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TRANSMUTATION OF LWR WASTE ACTINIDES IN THERMAL REACTORS

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ABSTRACT

Recycle of actinides to a reactor for transmutation to fission products is being considered as a possible means of waste disposal. Actinide transmutation calculations were made for two irradiation options in a thermal (LWR) reactor. The cases considered were:

- All actinides recycled in regular uranium fuel assemblies.
- Transuranic actinides recycled in separate mixed oxide (MOX) assemblies.

When all actinides were recycled in a uranium lattice, a reduction of 62% in the transuranic inventory was achieved after 10 recycles, compared to the inventory accumulated without recycle. When the transuranics from 2 regular uranium assemblies were combined with those recycled from a MOX assembly, the transuranic inventory was reduced 50% after 5 recycles.

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INTRODUCTION

Partitioning the actinides in fuel wastes from light water reactors and transmuting them to fission products in power reactors represents a waste management concept which could reduce the long-term risk associated with geologic isolation of the wastes (Slide 1).

- Partitioning is defined as the chemical process in which the level of actinides in radioactive waste is reduced to a greater extent than dictated by normal economic consideration, and where effective recovery of the actinides is achieved.
- Transmutation is defined as the irradiation process in which the actinides are converted to fission products in a reactor.

The Chemical Technology Division of Oak Ridge National Laboratory (ORNL) is coordinating a program[1] involving several national laboratories and commercial organizations to evaluate the feasibility and incentives that may exist for implementing the concept. Studies and tests pertaining to chemical partitioning of the actinides, economic analyses, and risk assessments are being conducted at other sites, and are not discussed in this paper. The portion of the process considered in the Savannah River Laboratory (SRL) study pertains to neutron-induced transmutation in thermal reactors. Specifically, computations were made to assess methods in which the waste actinides would be recycled and reirradiated in various fuel or target forms.[2,3] Two of the options are described in this paper:

- The recycle of all actinides, with all assemblies in the lattice containing similar actinide loadings.
- The recycle of the transuranics in separate assemblies comprising 1/3 of the lattice, with the remaining 2/3 of the lattice made up of assemblies containing 3.2% ^{235}U.

SUMMARY (Slide 2)

Recycle of all actinides in a uniform lattice can be implemented by first removing about 10% of the uranium from the dissolver solution to permit addition of "new" ^{235}U at 20% enrichment as needed for reactivity. A reduction of 51% in the transuranic inventory was achieved after 5 recycles and 62% after 10 recycles, compared to the inventory accumulated without recycle.

The recycle of transuranics in separate mixed oxide (MOX) assemblies is an equally viable option. The transuranics from 2 regular uranium assemblies were combined each cycle with those recycled in a MOX assembly containing slightly enriched uranium. The transuranic inventory after 5 recycles was about 50% of the inventory without recycle.

The use of two or more enrichments may be necessary in MOX assembly fuel pins to reduce power peaking at the assembly edge, when MOX and uranium assemblies are present in the same lattice.

DESCRIPTION OF COMPUTATIONAL METHOD

The SRL GLASS code[4] was used for all neutronic and depletion calculations in this study. GLASS performs the following general operations (Slide 3).

> A basic lattice physics calculation with multigroup integral transport methods. Options include 37 or 84 energy groups.
>
> A Nordheim calculation of self-shielded resonance integrals.
>
> An isotope depletion and decay calculation for all actinides and selected fission products based on reaction rates from the physics calculation. The neutron energy spectrum calculation is repeated at regular intervals during the depletion calculation.

Input parameters (Slide 4) include isotopic compositions, temperatures, and spatial dimensions in annular, square, or hexagonal geometry. A standard cross section data base is part of the SRL library, which is regularly updated to provide good results for thermal and near-thermal reactors. The data base currently includes the Phase I ENDF/B-V data for the transplutonium actinides.[5]

A typical pin cell (Slide 5) consists of clad UO_2 fuel surrounded by H_2O coolant in a square pitch. The unit cell is translated in the XY plane to form an infinite lattice.

GLASS may also be used to simulate multi-region annular geometry within a cell, and different cells may be linked to form "supercells." A supercell is defined as a connected group of cells which form an array which repeats by translation in a lattice. Examples of supercells are shown in Slide 6.

Entire LWR fuel or target assemblies have been represented by annular mockups; these assemblies may have different properties. The use of supercells is important in the present study because it permits a realistic calculation of resonance self-shielding in target assemblies and at the same time yields a realistic multi-group spectrum that is dominated by the fuel environment.

In all calculations, the fuel was irradiated for 3 years to an exposure of 33,000 MWD/MTM; a one-year cooling period followed. Because reprocessing losses are expected to be small, it was assumed that 100% of the actinides were recovered from fission products and other waste. Only PWR systems were considered.

Option 1: Recycle of all Actinides in Uniform Lattice

In this recycle alternative, all actinides from each fuel assembly were recycled into a new assembly. The lattice model consisted of a uniform array of zirconium clad fuel rods in H_2O coolant-moderator. It was assumed that highly enriched uranium would not be available for LWR fuel fabrication, but rather uranium enriched only to 20% ^{235}U. Thus, the new ^{235}U added for reactivity control was accompanied by 4 times as much ^{238}U, necessitating the removal of 10% of the uranium in the spent fuel (Slide 7).

The cycle-ending transuranic contents obtained from these calculations are shown in Slide 8. None of the transuranics have reached equilibrium values after ten recycles, but the rates of increase per fuel cycle have fallen to 6% or less. The total

inventory of transuranics after ten recycles is only 38% of the inventory that would accumulate without recycle. The plutonium inventory is 31% of the no-recycle inventory.

In other results, the negative reactivity worth of neptunium, americium, and curium (collectively) was found to increase each recycle, to a value of 6% Δk_∞ at Recycle 10. Also, the burnup of ^{235}U in Recycle 10 is about 19 kg/MTM, compared with 24 kg/MTM in regular 3.2% enriched fuel with no actinide recycle. The difference is made up by fissions in the transuranics, primarily plutonium. ^{235}U loadings required for reactivity were between 2% and 5% for the 10 recycles.

Option 2: Recycle of Transuranics in Separate Mixed-Oxide Assemblies

As shown in Slide 9, the reactor contained two 3.2% enriched uranium assemblies for each mixed oxide (MOX) assembly. After irradiation to an exposure of 33,000 MWD/MTM in the uranium fuel, the two assembly types were discharged and reprocessed separately. The transuranics from both process streams were recovered and charged to new MOX assemblies with ^{235}U added as needed for reactivity. The lattice model consisted of a repeating array of uranium and MOX assemblies, in a 2/1 ratio.

Slide 10 compares the cycle-ending transuranic contents of a MOX assembly with those from Option 1. Results are given as grams per metric tonne of fuel in two uranium assemblies. After five recycles, the contents for the 2 options are very similar. ^{237}Np

production in Option 2 is lower because the average ^{235}U content is lower. The total inventory of transuranics after five recycles is about 50% of the inventory that would accumulate without recycle. By Recycle 5, less than 10% of the MOX assembly power is due to ^{235}U fission; plutonium is the primary fissioning material.

The specific powers of fuel pins at the edge of the MOX assembly were considerably higher than the average pin power, when all pins had the same enrichment or fissile content. Qualitatively, the power peaking at the assembly edge was the result of a higher thermal neutron flux in that region, relative to the inner regions. Examples of flux distributions across the two assembly types are given in Slide 11. All the distributions are flat except for the thermal flux in the MOX assembly, which has twice as large a value at the assembly edge as in the center.

Pin powers for a uniform Pu loading are shown in Slide 12, with a peak power of 7.1 kw/ft per pin at the assembly edge. (The ratio of total Pu/total actinides was 8%.) Pin powers for the uranium assembly are relatively flat. A reduction in plutonium content of the outer two rows of pins to 5.0% and 3.0%, respectively, reduced the pin powers of the MOX assembly to about the same as those for the uranium assembly. The Pu enrichment of inner pins was raised from 8 to 10.3%, to maintain the same loading in the assembly.

Although the use of several fuel pin enrichments in the same assembly will create special problems of quality assurance and production control, such a technique will be necessary in mixed lattices of MOX and enriched uranium assemblies if efficient full power operation is to be achieved.

OVERALL CONCLUSIONS OF THE PARTITION-TRANSMUTATION STUDY

The results presented in this paper represent only part of the total number of recycle options considered. Several conclusions can be drawn from the completed studies, as follows:

- The reduction in transuranic inventory achieved by actinide recycle in LWR systems would be on the order of 5 to 10% per fuel cycle over the first several cycles.
- Recycle of waste actinides (Pu excluded) uniformly distributed throughout the fuel assemblies would result in the smallest perturbation in reactor operation, of all cases studied. Increases in ^{235}U content of a few percent would provide the needed reactivity.
- Major changes in reactor operation would be required if Pu were added to the waste actinides, either as separate MOX assemblies or uniformly distributed in all assemblies. The reduced thermal flux in the Pu-containing fuel and other changes in neutronics would result in significant changes in many operating and safety related parameters.

- The uniform loading option offers advantages over the separate assembly option during reactor operation. Power peaking in outer fuel rods is minimal in the first option but could be severe in the second. However, with the separate assembly option, only a third of the fuel assemblies require special fabrication techniques and shielding during fabrication and charging. If the choice of recycle option is made primarily from operating or from fabrication considerations, the MOX assembly is probably more promising.
- Use of uranium as a diluent rather than zirconium is preferred. Difficulties encountered during dissolution of zirconium targets would be severe.

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4. H. C. Honeck. **The JOSHUA System.** USAEC Report DP-1380, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, SC (April 1975).

5. R. W. Benjamin, F. J. McCrosson, V. D. Vandervelde, and T. C. Gorrell. **A Consistent Set of Heavy Actinide Multigroup Cross Sections** USERDA Report DP-1394, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, SC (1975).

SLIDE 1

INTRODUCTION

Definitions

Partitioning - Chemical Process

Transmutation - Irradiation Process

Two Options

1. Recycle of all actinides in uniform loading
2. Recycle of transuranics in separate assemblies

SLIDE 2

SUMMARY

Option 1 - A 62% reduction in the transuranic inventory after 10 recycles

Option 2 - A 50% reduction in the transuranic inventory after 5 recycles

SLIDE 3

OPERATIONS PERFORMED BY GLASS

- A basic lattice physics calculation with multigroup integral transport methods.
- A Nordheim calculation of self-shielded resonance integrals.
- An isotope depletion and decay calculation for all actinides and selected fission products.

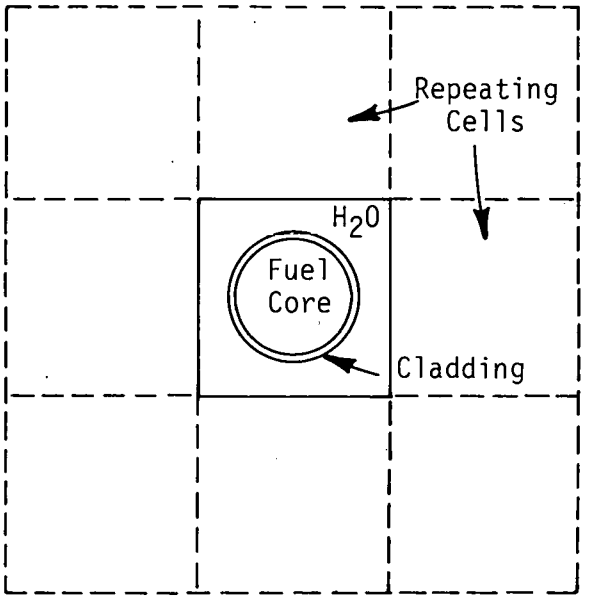
SLIDE 4

GLASS INPUT PARAMETERS

- Isotopic compositions
  - Fuel
  - Cladding
  - H_2O
- Temperatures
- Dimensions
  - Annular
  - Square
  - Hexagonal

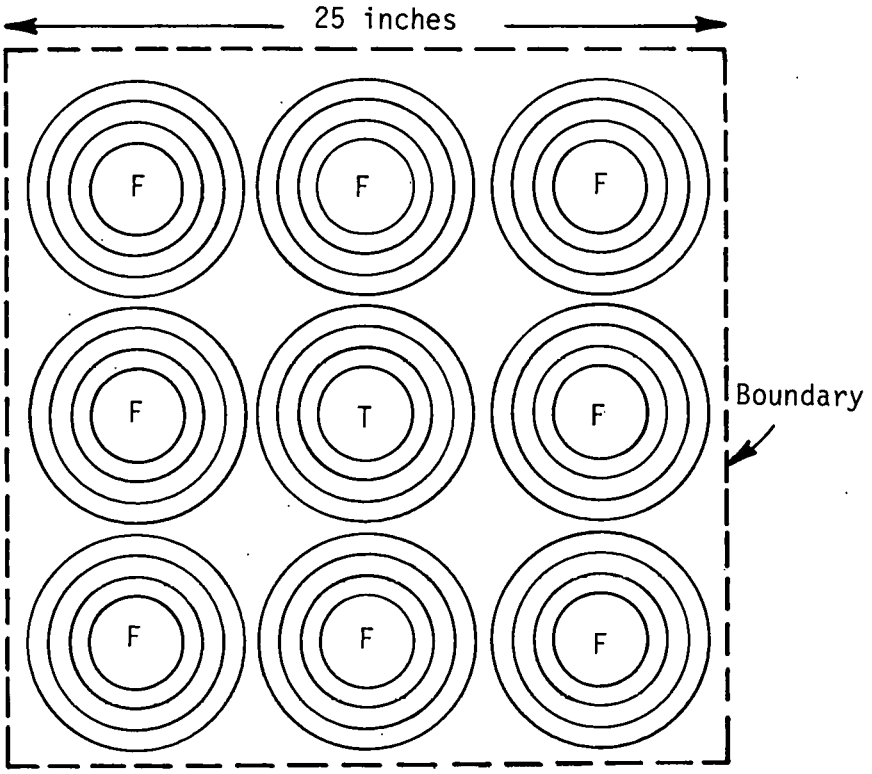
SLIDE 5

TYPICAL LWR PIN CELL



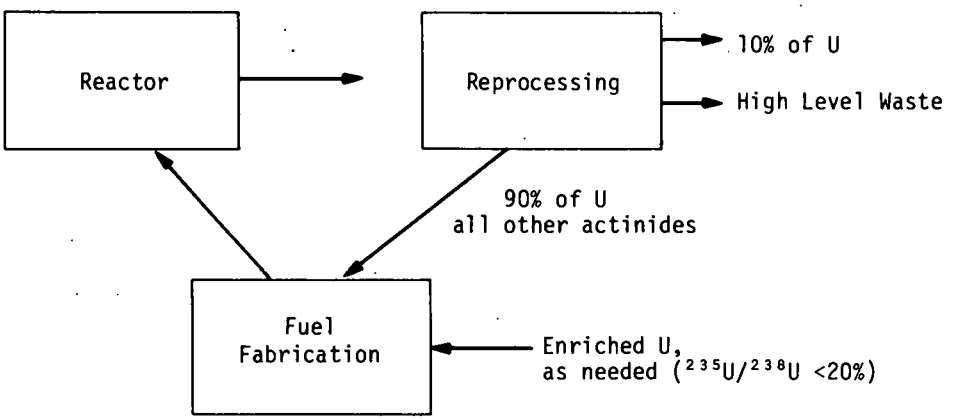
SLIDE 6

9-ASSEMBLY SUPERCELL



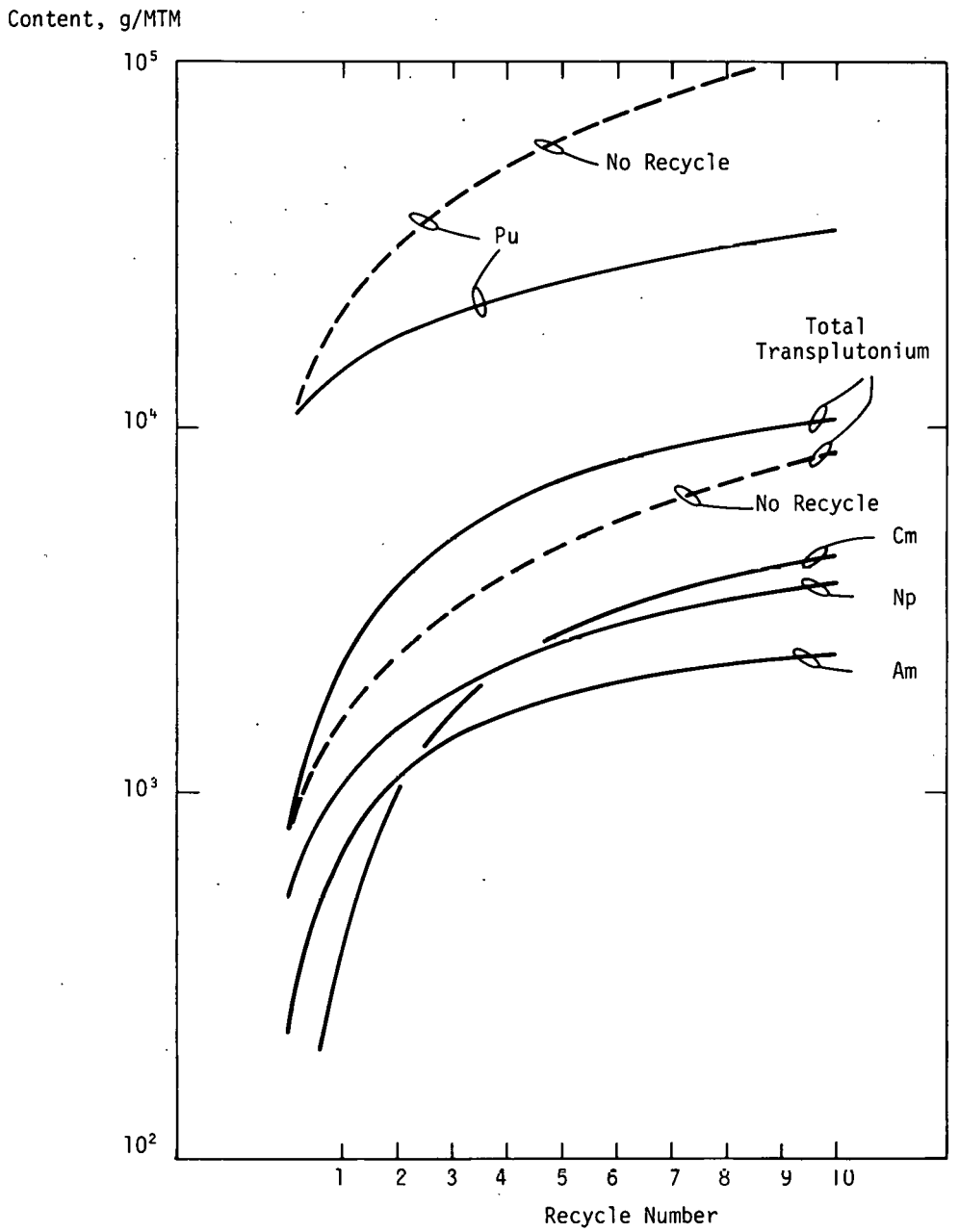
SLIDE 7

Flow Diagram for Recycle Option 1



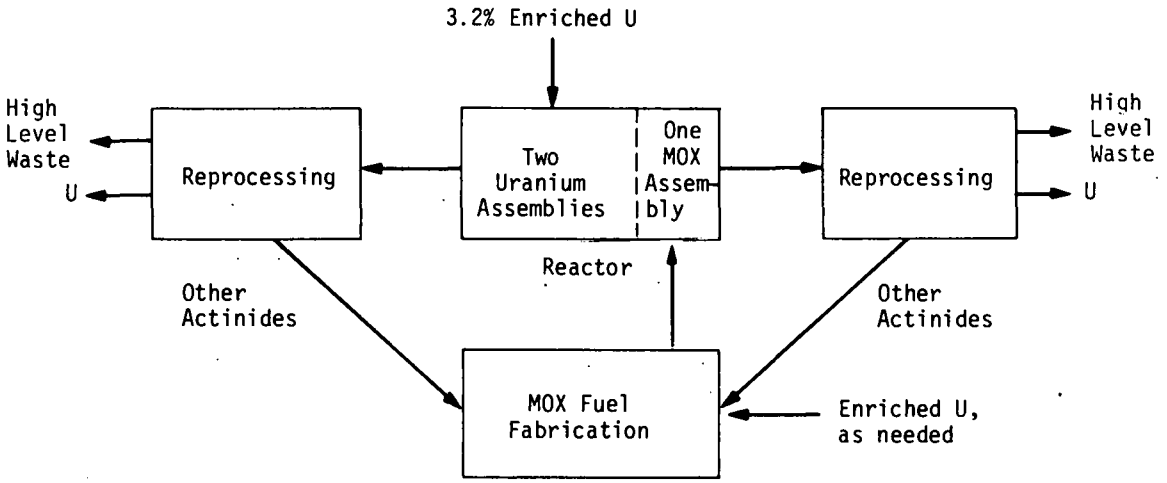
SLIDE 8

Transuranic Content vs. Number of Recycles



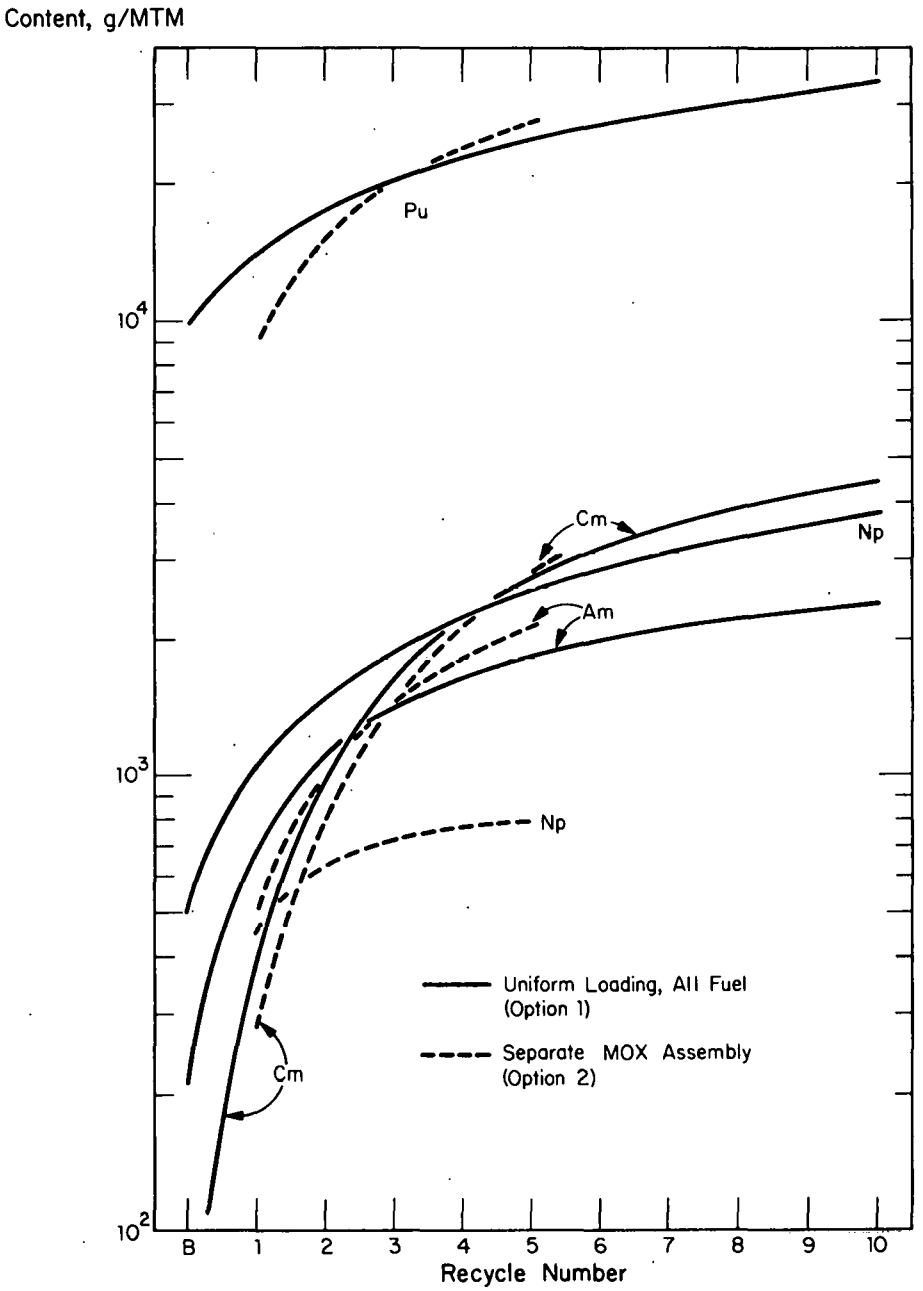
SLIDE 9

Flow Diagram for Recycle Option 2



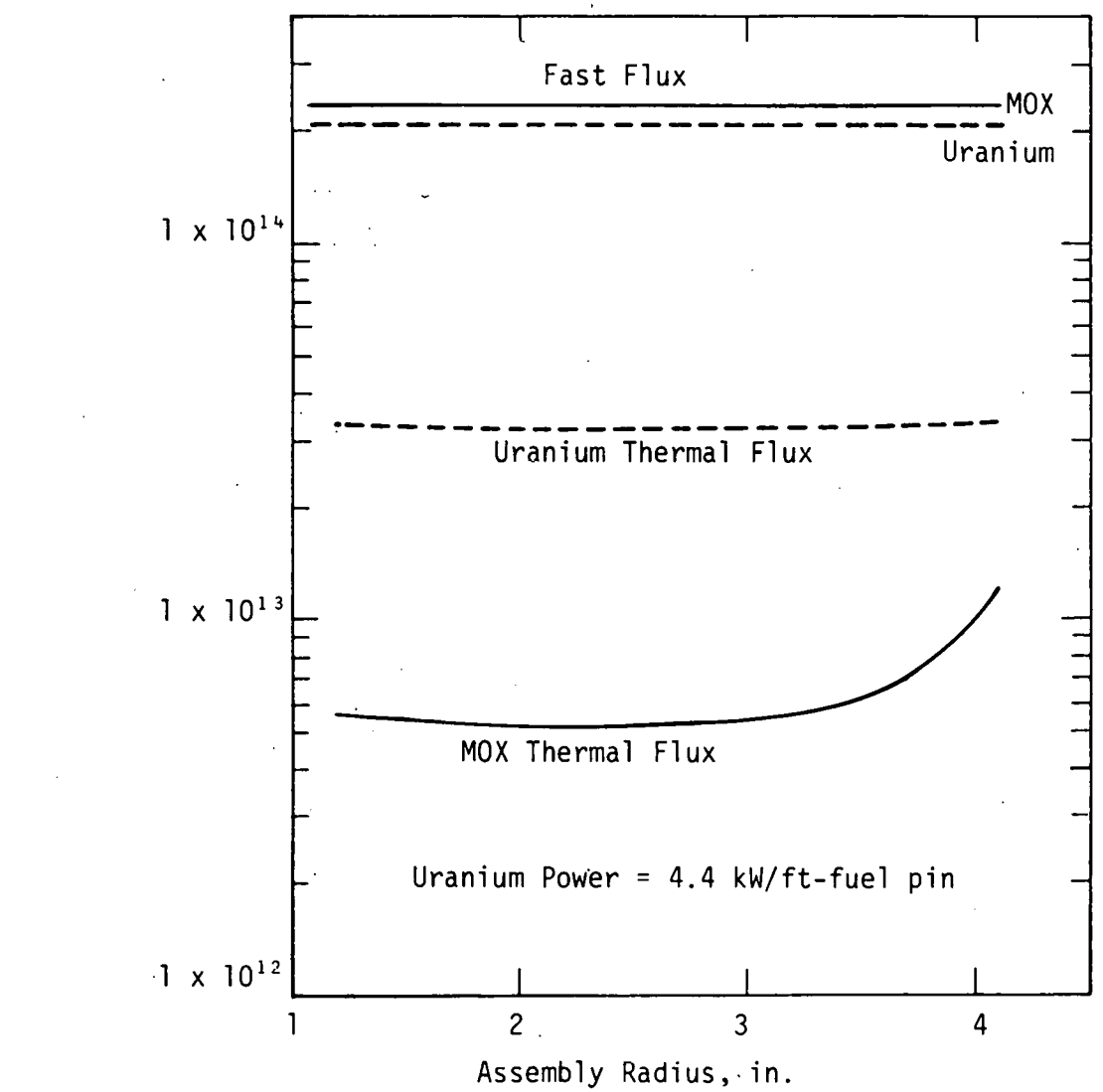
SLIDE 10

Transuranic Content versus Recycle Number



SLIDE 11

Flux Distributions in Uranium and MOX Assemblies



SLIDE 12

Average Pin Power vs. Assembly Radius