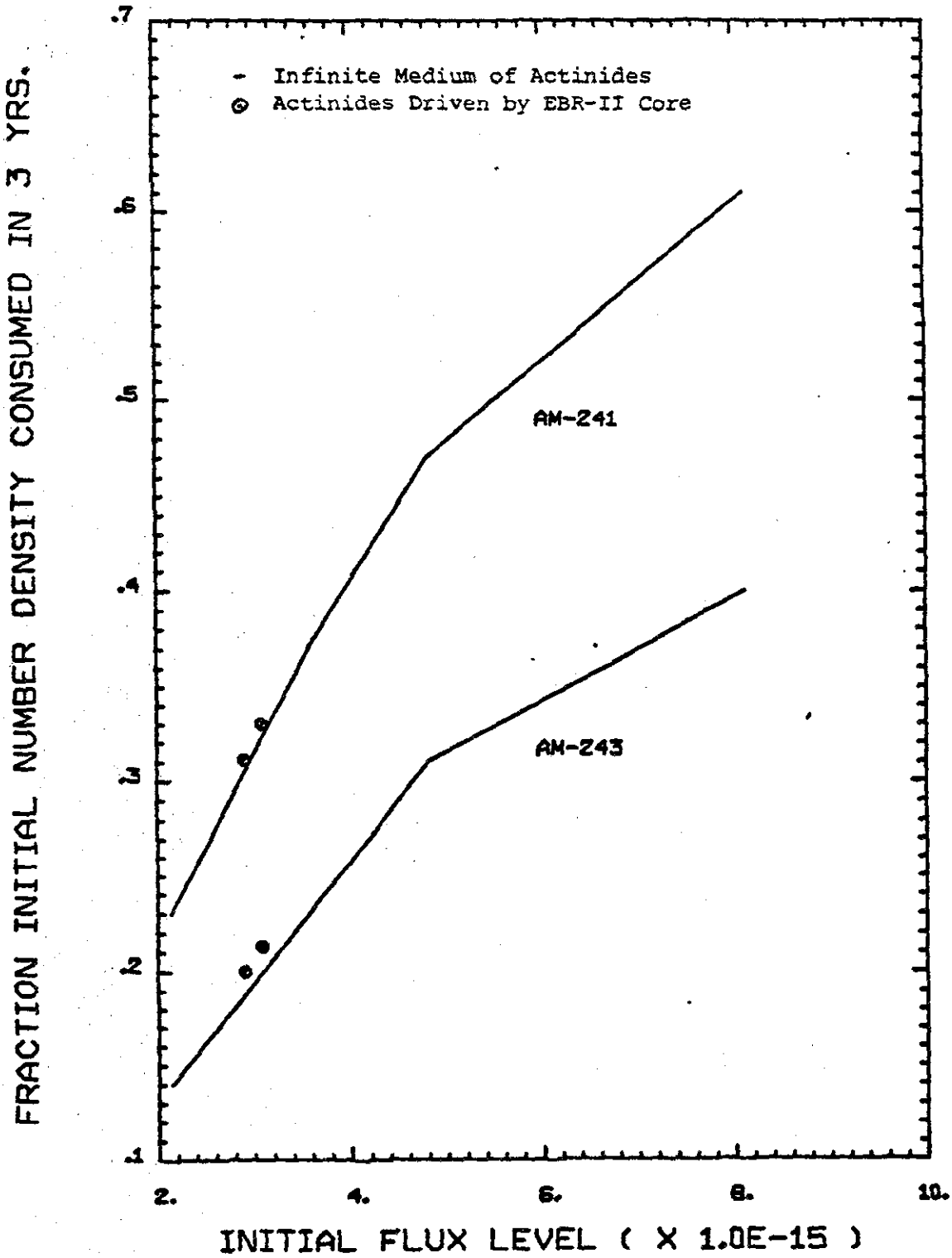

FIGURE 1