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HIGH FLUX PARTICLE BED REACTOR SYSTEMS FOR RAPID TRANSMUTATION OF ACTINIDES AND LONG LIVED FISSION PRODUCTS

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ABSTRACT

An initial assessment of several actinide/LLFP burner concepts based on the Particle Bed Reactor (PBR) is described. The high power density/flux level achievable with the PBR make it an attractive candidate for this application. The PBR based actinide burner concept also possesses a number of safety and economic benefits relative to other reactor based transmutation approaches including a low inventory of radionuclides, and high integrity, coated fuel particles which can withstand extremely high temperatures while retaining virtually all fission products. In addition the reactor also posesses a number of "engineered safety features", which, along with the use of high temperature capable materials further enhance its safety characteristics.

INTRODUCTION

The radioactive waste from commercial and defense related nuclear facilities remains hazardous to humans and the environment for tens of thousands of years, thereby imposing significant burdens on potential waste disposal/long-term storage schemes. The primary contributors to the long term hazards are plutonium, the minor actinides (neptunium, americium and curium), and a few long-lived fission products (e.g., technetium, iodine). Consequently, if these isotopes could be removed from the bulk of the waste, or "neutralized," there would be a significant beneficial impact on the waste disposal/storage problem. In fact, studies have shown that if the Pu and minor actinides were burned/transmuted into stable (or short-lived) isotopes, the remaining waste product would have an ingestion toxicity below that of uranium ore after ~300 years.

Several options have been proposed for the transmutation of radioactive waste, including recycle into commercial reactors, special "burner" reactors, and accelerator based schemes. In this paper an initial assessment of a minor actinide (MA)/long-lived fission product (LLFP) burner concept based on the Particle Bed Reactor (PBR) is considered. A number of configurations are evaluated in terms of criticality, and characteristics related to performance as a waste burner, i.e., flux level, energy spectrum, etc.

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The PBR has neutronic, thermal-hydraulic, and safety characteristics which make it an attractive option to consider for the transmutation/burning of radioactive waste. While this paper focusses on the problem of waste from commercial reactors, a companion paper discusses a PBR based concept for the disposition of weapons grade plutonium.[1] A discussion of some of the safety related aspects of the PBR burner, including minimization of the radioactive inventory (and associated potential source term) and excess reactivity, the use of high temperature materials (to ensure large thermal margins under normal operating conditions), and provisions for "passive" cooling and criticality safety in postulated accident scenarios, is presented in more detail in an accompanying paper.[2] This paper, coupled with References 1 and 2, constitute a fairly complete review of the characteristics and current status of PBR systems for transmutation/burning of radioactive materials.

The PBR Burner concept is based on the PBR nuclear rocket system currently under development by the Air Force Space Nuclear Thermal Propulsion (SNTP) Program and draws on much of the technology that has been developed by the SNTP Program including fuel particles, frits, thermal hydraulics, neutronics, etc. In general, the operating parameters for the PBR Burner (power density, temperature, coolant corrosion, etc.), are much less stressing than those required for SNTP applications, and it is expected that the Burner components will be substantially simpler and easier to develop and qualify.

DESCRIPTION OF THE PARTICLE BED REACTOR

In this section some of the constituents of a conceptual PBR waste burner, i.e., the fuel particle, the fuel elements, and the over-all core design, will be outlined.

Fuel Particle

The basic building blocks of a PBR are the fuel particles. For these studies it was assumed that particles similar to the proven HTGR BISO technology could be utilized. The particles consist of a kernel surrounded by a coating layer of pyrolytic graphite or other appropriate materials to improve fission product retention. The fuel particles (OD ~0.5-1 mm) are directly cooled by pressurized helium gas in the baseline design. The large surface-to-volume ratio of the particles allows high power densities (~5Mw/ℓ), with associated high flux levels (~10^{15}

n/cm²-sec) which are necessary for good performance as a waste burner. Two types of particles would be employed: one containing a plutonium loaded kernel for the "driver" fuel elements, and one containing a minor actinide loaded kernel for the "target" elements. The isotopic distribution of the constituent nuclides in these particles for the present evaluations was based on 33,000 MWD/T burn-up PWR fuel after a decay period of 10-years as given in Reference 3. Based on experimental results and operational experience, these particles should retain their integrity at extremely high temperatures, and ensure large thermal margins under normal operating conditions. In addition, the natural porosity of the particles, coupled with the high density outer coatings, ensure that fission products will be released in significant quantities only if the coatings fail.

Fuel Element

As noted above, two types of elements are envisioned: "driver" elements containing Pu to control criticality, and "target" elements containing the MA or LLFP to be transmuted. The fuel elements consist of a bed made up of the appropriate particles, and constrained between two porous, co-axial cylindrical "frits". Two fuel element configurations are possible corresponding to pressure tube and pressure vessel core options. Figure 1 shows a fuel ement for the pressure vessel configuration. Coolant flow in the PBR is always radial (although it can be in either direction), passing through an inlet "cold" frit, flowing radially through the particle bed, and exiting through the "hot" frit. The bed thickness will depend on a number of variables, including the average bed power density, total reactor power and number of elements.

Zircalloy was selected for both the hot and cold frit material in the current studies since its melting point is sufficiently high to accommodate the maximum operating temperatures for most PBR actinide burners with a reasonable thermal margin. Furthermore, the capture cross section of Zircalloy is low, so its presence will not have an adverse impact on the neutron economy. This is especially important for the PBR burner concept which is designed for a low inventory and excess reactivity.

If a pressure tube is employed, it is also assumed to be Zircalloy. A carbon-carbon liner may be used on the inner surface of the pressure tube to thermally insulate it from the coolant. The liner would also serve to protect the pressure tube from hot fuel particles in the unlikely event of a frit failure.

Pressurized helium has been selected for the baseline coolant because it is both chemically and neutronically benign, and has good heat transfer characteristics. In addition, its previous use in power reactors is valuable for any power generation options based on the PBR burner. Cooling the particles directly with water is also an option.

Core Design

Several potential core designs were evaluated in this study, falling into two general categories: heavy water moderated, and solid moderator systems. Given the basic element design, and the decision to use separate Pu and minor actinide containing elements, the total number of elements and the ratio of driver-to-target, the configuration of the elements, and the moderator material were varied. The major bounding constraints were the desire to minimize the Pu/MA inventory in the reactor and the overall reactor size; therefore the total number of elements, the particle fissile and MA loadings, and the radial reflector were minimized. In general, the candidate configurations represent a compromise between initial excess reactivity, and the flux level and neutron spectra characteristics in the target elements necessary to improve burner performance.

The reactor is constructed of high temperature materials, and posesses a number of engineered safety features which further enhance its safety.[2]

A simplified flowsheet for a PBR Burner based system is shown in Figure 2.

NEUTRONICS METHODOLOGY AND RESULTS

Initial scoping analyses were performed with MCNP4 (for evaluation of initial criticality), and ORIGEN2 (for estimating burner performance) for a number of "seed-blanket" type configurations consisting of Pu-fuelled "driver" elements, and minor-actinide "target" elements. In addition, thermal-hydraulic calculations were performed to estimate particle temperatures, pressure drops, coolant flow characteristics, etc. to demonstrate operating/safety margins.

The MCNP4[4] calculations utilized ENDF/B-V pointwise cross sections with S(α,β) kernels for thermal scattering ott bound nuclei. The geometric modelling for the elements preserved the essential neutronic features of the PBR fuel elements (fuel bed, frits, plena, etc.), and reactor geometry. The flux in the target elements relative to that in the drivers was edited, along with four-group energy dependent spectra. These parameters were used in conjunction with the results from the ORIGEN2 calculations to estimate the changes in the isotopic inventories in the elements with time.

The ORIGEN2[5] program performs a one-group, point reactor depletion calculation accounting for all the relevant isotopic production/destruction chains. Different reactor types/characteristics are accommodated by selecting cross section data appropriate to the configuration being considered. For this study, burnup calculations were performed with the Pu/LWR, and FFTF data sets, thereby bracketing the expected burnup performance for the various configurations; however, this clearly represents an approximation to the true system behaviour.

The baseline reactor core design based on the fuel elements described above takes the shape of a series of hexagonal ring-shaped patterns contained in a moderator volume. This arrangement is shown schematically in Figure 3. The majority of the designs considered here assumed a beryllium carbide (Be_2C) moderator; however, other moderators with low neutron capture properties could be used. The pitch of the hexagonal rings can be varied, changing the neutron energy spectrum. Depending on design, the spectrum can range from thermal to epithermal, and would be chosen to yield optimum performance.

Adjunct calculations were performed with the two-dimensional depletion code 2DB to estimate the variation in core reactivity and burner performance with time.

The results of an initial scoping calculation for a heavy water moderated PBR burner operating at 5 MW/ℓ in the driver fuel, are summarized in Table-1. An ORIGEN2 calculation assuming that the flux level in the target elements equals that in the driver elements, and that a 20-day cycle can be sustained, shows that the burnup of MA is in the range of ~35% to ~60%, with the larger burnup for the FFTF spectrum, assuming a constant power level. However, the flux level associated with the FFTF cross sections is considerably higher than with the Pu/LWR data. Additional calculations were therfore done to isolate the spectrum effect, at a constant flux. Results for a constant flux of 10^{15} n/cm²-sec for the FFTF and Pu/LWR ORIGEN2 data are shown in Figures 4 and 5. Note that the burnup of MA under these conditions is much more effective in a thermal than a fast spectrum. This conclusion is consistent with results obtained by H. Kusters.[6]

The geometries for two solid moderator configurations are given in Figures 3a and 3b. These cores contain ~20% more fuel elements than the configuration considered in Table-1, and either 42 or 43 target elements. The moderator/reflectors in these cases were graphite or beryllium carbide. Some results for the solid moderator/reflector configurations are given in Table-2. Note that the eigenvalue (k-eff) for these systems depends on the pitch and the configuration, with the seed-blanket cores (cf. Fig. 3b) more reactive for the same pitch than the distributed cores (cf. Fig. 3a). In addition, the flux level in the target and the "hardness" of the spectra can vary significantly. While the flux level and spectral "hardness" in the targets is in general inversely proportional to the pitch, thereby reducing the excess reactivity, modification of the moderator surrounding the target elements can somewhat mitigate this effect (Case-1 vs. Case-3).

In order to take into account space-dependent effects on the core k_{∞} and burnup of Pu and MA's, a 2DB calculation was performed for Case-3 of Table-2. The results of that calculation are shown in Figure 6. Note that ~1%Δk of excess reactivity is required to sustain a cycle of approximately two weeks duration. Calculations of this type, eventually expanding to a full three-dimensional model, will be required to define a cycle, and configuration, which optimize burnup performance and cycle length.

TECHNICAL AVAILIBILITY AND R&D REQUIREMENTS

The following technology issues need to be addressed, and in most cases a development program will be necessary to bring the reactor components to a sufficient level of maturity in order to utilize the PBR for waste transmutation.

Moderator

No issues with heavy water. Heavy water is routinely used in a variety of operating reactors, i.e., CANDU, High Flux Beam Reactor (HFBR) and production reactors. In the solid moderator option (baseline) the operating temperature will be approximately 500K in a helium environment. It will be exposed to a neutron fluence of approximately 10^{23}. The nominal design involves beryllium carbide in a graphite structure. There is currently no manufacturing technique for Be₂C components of this size, and one would need to be developed. Other moderators can be used, since the temperature is relatively low. For the case of Be₂C/C, the operating temperature is ~2000 K below its limit.

Cold Frit

The cold frit operates at a temperature of approximately 400K in a helium (baseline) or water environment and is exposed to a neutron fluence of 10^{23}. Several candidate materials can be used in this application. The baseline design would use Zircalloy, which is a well understood material for reactor applications. Other materials include graphite or silicon carbide. The techniques for the manufacture of metallic frits is well understood, and they can be purchased from several vendors. Porous SiC frits would be manufactured by sintering. In the case of graphite a manufacturing process would have to be developed, since on the inlet side, porosity and the flow passages are small.

Fuel

The new particles will consist of two regions, with an inner kernel and an outer impervious coating of pyrographite. This design is similar to the conventional BISO HTGR particle, except that in this case the kernel will be composed primarily of graphite, with a small ad-mixture of Pu, MA or LLFP. The natural porosity of the kernel provides voids for the released fission products. Various options for kernel fabrication exist. An example is the LANL freeze/dry process; other options have also been developed. The outer pyrographite coat will deposited by present conventional CVD techniques.

Hot Frit

The hot frit will operate at the reactor outlet temperature which will be approximately 1000K for the baseline design. In the water cooled option the temperature will be approximately 600K. A hot frit is characterized by having a relatively high porosity, in order to minimize the pressure drop. In the case of a water cooled reactor a simple drilled zircalloy tube would act as the hot frit. The holes in the tube would have to be smaller than the particle diameter. In the case of the helium cooled option a higher temperature material is required. The baseline design assumes a drilled Inconel tube. Such techniques have been used to make hot frits for the SNTP program. The cost is low, and quality excellent. However either graphite, carbon/carbon or silicon carbide would be perfectly feasible and should present no manufacturing problems. In summary, none of the candidate designs for the hot frit should be difficult to manufacture.

Inlet and Outlet Plenum Grid Plate Structure

Incalloy-Be₂C structure needs to be developed. Prototype structure needs to be tested at operating temperature. It is felt that manufacturing the types of structure envisioned here will not be difficult.

Pressure Vessel

Needs to be mechanically and structurally strong at inlet temperatures. No mechanical or developmental problems are expected for this component.

Long-term Fuel Integrated Element Test to Determine Material Compatibility

Two issues need to be addressed in the development of an integrated fuel element. First, the loading and unloading of particulate fuel by hydraulic means needs to be confirmed. Second, fluid dynamics, heat transfer and material compatibility experiments need to be carried out for a integrated element. Initially the material compatibility tests will be carried out on test coupons in specially designed furnaces.

The first set of fuel element tests can be carried out using electrical heating. Subsequent tests would be carried out in an appropriate test reactor. The final tests at target power densities will be a challenge for all existing test reactors, since a substantial flux will be required to drive an element to the 5-10 MW/liter target range.

Frit Clogging and Particle Erosion

An experimental program needs to be designed and carried out to ensure acceptable fuel stability and frit porosity. An initial experiment carried out at Brookhaven National Laboratory using pyrolytic graphite-coated particles contained between commercially available steel frits in a helium loop showed acceptable results. No clogging of either frit was observed for the duration of this experiment.

Hot Frit Failure

In the case of a hot frit failure, it might be desirable to prevent particles and hot frit parts from entering the balance of plant. A possible remedy to this difficulty is to use low pressure drop filters at the end of each hot frit. This filter will have to be made of hot frit material to withstand the outlet temperature. Furthermore, the porosity and area will have to be large enough to prevent any large pressure drop from developing. Finally, it should be pointed out that in the event of a hot frit failure and the effective loss of a fuel element, the reactor might be subcritical and automatically shut down.

SUMMARY AND CONCLUSIONS

An initial assessment of several actinide burner concepts based on the Particle Bed Reactor has been performed. Configurations based on separate Pu fuelled driver and minor actinide loaded target elements, and moderated by heavy water or beryllium carbide, have been evaluated in terms of initial criticality and anticipated transmutation performance. The results show that varying the number and mix of driver and target elements, the moderator, and the core configuration can modify the reactor characteristics (e.g., k-eff, flux levels and spectra) so as to improve the expected performance as an actinide burner. The configuration of Table-1, for example, was configured so as to yield an ~epithermal spectrum in the target elements, with a flux level close to that in the drivers. In this configuration ~ 50 % of the actinides in the target elements are burned in 20 days. The ability to sustain such a cycle, however, and the details of the needed initial excess reactivity / assumed refueling scenario, require further study, and may require some compromises relative to the particle or element loadings. While these have been deliberately kept low to minimize the inventory of radionuclides, only a modest increase may be sufficient to sustain a reasonable cycle length.

The PBR based actinide burner concept also possesses a number of safety, and potential economic benefits relative to other reactor based transmutation approaches. Among these are a low inventory of radionuclides (~5-10% of that in a commercial LWR), and high integrity, coated fuel particles which can withstand extremely high temperatures, while still retaining virtually all fission products. This ensures large thermal margins under normal operating conditions, and minimizes the potential source term in postulated accidents. In addition, the pressure tube design, and the possibility of on-line refueling offer further potential safety and economic advantages. Finally, the helium coolant can be used for power generation.

While the above results suggest that the PBR is an attractive concept for further study as an actinide/ long-lived fission product burner, a number of technical issues require attention. These include:

- The desired actinide loading of HTGR-type fuel particles, and their subsequent performance must be demonstrated; however, the available operational experience with "conventional" particles vis a vis fission product retention, and integrity at high temperatures, is favorable.

- While operation of PBR elements at the desired 5 MW/l has been demonstrated in blow-down experiments, experience with a nuclear heated fuel element for high power-long term operation is needed.

- Hydraulic loading/unloading of particles from PBR elements has been successfully achieved in an experimental mock-up, but further work is necessary, particularly if on-line refuelling is required.

- The high flux levels associated with the PBR concept require that extra attention be devoted to the consideration of radiation damage/structural integrity issues, particularly for the pressure tubes and solid moderator.

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Table 1 Selected Characteristics of Heavy Water Moderated PBR Burner

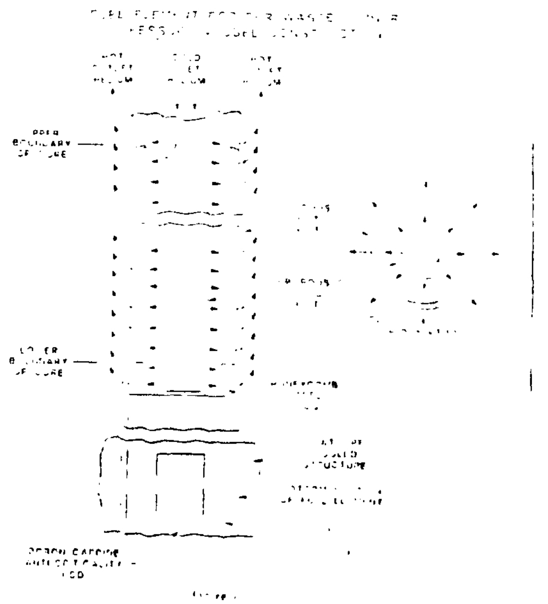
Moderator	D_2O	
Number of Pu Driver Elements	72	
Number of MA Target Elements	42	
Power Level with Driver Elements @ 5 MW/ℓ	1080	
K_{eff} (Clean)	1 040±0 005	
(Average Flux)$_{target}$/(Average Flux)$_{driver}$	0 86	
Target (Flux E>5 5KeV/Total Flux)	0 39	
Initial Loadings, Kg		
Plutonium	30 0	
Neptunium	7 3	
Americium	9 9	
Curium	0 3	
Total Minor Actinides	17 5	
Final Loadings after 20 days @ Φ(driver)=Φ(target), Kg		
Plutonium	8 77 (1)	9 86 (2)
Neptunium	2 46	1 64
Americium	1 54	2 10
Curium	6 37	3 45
Total Minor Actinides	11 19	7 17

(1) Pu/LWR Spectrum in ORIGEN2
(2) FFTF Spectrum in ORIGEN2

Table 2 Selected Results for Solid Moderator Options

	Configuration	K [illegible]	[illegible]	[illegible]
1	84 Driver-43-Target, Be-C Moderator Reflector, pitch = 20 cm, Fig 2a	[illegible]	0 [illegible]	
2	Same as (1), pitch = 15 cm	1 [illegible]	[illegible]	
3	Same as (1), graphite around target elements	[illegible] 051	0 78	[illegible]
4.	85 Driver/42-Target, Be-C Moderator/Reflector; pitch = 20 cm, Fig 2b	1 335	0 44	[illegible]
5	Same as (4), pitch = 15 cm	[illegible] 031	0 54	[illegible]
6	Same as (4), graphite axial radial reflectors	[illegible]	[illegible]	[illegible]

* Fractional standard deviation ±0 005
(1) Φ_T = Flux in Target, Φ_D = Flux in Driver
(2) Target Φ (E > 5.5 KeV) Φ (Total)



SIMPLIFIED OVERALL FLOW SHEET
FOR NUCLEAR WASTE BURNING

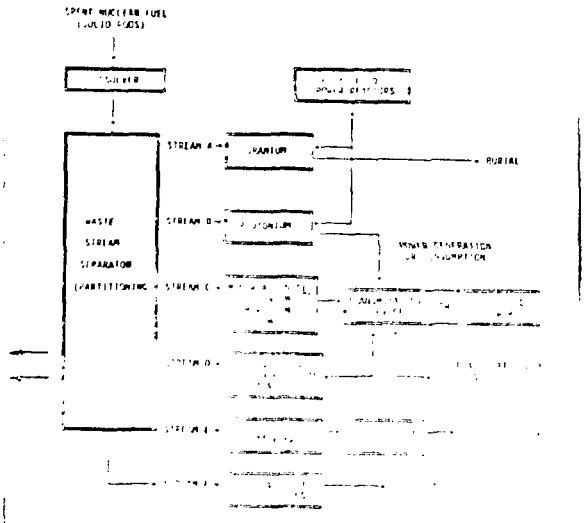
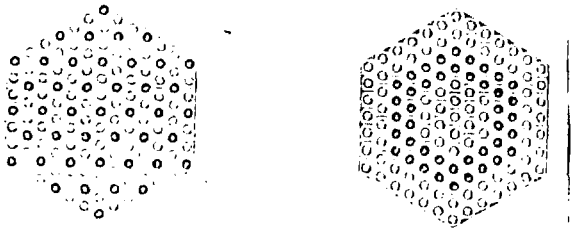


Figure 2

SOLID MODERATOR CORE CONFIGURATIONS



CONFIGURATION A

CONFIGURATION B

Figure 3

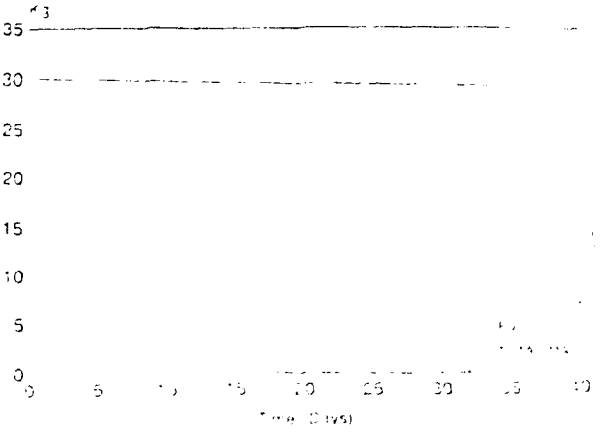


Figure 4 [illegible] in FBTR Spectrum in ORIGEN2
Flux = 1.0E+16 n/cm2-sec

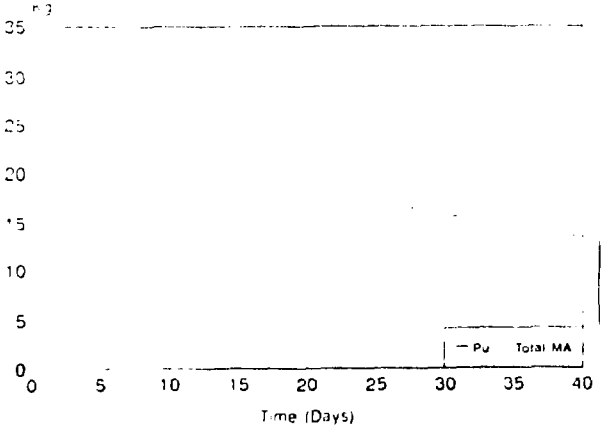
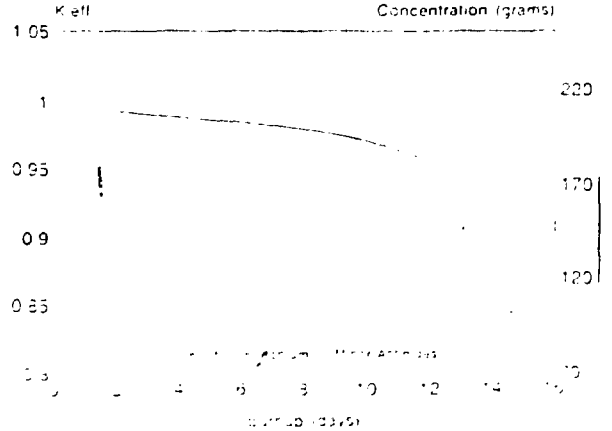


Figure 5 PBR Actinide Burner with Pu/LWR Spectrum in ORIGEN2
Flux = 1.0E+16 n/cm2-sec



[illegible] PBR Actinide Burner K-eff and Pu and Actinide Concentrations with Burnup