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STUDIES ON THE PROPERTIES OF HARD-SPECTRUM,

ACTINIDE FISSIONING REACTORS

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Final Report

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INTRODUCTION

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Idealized practice in any industry is to produce minimum effluents--to consume as much of one's own waste as is possible and to render what cannot be consumed as nearly innocuous as possible. Our initial motive in undertaking our studies was to examine possibilities for utilizing "waste" actinides from the nuclear industry within that industry. Since all such nuclides are fissionable, with effective thresholds for fission in the few hundred kev range, it occurred to us that they are in principle usable nuclear fuels for hard-spectrum reactors. Such actinide burning not only would recover useful values from these waste materials, but could also serve to render innocuous some of the more troublesome actinides. An example is ^{241}Am , whose half-life is long enough so that its hazards would persist after most fission product activities have decayed away, yet short enough so that it is intensely radioactive.

This opportunity does not present itself in reactors of the standard fast breeder type, such as LMFBF's of the PHENIX or Clinch River type. These reactors have median fission energies in the tens of kilovolts range, and consequently actinides do not fission directly, but only after neutron captures--which may be multiple--lead to a fissile or spontaneously fissioning nuclide.

However, hard-spectrum reactors exist and have been operated. The best known reactor of this type that is still operating is the EBR-II. Such reactors differ from conventional LMFBF concepts in having relatively low concentrations of fertile material and high concentrations of fissile material in their cores. The low fertile material concentration reduces neutron moderation by inelastic scattering, while the high fissile material concentration permits absorption of neutrons before many elastic collisions with medium-weight nuclides have taken place.

Favorable properties of such reactors include very high fissile material conversion ratios, small and mostly negative sodium void coefficients, and capability of structural design to exhibit large negative power coefficients. Unfavorable properties are low specific powers for given thermohydraulic conditions, small critical sizes, the essential absence of Doppler effect, and the need to achieve most of the conversion in external blankets. The drawbacks of small core size and external conversion can, however, be much mitigated by using a modular or parfait core design, which amounts to assembling a reactor that consists of several weakly coupled, subcritical cores, while the varying advantages and disadvantages with respect to reactor safety are generally compensating.

Low specific power remains an economic disadvantage of this reactor type. However, in the present era, the preferred fuel, plutonium, is a glut on the market and may be of negative value: that is, a good case can be made for maintaining large in-reactor inventories of plutonium as a cheap method of safeguarding it. Hard spectrum reactors show

good promise as converters of LWR plutonium to more usable fissile materials (1). For this use, the important criterion is high conversion rate rather than doubling time. One wishes to move blanket material, rather than core material, rapidly through the reactor.

These comments represent the context in which our studies of actinide-burning reactors were undertaken. The studies are specifically directed toward the possibility of reducing effluent hazards, but the reactor would be more of a curiosity than a concept if it did not have other potential merits.

ACTINIDE BURNER REACTOR (ABR) SYSTEMS

Previous studies (2-7) have investigated the feasibility of fissioning actinides in LWR's, LMFBF's, and even fusion devices. Because of the high energy fission thresholds of most of the actinides, the neutron spectra of both LWR's and LMFBF's are too soft to fission an appreciable quantity of the actinides. Placing such wastes in these reactors necessitates long residence times with an accompanying increase in production of higher actinides.

We have therefore used a totally different design philosophy. For burning actinides, a reactor should have the hardest possible neutron spectrum in order to produce the highest achievable actinide fission rate. We refer to such a reactor as an Actinide Burning Reactor (ABR). An ABR should, by virtue of its hard spectrum, and the high fission-to-capture cross section ratios of most actinides at high neutron energies, exhibit decreased production of new actinides in burner fuel as well as higher relative fission rates for the input wastes.

In addition, and in common with other actinide burning proposals, a high neutron flux is desirable, to decrease the system half-life of the actinides (i.e., the mean time for burning an atom) to the few-year value characteristic of most fission products.

This concept could be used in an energy park based on integrated fuel cycle facilities for a large number of power reactors. This application would minimize transport of radioactive wastes from the site and reduce the long-term hazard associated with any waste materials which do leave the site.

The actinide waste for the burner is assumed to consist of all those actinide nuclides that are not recoverable economically as "clean fuel" in the standard chemical processing steps. This includes some small fraction of the uranium and plutonium reprocessed, plus all neptunium and trans-plutonium elements.

The long-range goal is a fuel cycle and fuel management concept for an energy park utilizing one or more reactors whose function is, among others, to burn actinide wastes. To this end, our work has been to determine the feasibility of the burner concept and develop a preliminary reactor design based on the following criteria:

1. The object of the burner is, at equilibrium, to consume waste actinides at a rate equal to the waste actinide production rate from the LWR's, from such LMFBR's as are associated with them in the system fuel cycle, and in the burner itself. At equilibrium, transuranic waste inventory (in kg) in the park will be constant.

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2. The burner will initially be fueled with uranium, plutonium, or a mix, and as actinides become available, this conventional fuel will be replaced with fuel pins fabricated from actinide wastes.

Questions which our investigation has covered are:

1. Can a burner accept actinide wastes and maintain criticality throughout the operating life for various compositions of uranium and/or plutonium fuel and the included wastes?

2. Can a burner dispose of actinides at a rate which will establish an equilibrium for a park such that the actinide inventory can be maintained constant?

3. Once a preliminary design is determined, what will the equilibrium condition be? What will be the core composition for the condition of constant park actinide inventory?

4. How will the core composition vary during the period prior to equilibrium?

5. Since actinides will be loaded as an increasing segment of the burner core, at some point the burner will transit from being a net producer of actinide wastes to a net burner. How much operation and what inventory is required to reach this point?

6. What are the values (or even the signs; i.e., $\pm$) of the reactivity parameters associated with safety and control of ABR's?

To answer these questions, a program, NCINR8 (8, 9) has been developed to perform regional spectrum, one-dimensional diffusion, and burnup calculations. Energy spectrum and leakage are recomputed periodically during the burn, and core actinide composition is followed during a thousand year decay. Inventory is expressed in kg. A 29-group set of shielded cross sections from ENDF/B-V in the Bondarenko format is the primary data source (10-14).

Using pin-cells modeled after either EBR-II, a "typical" LMFBR of the PHENIX or Clinch River fuel design, or a GCFR, several assemblies of reprocessed actinides have been analyzed. Some preliminary conclusions which were drawn from our early results are:

1. A critical reactor fueled only with recovered wastes (all Np, Am, Cm, etc., plus 0.1% of the U and Pu in spent LWR fuel may be constructed. Due to a large fraction of the core being ^{238}U , this reactor acts as a breeder.

2. Further reprocessing of the input wastes to decrease the

uranium component significantly (by about two orders of magnitude) reduces the critical size of the burner and hardens its spectrum.

3. In order to achieve maximum relative burnup rate of the actinides it is necessary to use a diluent. This permits the required high fluxes while not exceeding a reasonable power density. EON PAGE 1

4. Key isotopes in the analysis are ^{238}U and ^{241}Am . ^{238}U significantly degrades the neutron spectrum. ^{241}Am provides approximately 90% of the total actinide hazard at 1000 years. Hence, reprocessing should concentrate on removing as much U as possible from the actinide fraction, subsequent to partitioning of actinides from FP. This would permit the most effective destruction of ^{241}Am .

Analysis of the reactor designs has demonstrated that the actinides may be loaded as metal, metal oxide, or metal carbide, and form critical configurations. All of the actinide configurations that we explored, except those using 50 w/o molybdenum as a diluent, had k_{∞} values greater than 1. However, the burnup rate of the actinide wastes is significantly improved by dilution with molybdenum, since higher fluxes may be attained.

Since k_{∞} for the diluted actinide mix is below 1, but its fractional burnup rate is by far the best, the idea of a driver-driven region reactor was studied. In this concept a central region composed of an actinide mix is surrounded by an appropriate fast reactor driver. EBR-II provides an excellent starting example of a suitable driver.

Most of the calculations reported here are for the same basic two-region core. The central, 5 cm radius region contains actinides in many different configurations. These consist of actinide metal, metal diluted with molybdenum, actinide oxide, and actinide carbide. The four different compositions were arranged in three different pin-cell dimensions. The isotopic composition of Mo-diluted actinide material and pin-cell configurations of the three reactor types are given in Tables I and II.

The driver region was modeled after EBR-II. The composition and standard dimensions of this driver region are given in Table III. Most of the calculations reported here for driver actinide systems used this driver as a bare reactor, without external reflector or blanket. In general, the reactor was not adjusted to be critical, but each loading had its k_{eff} calculated.

As will be shown later in this paper, for small driven actinide regions the spectrum and flux level in the actinides is completely dominated by the driver region. Thus, it is the driver region that must be optimized to burn actinides effectively in a two zone core.

This leads us to two different types of ABR's for study; the all actinide reactor and the driver region actinide burner.

The characteristics of the first are determined only by the actinides while the characteristics of the second are independent of them. Thus, our work has followed two paths. Both of these are reported here.

The advantage of the actinide burner was initially assumed to be its ability to burn the most hazardous actinides. Thus, the actinide burner could eliminate the necessity for disposing of the transuranic isotopes. As a result, the radioactive waste disposal problem would be limited to isolation of fission products from the environment for a few hundred years.

SPECTRA

We have stated that a hard spectrum is a necessity for fissioning the more troublesome actinide nuclides directly and preventing the formation of still higher actinides. Figure 1 illustrates this point. In Figure 1, we exhibit the fission and capture cross sections of ^{241}Am , which is both a troublesome nuclide and one whose cross section behavior is typical of the class. With these cross section curves are plotted the multi-group spectrum of a large LMFBFR and of the driver region of the reactor (adapted from the original EBR-II design) that we have used as a driver system for an actinide target region. The large LMFBFR has a major flux component below the effective fission threshold of ^{241}Am , and reacts with that nuclide primarily by capture. In contrast, our driver spectrum exhibits very little flux at low energies and a more typical ^{241}Am reaction is fission.

Figures 2, 3, 4, and 5 display the spectra of infinite medium, all-actinide cores made of various possible materials. These are all for a geometric pin design similar to that of a large LMFBFR. The spectra are hardly affected by the pin design, and the "EBR-II" and "GCFFBR" pin designs lead to very similar results. Again, the ^{241}Am fission and capture cross sections are drawn onto these curves for reference. The GCFFBR pin configuration has slightly harder spectra due to the absence of sodium, but the difference is small. Of the all-actinide cores, the one with metal fuel exhibited the hardest spectrum, but is possibly not practical to build. The one with carbide was the next hardest, and with oxide the Mo-diluted metal the spectra were softest. The large dip in the oxide spectrum at 350 keV is due to the large elastic-scattering cross section of oxygen at that energy.

In order to simplify our interpretation, we have examined ways of describing the spectrum to get some measures of spectrum effectiveness for fissioning actinides. The first way chosen was to describe the spectrum in terms of a single number, the average neutron energy. This technique was used since it describes the reaction rate, approximately, if it is assumed that cross sections are approximately proportional to $1/\sqrt{E}$. This result is seen in Equation 1.

$$\sigma(E) \phi(E) = (\sigma_0(E)/\sqrt{E})(\sqrt{E} n(E)) = c n(E) \quad (1)$$

where $\phi(E)$ is the energy dependent flux,

$$n(E)$$
 is the energy dependent neutron density

E is the energy

$\sigma(E)$ is energy dependent cross section

$\sigma_0(E)$ is cross section at a fixed energy

Although actinide fission cross sections bear no resemblance to $1/\sqrt{E}$ behavior, capture cross sections do. Thus, mean energy,

$$\bar{E} = \frac{\int E n(E) dE}{\int n(E) dE} \quad (2)$$

measures the "reluctance" of the actinides in the system to capture neutrons.

Other measures that are possible include integral spectral cross sections of actinides. Finally, since k_{∞} of an all-actinide system is a sensitive measure of fission-to-capture ratio, this, too, was examined.

Figures 6 and 7 show cross correlations of several of these measures. k_{∞} , the average fission cross section of the given actinide mix, and the fission-to-capture ratio of that mix are plotted against $\bar{E}$ for a number of systems whose actinide mixes are the same. The actinide concentrations are given in Table IV for the two mixes used.

The results show an almost linear improvement of each of these parameters with $\bar{E}$. The two cases which fall below the general correlation (between $\bar{E} = 0.2$ and 0.25 MeV) are cases of molybdenum diluted metal systems, for which specific inelastic effects in Mo may be responsible. In general, it appears that $\bar{E}$ is a valid spectral criterion. The individual points in Figures 6 and 7 are for oxide, carbide, actinide metal and Mo-diluted metal, for the three different pin designs looked at, in variable infinite lattice configurations. The points to the right, which make for the best actinide burning, are for actinide metal systems. It appears that $\bar{E} = .3$ MeV or more is most desirable for efficient actinide burning.

PRELIMINARY CYCLE STUDIES FOR INFINITE MEDIUM ABR'S

For each of the reactor configurations given in Table II, four fuel compositions were burned over a three-year period at a constant power density of 600 kW/l. The four fuel compositions were pure actinide metal, actinide diluted with 50 w/o molybdenum, "actinide oxide" (two oxygen atoms for each actinide), and "actinide carbide" (one carbon for each actinide).

Table V shows the calculated values of k_{∞} as a function of exposure for five of the cases. The first four are different fuel compositions in an EBR-II configuration. The last column is actinide metal in GCFR configuration. With the exception of the 50 w/o Mo fuel, all are critical at the beginning of the cycle. In every case k_{∞} increases with exposure. The reason for this appears to be that as the americiums burn out ^{239}Pu and higher actinides are produced, which make the fuel "better".

All three of the reactor configurations show the same general behavior. Thus, we will discuss only the EBR-II pin-cell configuration.

Figures 8 through 11 show the fraction of ^{241}Am and ^{243}Am burned

in each case as a function of exposure. (See description on page 1)

The pure actinide metal case burns only about 20% of the ^{241}Am . By adding molybdenum as a diluent to 50 w/o, about 60% of the ^{241}Am is consumed (9, 15). This shows the importance of a diluent. It allows much higher fluxes at the same power density.

There is an apparent paradox between the oxide and carbide cases. One would expect that with two oxygen atoms present instead of one carbon atom per actinide that the americium would burn slower in the softer spectrum. However, this effect is more than offset by the higher fluxes in the oxide case. What happens is that in the oxygen case the americium is more dilute.

One should not make too much of the dilution effect, however. While it is true that, for constant power density, high dilution implies higher flux and higher destruction rate per atom loaded, more concentrated actinides would actually exhibit a higher rate of actinide destruction by the measure of grams destroyed per unit power. Indeed, for a pure actinide-fueled reactor, one would expect the largest amount of actinide burning to be accomplished in that core which had the highest actinide concentration.

These studies indicated to us that a driver-driven system would be a more likely initial venture for actinide burning. At the start, one would have a reactor with a small central actinide region. As the nuclear industry provides more actinides, the central region grows. However, it might take quite a while before the actinide region becomes most of the core. For these reasons, we have performed most of our studies on the basis of driver-driven ABR's.

ACTINIDE RECYCLE STUDIES--DRIVEN SYSTEMS

Steady-state recycle of actinides through a driver-driven ABR was studied. Original actinide fuel was inserted into the central region for a three-year period, removed, cooled for two years, reprocessed and reconstituted by one of two different methods, and reinserted into the central region. This process was repeated through ten cycles, so that the in-core time period was 30 years. This arbitrary number was used only to study trends, as theoretically the actinides could be recycled indefinitely. In this study, fuel of only one cycle was in a given reactor for each three-year burn, although a mixture of generations would most likely be used. Results for such a mixture could be obtained by averaging the results of different cycles in accordance with their respective constitutional ratios and quantities at steady state.

The original actinide fuel was the discharge of an LWR, reprocessed after a two-year cooling period with 99.9% of the Pu and U removed. Two methods of reprocessing were followed through subsequent cycles. In the first, the levels of Pu and U were reset after the cooling period to the levels of the first cycle. In other words, it was assumed that the Pu and U could be removed in future cycles so that they constituted

the same percentage as at the start of the first cycle. In the second approach, 90% of the Pu was removed at the end of each cooling period, but the U remained with the other actinides. The Pu level at the start of each cycle was never less than that of the first cycle. The first method mentioned is probably more realistic, as most common processing techniques remove U and Pu simultaneously; and indeed U is the easiest of the actinides to separate from the others. In both methods, it was assumed that all the fission products were removed.

Because the type of fuel used proved to have little effect on burn rates, the first reprocessing method was studied only with oxide fuel at a central flux level of 3.2 to 4.1×10^{15} n/cm²-sec (135 MW of thermal power for the entire core). Three different types of fuel at two different power levels were used with the second reprocessing approach. The types of fuel used were oxides, carbides, and metal. The lower power level used was that just listed for the calculations using the first reprocessing approach; the higher power level was 150-200 MW, varying to give a constant flux of 5.0×10^{15} n/cm²-sec. This higher flux level resulted in a peak power density of about 1900 kW/l, excessively high for an EBR-II type reactor. The limiting condition, however, is not in the actinide region, but rather at interface of the driver region. The power density in the actinide region was only 400 kW/l for oxide fuel, well below the 1400 kW/l design capability of the EBR-II. The high flux was studied with the idea that perhaps the driver region could be tailored to obtain a flux of this magnitude. One hundred thirty five MW results in a peak power density of 1240 kW/l, about 10% below the design capability.

The ABR is designed so that the spectrum to which the actinides are subjected is determined by the driver region. Figure 1 has already shown the beginning of cycle (BOC) spectrum of the first cycle for each fuel type at 135 MW. The spectra are nearly independent of the fuel type particularly at the high energies with which we are concerned (at lethargy unit 7, $E = 9.1$ keV). The driver effectively controls the spectrum.

Figure 12 shows the BOC spectrum for the 10th cycle, carbide fuel, according to the second reprocessing mode. The actinide region here contains a much higher percentage of ²³⁸U than the first cycle, but again, the two spectra are nearly identical.

Figure 13 shows the end-of-cycle (EOC) spectrum for carbide fuel, first cycle. A softening of the spectrum occurs as fission products build in, and, of course, this effect is greater at the higher flux. Nonetheless, the spectrum is quite constant at the higher energies over the three year burn. In actual practice, fuel rods are replaced every 12-18 months, and this would result in an even more constant spectrum. With this as a justification, several cycles were simulated by inputting a representative flux which was held constant through the three year burn in order to save computer time and money. This resulted in a change of less than 3.6% in the burn rates of the major isotopes.

All the runs discussed to this point were done without the

surrounding blanket regions that breeder reactors contain. An NCINR8 run was performed which included a blanket of 25,000 kg of ^{238}U . Although the radial flux profile was altered increasingly towards the outer edges of the driver region, the actinide region was by-and-large unaffected. The spectrum of the actinide region when the driver has a surrounding blanket is shown in Figure 14. The results justify the exclusion of the blanket region to determine the burn rates in the actinide region, although the rate that actinides build into the driver region is affected.

The radial flux profile for the first four groups of a collapsed 9-group structure is shown in Figure 15. The flux is somewhat depressed in the actinide region. This is due to the relatively small amounts of fissionable material, so that the source of fission neutrons is depressed.

An inherent flaw in the NCINR8 code is that it fails to account for axial flux and spectrum variations. Since the flux decreases and the spectrum softens near the top and bottom of the core, slower burn rates would be expected in these regions. Thus, the burn rates that would be expected in these regions. Thus, the burn rates that are presented in this paper are representative only of the central region of the core.

During the course of the three-year burn, the driver region becomes less enriched as the fuel burns out. To maintain a constant power, the flux must be increased to compensate for the loss of fuel. In the actinide region, the flux typically increases from 3.2 to 4.1×10^{15} n/cm² - sec at 135 MWth. This should result in an increased burn rate late in the cycle, depending on the competing affect of the softer spectrum. By calculation, we found that the EOC burn rate in the actinide zone was not much different from that at BOC.

The following figures graphically show the results of the burns. Figure 16 shows the remaining quantities of the major isotopes from an original 10 kg sample of actinides in the form of oxide at the end of each five year cycle (three-year burn + two year decay) at 135 MW. Virtually no difference exists between the three fuel types (because of the near-identical spectra). This means that the type of fuel can be determined by factors such as thermal considerations, compatibility, manufacturing ease, etc., rather than burn rates. The number density of actinides, however, is greatest in the metal fuel (followed by the carbide fuel), and the quantity of actinides burned is directly proportional to this.

Essentially the same results are obtained using the first reprocessing scheme. This is somewhat unexpected in that if uranium is not removed, the ^{238}U builds up to 93% of the fuel by the 10th cycle due to its relatively slow transmutation rate. This makes for a good breeder, but is not germane to the main purpose of the ABR. Table VI shows the different isotopic constitution per cycle depending on which reprocessing scheme is followed. Although ^{238}U actually depletes during each cycle, it burns out at a slower rate than the other actinides, and therefore, if not removed, constitutes a greater proportion with each cycle. However, this apparently makes no difference in the rate

that the transplutronics burn out. Again, this latter is a function chiefly of driver flux and spectrum.

In fact, the results indicate that two nearly independent events are taking place. The first is the conversion of U to Pu, and the second is the fissioning of the higher actinides (transplutronics). In other words, while some U is converted to Pu, very little buildup occurs past ^{242}Pu . The major transplutronics, such as ^{241}Am and ^{243}Am burn out independently of the U and with very little buildup of Cm, Bk, or Cf. The actinide region serves as both a breeder and a burner. To confirm this, runs were performed where exclusively ^{238}U and exclusively ^{239}Pu were subjected to the constant flux discussed previously. With 10 kg of ^{238}U initially in the central region, 355 grams of ^{239}Pu built in after three years or 3.55% of the original ^{238}U initially loaded, .55 grams of ^{241}Am built in, but only traces of higher actinides. Of the original 1,000 grams, 5,387 grams of ^{239}Pu remained, (along with 375 g of ^{240}Pu , 11 g of ^{241}Pu and .3 g of ^{242}Pu); the rest had fissioned. This explains why the same percentage of major actinides burned out on every run, regardless of the initial loading of U and/or Pu. As seen in Table VII, the amount of ^{239}Pu that built in after each three year burn at 135 MW, assuming that only 53.9% of the original ^{239}Pu remained, was 3.5 to 3.6% of the BOC ^{238}U amount. The slight differences can be ascribed to small spectra variations and initial loadings of ^{237}Np and other isotopes of U and Pu.

In terms of the burning of the actinides, each five year cycle saw the destruction of approximately 36% ^{241}Am , 23% ^{243}Am , and 34% ^{237}Np as seen in Table VIII. In addition, approximately 21% ^{244}Cm disappeared but this was mostly due to natural decay (17.91 yr. half life). Thus, after 10 cycles, the percentage of the initial quantities remaining were: ^{241}Am , 1.1%; ^{243}Am , 7.4%; ^{237}Np , 1.9%; ^{244}Cm , 9.7%.

Although the relative rates at which the isotopes burn out are the same with the two reprocessing schemes, the quantities that burn out are different. Table VI shows the isotopic compositions according to the two schemes, and it is seen that large differences exist. If uranium is not removed in the reprocessing, the ABR soon contains so little transplutronics that the quantity destroyed is not worth the effort. Justification cannot be given for running a reactor three years to destroy 36% of only 41 grams of ^{241}Am , or 23% of 140 grams of ^{243}Am . However, if uranium is removed, the quantity of ^{243}Am actually increases per cycle because of a relatively slow burn rate and the fact that in this conceptual scheme the uranium is replaced by more of the remaining actinide mix--for example, from other ABR's. Thus, the 10th cycle contains 1,183 grams of ^{243}Am , and the amount destroyed is greater than that contained in the region if uranium is not removed. This clearly illustrates the importance of removing as much U and Pu as possible during each reprocessing cycle.

The destruction of actinides in the central region is irrelevant if large amounts are to be created in the driver and blanket region. The amount created in the driver region is shown in Table IX. After three years a large amount of Pu and Np build in, but only a trivial amount of transplutronics. The Np-Pu increase comes from ^{235}U - ^{238}U

fuel in the driver. Although a radial blanket surrounded the driver for this run, the actinides that built into the blanket were not included because the exclusion of an axial blanket would make these numbers meaningless.

The quantity of actinides destroyed obviously depends on the cycle, the reprocessing scheme followed, and the type of fuel used. The metal fuel has the largest number density of actinides followed by the carbides. Table IX also shows the amount of actinides destroyed in the actinide region, oxide fuel, fifth cycle. It can be seen that a net destruction of transplutronics exists, and thus it can be concluded that the ABR succeeds in burning the actinides of greatest concern.

The rate at which the higher actinides (^{247}Cm , Bk, Cf) build in are shown in Figure 17. Although these isotopes do increase, the quantities are still insignificant in comparison to other actinides. The fission to capture ratio for the major isotopes in such a hard spectrum is such that the production of Cm, Bk, and Cf is not a legitimate concern.

Table VIII also shows the rate at which the major isotopes burn out in a flux of 5×10^{15} n/cm²-sec. As expected, the burnout rates are almost directly proportional to the power levels and/or flux levels used. Again, the conversion of U to Pu occurs independently of the burning of the higher actinides. The production of Cm, Bk, and Cf remains a problem of very minor concern.

In summary then, it appears that, at least theoretically, the major actinides can be largely eliminated by the continuous recycling in the ABR. The removal of as much U and Pu as is feasible is desired with each recycling, not because of the buildup of higher actinides, but so that more of the transplutronics can be loaded into the core. To remove only Pu and fission products at each cycle results in the eventual transformation to a pure breeder reactor in which ^{238}U dominates the actinide region. The fear of producing large amounts of Bk, Cf, and Cm is unfounded due to the high fission to capture ratio of most actinides in the ABR.

REACTIVITY COEFFICIENTS

We report here on calculations made to check statements in the introduction, namely that hard spectrum reactors of the ABR types have essentially zero Doppler coefficients and tend to have negative sodium void coefficients. We can not make a general statement about temperature coefficients arising from thermo-mechanical effects, as these are dependent on detailed mechanical design.

Doppler Effect

The seven actinide isotopes for which temperature-dependent cross section data were available to us are: ^{238}U , ^{239}Pu , ^{237}Np , ^{241}Am , ^{243}Am , ^{244}Cm , and ^{245}Cm . We could not at the time the calculations were done lay our hands on similar data for ^{240}Pu , ^{241}Pu , and ^{242}Pu .

which are the chief recycle actinides omitted from the previous list, so we used surrogates from those available for these (relatively low concentration) nuclides.

Zero dimensional NCINR8 runs were made for 3 x 9 actinide metal fuel and 3 x 9 actinide-50 w/o molybdenum fuel at the three temperatures 300 K, 900 K, and 2100 K. The results are summarized in Table X. The results show that k_{∞} does not change with temperature for either fuel type, and this implies that the Doppler effect is zero for ABR's. A Doppler coefficient of zero is not unexpected for the ABR's, since fission occurs in the actinides at neutron energies of 500 keV or greater, which is well into the unresolved resonance region where Doppler broadening has no appreciable effect.

Sodium Void Effect

For all-actinide sodium-cooled ABR's calculations were performed comparing k_{eff} with sodium density for a variety of core types. These included 3 x 9 metal fuel, 3 x 9 metal diluted with 50 w/o molybdenum, and the equivalent 6 x 9 fuel systems (see Table IV for compositions). Results of these calculations are shown in Table XI. The corresponding initial void coefficients defined as

$$\alpha_v \equiv \frac{1}{k} \frac{dk}{dx} \Big|_{x \rightarrow 0} \quad (3)$$

where x is void fraction, are given in Table XII.

The trends seen in Tables XI and XII are: First, as expected from the fact that the negative component of the sodium void effect arises from neutron leakage, the void effect, strongly positive for infinite core heights, decreases and becomes either much smaller, or negative for the 30 cm core. Second, the more reactive (higher k_{eff}) the infinite core, the smaller the void coefficient; k_{eff} is highest for the unvoided 6 x 9 all-metal core, followed in order by the 3 x 9 all-metal, the 6 x 9 moly and the 3 x 9 moly core. Third, it should be noted that even the 6 x 9 all-metal core is subcritical when its height is reduced to 30 cm. In order to have an all-actinide ABR that is critical when the leakage is large enough for the reactor to have a negative void coefficient, it appears that some spiking of the waste actinide mix with fissile material would be necessary.

For driver-driven ABR's, the system is dominated by the driver reactor. Besides neutron leakage from the ends, the critical driver has large radial neutron leakage as well. Every critical driver-driven case examined had a small negative void coefficient, the range of values being from -0.0005 to -0.0009. They are not tabulated as the various cases are not comparable in detail.

CONCLUSIONS AND DISCUSSION- section page 1

It is technically feasible to construct an operable (e.g., safe and stable) reactor to burn waste actinides rapidly. The heart of the concept is a driver core of EBR-II type, with a central radial target zone in which fuel elements, made entirely of waste actinides are exposed. This target fuel undergoes fission, as a result of which actinides are rapidly destroyed. Although the same result could be achieved in more conventionally designed LWR or LMFBR systems, the fast spectrum reactor does a much more efficient job, by virtue of the fact that in both LWR and LMFBR reactors, actinide fission is preceded by several captures before a fissile nuclide is formed. In the fast spectrum reactor that we have called ABR (actinide burning reactor), these neutron captures are short-circuited.

Originally, we had hypothesized that the direct destruction of actinide waste would be the chief advantage of the ABR. Considerations of waste management and chemistry have led us to alter our views on this point, slightly. The pertinent considerations are:

With regard to waste management, it is not the higher actinides (Am, Cm, etc.) that are a major problem, provided that they are not formed in large concentrations. While ^{241}Am may produce the "dominant" radioactivity in waste that is about 1000 years old, plutonium isotopes have comparable radioactivities, that persist for longer times. In short, the plutonium content of the waste is the most troublesome after all. In the long run, the waste actinide ^{237}Np is almost the only major, long-lived alpha emitter of high radioactivity.

With regard to chemistry, recent work by Croff, Blomeke and Finney (16) has illustrated the difficulty of recovering the trivalent, trans-plutonic elements from the lanthanides in the waste. The lighter actinides U, Np, and Pu can be recovered by ensuring that the usual quantities entrained in solids are dissolved and added to PUREX raffinates. A second solvent extraction cycle on these raffinates can effectively purge the U, Np, and Pu from them, to be added to the product side of the cycle. Only the Np so obtained can be considered a waste element. If Np is repeatedly recycled, it can become a major source of the nuclide ^{238}Pu , which is both a useful and a troublesome isotope. Thus, useful actinide burning is reduced to destruction of ^{237}Np .

An ABR could be constructed for this purpose. However, the virtues of ABR's are not limited to their ability to fission actinides. Their more generalized advantage is that this destruction capability short circuits the buildup of higher actinides. We avoid the problem of excessive buildup of transplutonic isotopes if we never form them.

Thus, at this point, we can conclude that the advantages of hard-spectrum breeder reactors (of which the parfait core LMFBR with small, highly enriched fissile zones is an example from current design concepts) includes the avoidance of heavy actinide formation, and the ability to get rid of ^{237}Np by using it virtually as a fuel, if it is desirable to burn that nuclide.

Final line of text ACKNOWLEDGMENTS : see caption on page 1

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Table I

COMPOSITION OF THE ACTINIDE WASTE
DILUTED WITH 50 w/o MOLYBDENUM(a)

Isotope	Number Density (atoms/barn cm)	
U-234	2.80	-9
U-235	1.79	-7
U-236	1.00	-7
U-238	2.11	-5
Np-237	1.13	-2
Pu-238	3.60	-6
Pu-239	1.07	-4
Pu-240	4.32	-5
Pu-241	1.78	-5
Pu-242	7.63	-6
Am-241	2.59	-3
Am-242	1.48	-5
Am-243	1.60	-3
Cm-242	1.58	-6
Cm-243	1.18	-6
Cm-244	3.92	-4
Cm-245	2.93	-5
Cm-246	3.51	-6
Cm-247	4.46	-8
Cm-248	2.90	-9
Cf-250	3.45	-11
Cf-252	5.70	-13
Mo	4.02	-2

(a) One part in 10^6 of the U and one part in 1000 of the plutonium in spent LWR fuel are counted in waste; It is assumed that the remaining U and Pu are returned to the standard fuel cycle.

Table II.

PIN-CELL GEOMETRY OF VARIOUS
ACTINIDE BURNING REACTOR TYPES

	Reactor Type ^(a)		
	<u>EBR-II</u>	<u>LMFBR</u>	<u>GCFR</u>
Fuel Pin Pitch ^b	5.66	8.64	11.4
Pin OR	2.21	3.75	4.11
Pellet OR	1.83	2.75	3.69
Clad Thickness	0.23	0.55	0.30
Gas Gap	0.15	0.45	0.12

^a The reactor type is characterized by a coolant--Na for EBR-II and LMFBR; He for GCFR--and by the pin and pitch geometries listed here. Enrichments (i.e., fissile fraction of the heavy atoms) and chemical compositions are variable for each type.

^b Dimensions are in millimeters.

Table III
EBR-II DRIVER REGION CHARACTERISTICS

Fuel (Volume fraction = 0.3782)

<u>Isotope</u>	<u>Number Density</u>
U-235	2.246-02
U-238	2.081-02
Mo	5.711-03

Clad (Volume fraction = 0.2397)

CR-52	1.668-02
FE	5.934-02
Ni	9.132-03

Coolant (Volume fraction = 0.3822)

Na-23	2.310-02
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Driver Region Radius = 40 cm
Actinide Region Radius = 5 cm
Core Height = 40 cm

Table IV

COMPOSITION OF THE PURE METAL ACTINIDE FUEL
IN ATOMS/BARN-CM FOR TWO STANDARD MIXES

Isotope	Mix Designation	
	ACT6x9	ACT3x9
U-234	7.5336E-9	3.4029E-6
U-235	4.8216E-7	2.1775E-4
U-236	2.7130E-7	1.2256E-4
U-238	5.6822E-5	2.5670E-2
Np-237	3.0430E-2	1.3742E-2
Pu-238	9.5847E-6	4.3284E-6
Pu-239	2.8770E-4	1.2993E-4
Pu-240	1.1644E-4	5.2544E-5
Pu-241	4.7963E-5	2.1650E-5
Pu-242	2.0528E-5	9.2696E-6
Am-241	6.9852E-3	3.1576E-3
Am-242M	3.7752E-5	1.7953E-5
Am-243	4.2981E-3	1.7411E-3
Cm-242	4.2621E-6	1.9729E-6
Cm-243	3.1836E-6	1.4383E-6
Cm-244	1.0570E-3	4.7702E-4
Cm-245	7.8954E-5	3.5662E-5
Cm-246	9.4360E-6	4.2621E-6
Cm-247	1.2007E-7	5.4195E-8
Cm-248	7.7944E-9	3.5231E-9
Cf-250	9.2847E-11	4.1937E-11
Cf-252	1.5345E-12	6.1375E-13

Table V

 k_{∞} OF FIVE ABR FUEL LOADINGS VS. EXPOSURE

Exposure (Days)	Metal	Metal +Molybdenum (a)	Metal Oxide	Metal Carbide	GCFR Metal (b)
0	1.4115	0.8022	1.0232	1.1705	1.4859
120	1.4287	0.8818	1.0787	1.2073	1.5008
240	1.4395	0.9318	1.1141	1.2308	1.5096
360	1.4556	0.9855	1.1592	1.2628	1.5236
720	1.4725	1.0336	1.2033	1.2952	1.5380
900	1.5008	1.0832	1.2649	1.3436	1.5618
1080	1.5121	1.0904	1.2850	1.3609	1.5713

(a) The actinide metal was diluted with 50 w/o molybdenum for this case.

(b) All of the reactors used EBR-II type pins except GCFR which used .74 cm OD pins.

Table VI

BOC COMPOSITION OF 10 kg OF ACTINIDES
ACCORDING TO TWO REPROCESSING SCHEMES

Isotope	Cycle 1	Cycle 5		Cycle 10	
	Scheme	Scheme		Scheme	
	<u>I & II</u>	<u>I</u>	<u>II</u>	<u>I</u>	<u>II</u>
U-234	.73	39.5	37.3	50.8	26.8
U-235	47.1	11.6	15.3	10.0	8.08
U-236	26.6	25.0	35.0	19.3	30.4
U-238	5621.0	5645.0	8027.0	5616.0	9353.0
Np-237	2997.0	2700.0	1151.0	2153.0	279.1
Pu-238	.95	30.7	29.7	26.8	7.69
Pu-239	28.6	14.9	32.8	16.3	41.9
Pu-240	11.6	2.54	3.08	4.75	2.65
Pu-241	4.80	.05	.07	.08	.06
Pu-242	2.07	.01	.01	.01	.01
Am-241	700.4	536.5	227.0	348.8	41.0
Am-242	4.0	49.5	22.1	36.3	4.39
Am-243	434.1	708.4	299.8	1183.0	140.0
Cm-242	4.39	1.37	.59	.91	.10
Cm-243	.32	5.18	2.22	4.08	.50
Cm-244	107.1	215.8	92.8	385.6	48.1
Cm-245	8.04	4.82	20.7	98.2	12.4
Cm-246	.97	8.89	3.82	42.6	5.30

Table VII

Fuel	Cycle	Reprocessing Scheme	$^{239}\text{Pu}_{\text{BOC}}$ (g)	$^{239}\text{Pu}_{\text{EOC}}$ (g)	$^{238}\text{U}_{\text{BOC}}$ (g)	$\frac{^{239}\text{Pu}_{\text{EOC}} - .539 ^{239}\text{Pu}_{\text{BOC}}}{^{238}\text{U}_{\text{BOC}}}$
Oxide	1	---	29.93	229.2	5889	3.6%
Oxide	10	II	45.29	379.5	10100	3.5%
Oxide	10	I	28.12	221.9	5882	3.5%
Metal	1	---	55.81	411.8	10980	3.5%
Carbide	5	II	48.04	442.8	11920	3.5%

Table VIII

Isotope	<u>135 MW</u>		<u>5·10¹⁵</u>	
	<u>% Destroyed/Cycle*</u>	<u>% Remaining After 10 Cycles</u>	<u>% Destroyed/Cycle*</u>	<u>% Remaining After 10 Cycles</u>
U-234	---	868 %	---	339 %
U-235	---	4.1%	---	1.7%
U-236	---	23 %	---	16.8%
U-238	7.5%	46 %	10%	34 %
Np-237	32 %	1.9%	41%	0.6%
Am-241	36 %	1.1%	46%	0.2%
Am-242	---	21 %	---	4.0%
Am-243	23 %	7.4%	30%	2.7%
Cm-242	---	0.5%	---	---
Cm-243	---	31 %	---	---
Cm-244	21 %	10.3%	27%	4.4%
Cm-245	---	36 %	---	17 %
Cm-246	---	148 %	---	104 %

* If Consistent.

Table IX

	<u>Net Gain or Loss In Driver Region (g)</u>	<u>Net Gain or Loss in Actinide Region (g)</u>	<u>Total Gain or Loss (g)</u>
U-234	+176	-5.5	+170.5
U-235	-203,000	-2.2	-203,000
U-236	+28,470	-3.2	+28,470
U-238	-32,300	-446	-32,740
Np-237	+1,324	-948	+376
Pu-238	+63.13	+398	+461
Pu-239	+15,430	+207	+15,637
Pu-240	+265.5	+28	+291.5
Pu-241	+3.43	+8	+4.2
Pu-242	+4.03 (-2)	.11	+1.15
Am-241	+1.104	-202	-202
Am-242	+7.21 (-4)	-16.5	-16.5
Am-243	+2.38 (-4)	-170	-170
Cm-242	+1.77 (-3)	+17.29	+17.29
Cm-243	+2.54 (-5)	-1.5	-1.5
Cm-244	---	-34	-34
Cm-245	---	-8.8	-8.8
Cm-246	---	+12	+12

Table X

EFFECT OF TEMPERATURE DEPENDENT CROSS SECTIONS
(DOPPLER EFFECT) ON k_{∞} OF TWO TYPES OF ACTINIDE FUELS

Fuel Mix ^(a) and Type	k_{∞} at		
	300k	900k	2100k
3 X 9, Metal	0.94786	0.94786	0.94786
3 X 9, Moly-diluted	0.47872	0.47872	0.47872

(a) See Table IV for description of the 3 X 9 mix.

Table XI

SODIUM VOID EFFECTS IN ALL-ACTINIDE ABR's

Pin Cell: EBR-II Type; Infinite Core Radius

Fuel Mix	Fuel Type	Core Height	k_{eff} for Listed % Na Voids				
			0	25	50	75	100
3 X 9	Metal	∞	0.94766	0.96771	0.99124	1.01643	1.04344
		40 cm.	0.69917	0.70296	0.70665	0.70970	0.71197
		30 cm.	0.58225	0.58158	0.58023	0.57813	0.57521
	Moly Diluted	∞	0.47334	0.48761	0.50492	0.52421	0.54589
		40 cm.	0.32107	0.32389	0.32684	0.32956	0.33196
		30 cm.	0.25780	0.25794	0.25782	0.25735	0.25649
	Metal	∞	1.41153	1.43343	1.45845	1.48474	1.51204
		40 cm.	1.09318	1.09713	1.10074	1.10342	1.10498
		30 cm.	0.93042	0.92857	0.92560	0.92156	0.91630
6 X 9	Metal	∞	0.77363	0.79600	0.82283	0.85231	0.88486
		40 cm.	0.53230	0.53644	0.54060	0.54422	0.54714
		30 cm.	0.42747	0.42713	0.42620	0.42461	0.42227
	Moly-diluted	∞	0.77363	0.79600	0.82283	0.85231	0.88486
		40 cm.	0.53230	0.53644	0.54060	0.54422	0.54714

Table XII
INITIAL SODIUM VOID COEFFICIENTS, α_v

<u>Fuel Type</u>	<u>Base System Height, Cm.</u>	<u>α_v</u>
3 X 9, Metal	∞	0.0822
	40	0.0245
	30	-0.0017
3 X 9, Moly	∞	0.1130
	40	0.0380
	30	0.0054
6 X 9, Metal	∞	0.0619
	40	0.0169
	30	-0.0055
6 X 9, Moly	∞	0.1102
	40	0.0346
	30	0.0004

Figure 1
SPECTRA OF ACTINIDE REGIONS IN EBR-II AND PHENIX
COMPARED TO CAPTURE AND FISSION
CROSS SECTIONS OF AM-241

Figure 2
SPECTRUM FOR ACTINIDE METAL IN AN
LMFBR PIN CELL DESIGN

Figure 3
SPECTRUM FOR ACTINIDE OXIDE IN AN
LMFBR PIN CELL DESIGN

Figure 4

SPECTRUM FOR 50 W/O MO WITH ACTINIDES IN AN
LMFBR PIN CELL DESIGN

Figure 5
SPECTRUM FOR AN ACTINIDE CARBIDE IN AN
LMFBR FUEL PIN DESIGN

Figure 6
SPECTRAL PARAMETER CORRELATIONS FOR 6 x 9 ACTINIDE MIX

Figure 7
SPECTRAL PARAMETER CORRELATIONS FOR 3 x 9 ACTINIDE MIX

Figure 8

FRACTION OF AM BURNED IN AN ACTINIDE METAL
FUELED EBR-II CONFIGURATION

Figure 9

FRACTION OF AM BURNED IN A MOLY DILUENT
FUELED EBR-II CONFIGURATION

Figure 10

FRACTION OF AM BURNED IN AN ACTINIDE
OXIDE FUELED EBR-II CONFIGURATION

Figure 11

FRACTION OF AM BURNED IN AN ACTINIDE
CARBIDE FUELED EBR-II CONFIGURATION

Figure 12
BOC SPECTRUM, CYCLE 10, 3 x 9 OXIDE FUEL

Figure 13

SPECTRUM AT END OF FIRST CYCLE, 3×9 OXIDE FUEL

Figure 14
SPECTRUM WITH BLANKET SURROUNDING CORE

Figure 15
RADIAL FLUX PROFILE IN ACTINIDE REGION

Figure 16
DECAY OF MAJOR ACTINIDES FROM INITIAL
10 KG SAMPLE, AT 135 MW DRIVER POWER

Figure 17
BUILDUP OF MINOR ACTINIDES FROM INITIAL
10⁻⁴ KG SAMPLE AT 135 MW