PAUL E. SCHOFIELD

for the degree of MASTER OF SCIENCE

A STUDY OF THE BEHAVIOR OF ACTINIDES CONTINUOUSLY
RECYCLED IN A HARD SPECTRUM REACTOR

DO NOT MICROFILM
THIS PAGE

OSU-NE reports are the result of work performed in the Nuclear Engineering Department of Oregon State University which, in the opinion of the departmental faculty, contain information useful to other researchers. A limited number of copies are available from the Department. The report series includes theses (available from University Microfilms, Ann Arbor, Michigan), documents which may be reissued under another report number by a sponsoring agency, preprints, some student project reports, and archival data.

MASTER

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

DOE/RL/74032--7

DE83 017453

A STUDY OF THE BEHAVIOR OF ACTINIDES CONTINUOUSLY
RECYCLED IN A HARD SPECTRUM REACTOR

by

Paul Edwin Schofield

A THESIS

submitted to

Oregon State University

in partial fulfillment of
the requirements for the
degree of

Master of Science

Completed October 8, 1980

Commencement June, 1981

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

MASTER

EDB

AN ABSTRACT OF THE THESIS OF

Paul Schofield for the degree of Master of Science
in Nuclear Engineering presented on October 8, 1980.

Title: A Study of the Behavior of Actinides Continuously
Recycled in a Hard Spectrum Reactor

Abstract approved: _____

Dr. Alan H. Robinson

The behavior of actinides continuously recycled through the central region of an EBR-II type reactor was studied. Such a reactor would convert long-lived nuclear "wastes" to short-lived isotopes, and simultaneously produce useful power. This process is proposed as an alternative to the geological isolation of long-lived actinide "wastes". A driver region of 50% U-235 enriched fuel provided a nearly-constant spectrum and flux that was extremely hard compared to standard LMFBRs. This resulted in a high fission-to-capture ratio for most isotopes.

The original actinide fuel was the discharge from a LWR, cooled for two years, with 99.9% of the uranium and plutonium removed by chemical processing. Comparison was made between removal of both Pu and U and removal of only U in subsequent cycles. The latter case resulted in substantial quantities of trans-plutonics "burned" per cycle.

Operating the reactor at an average flux of

$3.6 \times 10^{15} \text{ n/cm}^2\text{-sec}$ resulted in 36% of the beginning of cycle (BOC) Am-241 and 23% of the BOC Am-243 burning out each three year cycle. These burn rates were established regardless of the original U and Pu content. The rate of actinide burnup was independent of the fuel type and the reprocessing method used. However, U-238 increased from 56% to 93% of the actinide mix when not removed during reprocessing. Thus little room remained for the trans-plutonics, with the result that the quantity of trans-plutonics burned decreased significantly using this reprocessing method.

Although 3.55% of the BOC U-238 was converted to Pu-239, a minimal buildup of trans-plutonics occurred. Isotopes of berkelium and californium continued to build up through ten cycles, but constituted less than 0.0001 grams per kilogram of original actinide material.

The power density of the actinide fuel was less than half that of the driver region. This means that an improved core could result in an increased flux in the actinide region. Increasing the flux to $5.0 \times 10^{15} \text{ n/cm}^2\text{-sec}$ resulted in burn rates slightly less than proportional to the increase in flux. This higher level of flux still produced negligible trans-plutonics. The hardness of the spectrum of the driver region indicates that actinides could be distributed throughout the core. In this case, it is expected that a 1000 MWe reactor could

burn the waste of two or more LWRs. A reactor similar to that described in this thesis could help eliminate the long term storage presently planned for the discharge of LWRs. Actinide burning reactors would prevent the initial buildup of actinide waste if used exclusively.

APPROVED:

Professor of Nuclear Engineering
in Charge of Major

Head of Department of Nuclear Engineering

Dean of Graduate School

Date thesis is presented October 8, 1980

Typed by Paul Schofield for Paul Edwin Schofield

TABLE OF CONTENTS

INTRODUCTION	
DESCRIPTION OF THE ACTINIDE BURNING REACTOR	10
The ABR	10
The NCINR8 Code	13
The decay and buildup of actinides	14
Reprocessing Schemes	16
Fuel Types and Power Levels	21
Assumptions	22
NEUTRON SPECTRA	25
Spectra in the ABR	25
Collapsed Cross-sections	35
Radial Flux Profiles	36
Alternative Arrangements	40
RESULTS AND DISCUSSION	45
Burn Rates in the ABR	45
Activity Levels of Trans-plutonics	52
Comparison of Reprocessing Schemes	54
Breeding of Pu in the actinide region	61
Actinide Buildup in the Driver Region	68
Buildup of the Higher Actinides	70
Burnup at the Higher Flux Level	70
Optimizing the ABR	75
SUMMARY AND CONCLUSIONS	82
REFERENCES	85
APPENDIX - DESCRIPTION OF NCINR8	88

LIST OF ILLUSTRATIONS

<u>Figure</u>		<u>Page</u>
1	Fission to capture ratio for Am-241	11
2	Fraction of Am remaining in actinide region with driver power of 600 kw/l	17
3	Spectra of Phenix and actinide region (metal fuel) in ABR compared to capture and fission cross-sections of Am-241	26
4	Spectrum of metal fuel in ABR configuration without driver	29
5	Spectrum for carbide fuel, BOC, first cycle	31
6	Spectrum for oxide fuel, BOC, first cycle	32
7	EOC spectra for oxide fuel at 135 MW and at 5.0E15 flux	34
8	Radial flux profiles for coarse groups 1-5	39
	Radial flux profiles for coarse groups 6-9	39a
10	Spectrum averaged over driver region and spectrum at outer edge of driver region	42
11	Decay of major actinides from initial kg sample, at 135 MW driver power	46
12	Activities initiated by Np-237 and transplutronics from original actinide material	55
13	Activities initiated by Np-237 and transplutronics from original actinide material after 5 cycles	56

14	Activities initiated by Np-237 and transplutonics from original actinide material after 10 cycles	57
15	Buildup of minor actinides from initial kg sample, at 135 MW driver power	71
16	Decay of major actinides from initial kg sample, at 5.0×10^{15} flux	73
17	Spectrum with actinides distributed throughout core	79

LIST OF TABLES

Table

I	Parameters for ABR based on EBR-II	12
II	NCINR8 Isotopes and half-lives	15
III	NCINR8 Alpha decay chains	18
IV	Comparison of fuel types	22
V	Group fluxes in actinide and driver regions	27
VI	Constant spectrum run compared to variable spectrum run	37
VII	Cross-sections collapsed in constant spectrum	38
VIII	Coarse group structure	40
IX	Percent of major actinides remaining after ten cycles	47
X	Quantity of actinides remaining from kg sample	48
XI	System half-lives of major isotopes	51
XII	BOC composition of kg of actinides according to two reprocessing schemes	59
XIII	Quantities of trans-plutonics burned per kg of material	60
XIV	Power density for metal fuel at 135 MW	63
XV	Composition of kg of Pu after exposure to flux for three years	63
XVI	Buildup of plutonium in various cycles	66
XVII	Isotopic Pu concentration using first reprocessing method	66

XVIII	Net gain or loss of actinides in ABR	69
XIX	Comparison of two power/flux levels	74
XX	Buildup of higher actinides at 135 MW and 5×10^{15} n/cm ² -sec	76
XXI	Percent of actinides burned when distributed throughout core	78

A Study on the Behavior of Actinides Continuously Recycled in a Hard Spectrum Reactor

INTRODUCTION

The present nuclear policy in the United States is that of a "once through" reactor - the fuel for the reactor is used one time, and then considered waste. This situation is analogous to what is now considered the out-dated idea of the throw-away aluminum can - not only is a valuable resource being unnecessarily discarded, but unpleasant garbage accumulates as well. While the recycling of aluminum, tin, glass, and other commodities has become an obvious solution to reducing waste and to saving a critically short supply of resources, the recycling of spent fuel, an energy source in itself, has been forbidden by government action. Moreover, whereas the waste generated by aluminum is at worst an eyesore to the public, the by-products generated by reactors could constitute a potential hazard to that public for thousands of years. The political repercussions of this problem have threatened a moratorium of the entire nuclear industry. Even as garbage itself is being considered a potential energy source, nuclear "garbage" is overlooked outside the nuclear community. Previous studies have shown that the longest lived of these by-products, the actinides, can be

used to fuel a nuclear reactor (1-5). This not only eliminates long term storage considerations for the "wastes", but produces useful power as well. This paper studies the behavior of actinides continuously recycled in a hard spectrum reactor.

The light water reactors (LWRs) utilized in the United States contain fuel rods typically enriched to 3% U-235. Natural uranium contains 0.01% U-234, 0.72% U-235, and 99.27% U-238. Spent fuel rods from a LWR contain, in addition to fission products, isotopes of plutonium, americium, curium, and heavier elements. These isotopes are formed when one or more neutron captures occur instead of fission. It is the isotopes of these elements, rather than fission products, that have half-lives long enough to suggest isolation for several millenia.

Actinides are those elements with atomic numbers greater than that of actinium, ^{89}Ac . Of particular radiological concern are the trans-plutonics having half-lives of 100 to 100,000 years. An example of this is Am-241 which, with a half-life of 434 years, is sufficiently radioactive to warrant legitimate concern for thousands of years. Isotopes with half-lives of less than 100 years will have activities of less than 0.1% of the original amount after 1000 years. Isolation for such a time period is not a serious technical problem within a

stable social situation. It is in this time range that the half-lives of most fission products fall. Only three fission products of significant production (fission yield) have half-lives between 10 and 100 years. No fission products have half-lives between 100 - 100,000 years, and only two have half-lives between 10^5 - 10^6 years. Isotopes with half lives greater than 100,000 years are not a serious radiological problem, due to their low specific radioactivity. This can be seen by considering the extreme case of those isotopes with infinite half lives, which, of course, are defined as stable. All actinides continue to decay through α and β emission until the stable isotopes of Pb-206, Pb-207, Pb-208, or Bi-209 are reached.

The best method for disposing of long-lived by-products is a subject of debate. Methods other than transmutation that have been suggested include placement in mined repositories, placement in deep ocean sediments, placement in very deep drill holes, and ejection into space (6). Disposal under the polar ice caps and the earth's tectonic plates have also been proposed. The principle in each of these methods is the isolation of the wastes from man and the aquifers that may be the vehicle for returning radioactivity to the environment.

The isolation time necessary to render these wastes innocuous has also been subject to debate and

misconception. The specific radioactivities of the wastes can be put into perspective through a comparison to natural uranium and thorium. These naturally occurring elements have specific radioactivities of approximately 10 curies/tonne. After 1000 years, spent fuel has a specific radioactivity of approximately 1000 curies/tonne (8). The solid discharge from a reprocessing plant has a specific radioactivity of approximately 5000 curies/tonne (8). The reprocessing plant discharge has a higher specific radioactivity than spent fuel because of the removal of long-lived U and Pu isotopes, which act as diluents. After 1000 years, the fission products have decayed away, and the majority of the activity is from Am-241. After a period of 10,000 years, the specific radioactivities of spent fuel and of reprocessing plant discharge are approximately 500 and 1200 curies/tonne, respectively. After one million years, the specific radioactivities are less than those of natural uranium and thorium.

Diluting long-lived nuclear waste to one part per thousand with an inert substance such as sand would reduce the specific radioactivity to less than that of natural uranium within several thousand years. The waste would be processed into an inert glass and/or enclosed in metal casing for disposal in deep, geologically stable, water-free salt or granite formations (9). Studies have

shown that if all U.S. electrical power were generated by LWRs for the next 1000 years, the undiluted actinide waste created would occupy only 12 square miles if stored in canisters spaced ten meters apart (7). This assumes a 99.5% reprocessing of all Pu and U.

It is difficult, however, to convince a skeptical public that disposal sites will remain undisturbed for several thousand years (6). The probability of the wastes leaching to the environment or being disturbed through natural phenomena such as volcanos at the proposed sites is small (6). However, the intentional or unintentional intrusion of man cannot be ignored. Even if isolation is not a technical problem, it is certainly a public relations and political one. An alternative to isolation, such as an actinide burning reactor, would remedy this situation.

Regardless of the feasibility of burial, it is nonetheless undesirable for any industry to produce large quantities of waste. An industry should attempt to consume as much of its own waste as possible. Currently, 8300 kg of actinide wastes (all isotopes of Np, Am, and Cm) have accumulated. This total is expected to reach 155,000 kg by the year 2000 (2). Actinide "wastes", however, need not be environmentally isolated until they decay naturally to a stable state. Most actinides have an effective fission threshold of several hundred kev and can therefore be

considered potential fuel in a reactor that utilizes high energy, or "fast" neutrons. This is known as a "hard" spectrum reactor. Light water reactors have most of their neutrons in the thermal range, with energies less than 1 ev. Low energy, or "slow" neutrons are far more likely to be captured than to cause fission in collision with heavy nuclides that are not considered fissile. A spectrum is made "softer" if neutrons experience inelastic scattering or multiple elastic scatterings, particularly with light atoms.

Harder spectra are found in liquid metal fast breeder reactors (LMFBRs) than in LWRs. With average neutron energies in the tens of kev range, however, adequate actinide fission still does not exist in the "typical" LMFBR. Some fissioning can occur after multiple neutron captures produce fissile or spontaneously fissioning nuclides. However, the benefits of this fissioning can be offset by the high radioactivity and longer isolation times required of isotopes that absorb neutrons. Furthermore, it is not as efficient to achieve a fission by first capturing several neutrons as compared to achieving a fission right away. Studies at Oak Ridge have shown the benefits derived from recycling actinides in an LMFBR to be minimal (10). However, these studies include time periods up to one million years, and probabilities on such a time scale are

considered invalid in this paper.

Reactors do exist, however, with neutron spectra that have a median energy value in the range of several hundred kev. An example of this is the EBR-II, which started operation in Idaho in 1964 and has a median neutron energy on the order of 500 kev (11,4). The hardness of the spectrum is due to the large proportion of the fissile isotope U-235 in the core (50% enrichment). Neutrons are absorbed by the U-235 before many elastic collisions with medium weight isotopes can occur. The U-235 atom produces fast neutrons when it fissions. The comparatively small proportion of fertile isotopes, such as U-238, reduces the amount of inelastic scatterings that occur. This results in a fast reactor well-suited to the purpose of transmuting the actinides. It is the extremely hard spectrum of such a reactor that separates this thesis and previous work at Oregon State University (3,4,5) from studies that have occurred elsewhere using conventional LWRs and LMFBRs (1,2,10,12,13).

The Actinide Burning Reactor (ABR) is a reactor similar in nuclear design to the EBR-II, but the ABR utilizes the waste from LWRs as part of its fuel. The ABR conceptually could be part of a nuclear park that also contains several LWRs and a reprocessing plant (14). The discharge from a LWR would then be reprocessed and

transported locally to fuel the ABR. This would reduce transportation hazards and the volume of spent material that is to be geologically isolated. The fuel that is discharged from the ABR could then be reprocessed to remove fission products and inserted back into the ABR. In theory, this recycling could continue indefinitely, so that the actinides need never be considered waste. Geological isolation would only be necessary for periods of time appropriate to fission product decay. This paper studies the behavior of actinides as they are repeatedly cycled through an actinide burning reactor, as modeled by the computer code NCINR8 (3,15).

The next chapter contains a description of the ABR and the various considerations that went into its design. The properties of actinides are discussed, including decay schemes, half-lives, and cross-sections. Fission and capture cross-sections as a function of neutron energy are discussed, and why this energy dependence necessitates an EBR-II type reactor. The chapter also discusses the fuel types, reprocessing schemes, and power levels used in this study.

Because of the importance of a hard spectrum, the third chapter is devoted entirely to this subject. Comparison is made between the spectrum of the ABR and that of the Phenix reactor. The ABR is shown to be superior to

the Phenix in terms of fissioning actinides. Spectra are plotted for the various fuel types utilized in the ABR. It will be shown that the spectrum in the actinide region is determined almost entirely by the driver region. The fuel composition in the actinide region has little effect on the spectrum. It is thus possible to collapse the fission and capture cross sections of the actinides in the nearly-constant spectrum. The third chapter also discusses the radial flux profile and alternative arrangements in the ABR.

The fourth chapter shows that significant burnup of the actinides does occur in the ABR. The effect of fuel types, reprocessing schemes, and power levels on the burn rates is discussed. It will be shown that very little neutron absorption occurs, so that the fear of a buildup of the higher actinides is unfounded. It will be concluded that the extensive use of actinide burning reactors would eliminate the production and buildup of actinide wastes.

DESCRIPTION OF THE ACTINIDE BURNING REACTOR

The ABR

Figure 1 shows the fission to capture ratio ($1/\alpha$) of Am-241 as a function of the energy of the incident neutron, according to the data described in the Appendix. Am-241 is representative of most other actinides in that $1/\alpha$ increases several orders of magnitude as energy increases from the kev to the Mev range. With an incident neutron energy of 10 Mev, nearly 100 fissions occur for every capture in Am-241. If the incident energy is below 100 kev, the opposite is true. To efficiently "destroy" actinides then, a reactor should have a very hard spectrum.

As stated previously, a sodium cooled EBR-II type reactor is well-suited for this purpose. The important parameters of an ABR based on the EBR-II are shown in Table I (16). The average and peak power densities are well above average for a "typical" LMFBR, while the flux is below the 8.35×10^{15} n/cm²-sec found in a reactor such as Phenix (16). This is due to the fact that the ABR is 50% enriched, compared to the more standard enrichment of 20%.

The core of the ABR is 40 cm in radius, with an inner region of five centimeter radius containing the actinide fuel. The actinide mix of the first cycle was obtained from the spent fuel of a LWR. After a three year in-core

Figure 1

FISSION TO CAPTURE RATIO
FOR AM-241

TABLE I: Parameters for ABR Based on
EBR-II (16)

Height	40 cm
Radius	40 cm
Radius of actinide region	5 cm
Fuel pin radius	72 mills
Gap thickness	6.0 mills
Clad thickness	9.0 mills
Pitch	223 mills
Uranium weight percent	95%
Enrichment	50% U-235
Design avg. power density	860 kw/l
Design peak power density	1430 kw/l
Design flux	$3.7 \times 10^{15} \text{ n/cm}^2\text{-sec}$

irradiation period and a two year cooling period, the spent fuel was reprocessed by removing 99.9% of its plutonium and uranium and all the fission products. The remaining actinide mix was fabricated into rods and inserted into the central region. A mix such as this has a critical radius of 25.3 cm and a critical mass of 1220 kg. However, an undriven reactor fueled with this has a much softer spectrum than that of the EBR-II (3). The driver region was thus conceived to determine the behavior of the actinides in a hard, constant spectrum. The actual reactor may or may not have a defined actinide region as such, so the size of the actinide region was somewhat arbitrary: indeed, in actual practice the actinide rods would most likely be dispersed throughout the reactor. For conceptual and computational ease, the actinide rods were lumped into the central region, where the highest flux occurs.

The NCINR8 Code

The ABR was modeled by the computer code NCINR8, which was written specifically for this purpose. It uses multi-group shielded cross-section data to determine spectra, perform one-dimensional diffusion calculations, and simulate isotopic burnup for a given interval of reactor time. Each simulated run in this paper represents three years of reactor operation. In addition to cataloging the core inventory throughout the burn,

inventories for the 25 isotopes are also followed for a 1000 year period following each burn. The cross-section data, a preliminary version of ENDF/B-V, has 29 groups in the Bondarenko format (17-20). Input variables to NCINR8 include core size, reactor power or flux, reactor geometry, number and size of different regions, unit cell geometry, fuel pin radius, gap thickness, clad thickness, and array pitch. The fuel composition could include up to 25 isotopes, as listed with their half-lives in Table II. The code had the option of several coolants, cladding materials, and fuel types. This study, however, used exclusively Na-23 as a coolant and stainless steel as a clad (20% chromium, 70% iron, and 10% nickel). NCINR8 is described in more detail in Appendix 1.

The Decay and Buildup of Actinides

As opposed to fission products, even those actinides with short half-lives must also be considered long term storage problems, as eventually they decay into long-lived daughter isotopes. An example of this is Cm-242, with a half-life of 162.8 days, which has the following decay chain:

All actinides have several decays until a stable isotope is

TABLE II: NCINR8 ISOTOPES AND HALF-LIVES

<u>Nuclide</u>	<u>T_{1/2}(yrs)</u>	<u>Nuclide</u>	<u>T_{1/2}(yrs)</u>
U-234	2.442E+05	U-235	7.136E+08
U-236	2.341E+07	U-238	4.476E+09
Np-237	2.142E+06	Pu-238	8.792E+01
Pu-239	2.439E+04	Pu-240	6.542E+03
Pu-241	1.508E+01	Pu-242	3.870E+05
Am-241	4.335E+02	Am-242	1.521E+02
Am-243	7.376E+03	Cm-242	4.467E-01
Cm-243	2.804E+01	Cm-244	1.791E+01
Cm-245	7.739E+03	Cm-246	4.768E+03
Cm-247	1.541E+07	Cm-248	3.500E+05
Bk-249	8.519E-01	Cf-249	3.522E+02
Cf-250	1.311E+01	Cf-251	9.008E+02
Cf-252	2.632E+00		

reached, and so are not deactivated after one emission has occurred. The specific activity, however, decreases as the longer lived daughters build up, and any hazard is proportionately reduced. In this paper, Am-241 (433.5 y) and Am-243 (7376 y) will be considered representative of the actinides, due to their significant LWR production rates and the appropriateness of their half-lives.

Previous work has shown that 32% of the Am-241 loaded and 20% of the Am-243 are transmuted in a three year burn at a core average power density of 600 kw/l (4). This is shown in Figure 2. Uncertainty existed, however, to what extent the gains accomplished by fission were offset by neutron capture, and how continued recycling would effect this balance. Table III shows the decay schemes for NCINR8 actinides (although decay continues further than indicated). By producing Cm, Bk, and Cf, the isolation time required increases as several decays are necessary to return to the original isotopes. Radiation is released in the process. Of secondary importance, fission energy is lost as a result of parasitic capture. It is unacceptable to have capture occur instead of fission, and one criteria of the ABR is to prevent this. By following the inventory through several cycles, the extent to which these higher isotopes build up should become evident.

Figure 2

FRACTION OF AM REMAINING IN ACTINIDE REGION
WITH DRIVER REGION POWER OF 600 KW/L

TABLE III: NCINR8 ALPHA DECAY CHAINS

<u>Parent Nuclide</u>	<u>First Decay</u>	<u>Second Decay</u>	<u>Third Decay</u>	<u>Fourth Decay</u>
Cf-252	Cm-248			
Cf-251	Cm-247	Am-243	Pu-239	U-235
Cf-250	Cm-246	Pu-242	U-238	
Cf-249	Cm-245	Pu-241	Np-237	
Bk-249	Cm-245	Pu-241	Np-237	
Cm-247	Am-243	Pu-239	U-235	
Cm-246	Pu-242	U-238		
Cm-245	Pu-241	Np-237		
Cm-244	Pu-240	U-236		
Cm-243	Pu-239	U-235		
Cm-242	Pu-238	U-234		
Am-243	Pu-239	U-235		
Am-242	Pu-238	U-234		
Pu-242	U-238			
Pu-241	Np-237			
Pu-240	U-236			
Pu-239	U-235			
Pu-238	U-234			

Two different reprocessing schemes were followed for the actinides. As already noted, the fuel for the first cycle was obtained from the spent fuel of a LWR. Spent fuel rods were allowed to cool for two years, then reprocessed by removing all of the fission products and 99.9% of the uranium and plutonium. This is assumed to be an achievable extraction rate (21,22). The removal of plutonium was assumed to be necessary because it is a precursor to americium through neutron capture and through the β -decay of Pu-241. The buildup of Am is contrary to the purpose of the ABR. Furthermore, plutonium is valuable as fuel in LWRs and conventional LMFBRs, as well as for military purposes. Uranium was extracted because, as a naturally occurring and nearly stable element, it is not comparable to the actinides with which this paper is concerned. U-238 also contributes to a softer spectrum with its inelastic down-scattering. Moreover, the U-238 can be used effectively and with purpose in the ABR's fertile blanket. The remaining actinide mixture was reconstituted and placed into the central region of the ABR, where it remained for three years. In an actual reactor, one-third of the rods are usually replaced every year, but for computational purposes this was not done.

Future cycles all followed the three year in-core, two year cooling scheme, but differences occurred during reprocessing. In the first reprocessing method, the levels

of Pu and U were reset to the levels of the beginning of the first cycle. It was thus assumed that in future reprocessing cycles the levels to which Pu and U were reduced in the first cycle could be obtained. In the second scheme, 90% of the Pu was removed each cycle, but the U remained in the actinide mixture. The level of Pu was never less than that of the first cycle because of the breeding that occurred during the burns. The levels of uranium also increased with respect to the other actinides due to its relatively slow transmutation rate. The previous paragraph suggests that not removing the uranium is impractical, as well as chemically more difficult than removing uranium and recycling plutonium. It was thought, however, that uranium might reach an equilibrium level near its level in the first cycle. This would then eliminate the cost of removing uranium during reprocessing. This paper shows that the actinides could be recycled indefinitely, but as trends were well established after ten five-year cycles, the study was then terminated.

In this study, the fuel in each generation is placed in a separate reactor. This is not the way an actual cycle would work. The discharge from a given ABR, after reprocessing, could be inserted back into any other ABR, including the original one. Thus separate generations would not exist, as each ABR would be fueled with a mixture of several generations. The behavior of a mixed generation

reactor, however, can be determined by multiplying the quantities burned in separate generation reactors by the fraction of the fuel occupied by that generation. The make-up fuel for the mixed generation reactor would be provided by the discharge from LWRs. Studies have shown that using a similar concept, one 1200 MWe LMFBR can accomodate the discharge of three LWRs (1).

Fuel Types and Power Levels

Although metal fuel was used exclusively in the driver region, oxide, metal, and carbide fuels were all studied in the actinide region. It was unknown at the start how the fuel type would affect the burn rates. As shown in Table IV, a higher quantity of actinides can be loaded into the core using metal fuel: 1.86 times as much as oxide fuel. The melting point of metal fuel, however, is only 1155°C, 1600°C less than oxide fuel (23).

Two different power/flux levels were studied. The first was a core power level of 135 MWth. This resulted in an average power density of 670 kw/l and a peak power density of 1240 kw/l. The peak is 13% below the design peak power density. The peak occurs not in the actinide region, but at the inner boundary of the driver region. The power density is less in the actinide region because of the lower quantities of fissile material there. The

TABLE IV: COMPARISON OF FUEL TYPES

<u>Fuel Type</u>	Actinide Number Density (atoms/b-cm)	<u>N.D. (fuel)</u> <u>N.D. (ox. fuel)</u>	<u>Melting pt.</u> <u>For U</u> <u>Fuel (°C)</u>
Oxide	.022	1.00	2740
Carbide	.031	1.39	2480
Metal	.041	1.86	1155

N.D. = Number Density

beginning of the first cycle (BOC1) power densities for metal, carbide and oxide actinide fuels are 557 kw/l, 411 kw/l, and 288 kw/l, respectively. The central region flux averages 3.6×10^{15} n/cm²-sec over the three year burn. Because the limiting thermal conditions did not occur in the actinide region, a series of runs was performed with an increased central region flux of 5.0×10^{15} n/cm²-sec. Although this resulted in an unacceptable peak power density of 1920 kw/l in the driver region, the power density in the actinide region was well within allowable limits.

Assumptions

The following assumptions have been made in this paper:

1. It is possible during the first reprocessing to

remove all of the fission products and 99.9% of the uranium and plutonium isotopes. In future reprocessing schemes, the levels of Pu and U can be reset to these levels.

2. It is possible to fabricate oxide, metal, and carbide fuels from the actinide mix.

3. Reprocessing occurs two years after spent rods are removed from the core.

4. In the metal fuel, molybdenum was used as a diluent instead of zircalloy due to the unavailability of the latter's group cross-sections. Molybdenum has an atomic mass of 95.95 amu, compared to zirconium's mass of 91.22 amu. The two elements have similar total cross-sections within the range of the ABR's spectrum. Excluding inelastic scattering, the total cross-section of molybdenum fluctuates between 6 and 12 barns between 50 and 300 kev (24). At 300 kev, it has a value of 9 barns, which steadily decreases to 4 barns at 3 Mev, remaining constant until 10 Mev. Zirconium fluctuates between 7 and 9 barns in the 50 to 300 kev range, and is nearly identical to that of molybdenum above 300 kev. Such data that does exist indicates that zirconium will probably degrade the spectrum by inelastic scattering less than molybdenum does. It is thus assumed that any changes resulting from this substitution, although conservative in

nature, are insignificant.

5. A reactor in a given cycle may have its actinide fuel fabricated from the spent fuel of several reactors of the previous cycle. Thus, even though the usable actinide fuel diminishes in each cycle, there is always enough to fill the next cycle's actinide region.

NEUTRON SPECTRA

Spectra in the ABR

The ABR was designed so that the spectrum to which the actinides are subjected is determined by the driver region. The beginning of cycle (BOC) spectrum for the actinide region, metal fuel is shown in Figure 3. Also shown in Figure 3 is the spectrum of the Phenix reactor, as determined by NCINR8, and a representation of the capture and fission cross-sections of Am-241. This plot again emphasizes the importance of a hard spectrum reactor. Realizing that the horizontal axis is logarithmic, the ABR shows an obvious advantage over the Phenix reactor.

It is not possible to determine a definite median neutron energy with the NCINR8 code. It is only possible to narrow the median neutron energy down to a certain group which may span several hundred kev, as shown in Table V. A different criteria is thus needed to compare the hardness of various spectra. A flux averaged mean energy for each spectrum may be calculated by the following equation:

$$\bar{E} = \exp \left\{ \frac{\sum_{i=1}^{29} \phi_i \ln E_i}{\sum_{i=1}^{29} \phi_i} \right\} \quad (\text{eq. 1})$$

where E_i is the logarithmic mean energy in each group. The log of E_i is taken so that the higher energies are not weighted disproportionately.

FIGURE 3: SPECTRA OF PHENIX AND ACTINIDE REGION
(METAL FUEL) IN EBR-II COMPARED TO
CAPTURE AND FISSION CROSS-SECTIONS OF AM-241

TABLE V: NORMALIZED GROUP FLUXES IN
ACTINIDE AND DRIVER REGIONS

<u>Group</u>	<u>Upper Energy (ev)</u>	<u>Lower Energy (ev)</u>	<u>Lethargy Width</u>	<u>Flux in Act. Reg.</u>	<u>Flux in Driv. Reg.</u>
1	1.00E7	6.07E6	0.50	0.0043	0.0044
2	6.07E6	3.68E6	0.50	0.0193	0.0217
3	3.68E6	2.23E6	0.50	0.0496	0.0536
4	2.23E6	1.35E6	0.50	0.0769	0.0825
5	1.35E6	8.21E5	0.50	0.1049	0.1073
6	8.21E5	4.98E5	0.50	0.1441	0.1434
7	4.98E5	3.88E5	0.25	0.1516	0.1497
8	3.88E5	3.02E5	0.25	0.1445	0.1427
9	3.02E5	1.83E5	0.50	0.1152	0.1120
10	1.83E5	1.11E5	0.50	0.0785	0.0756
11	1.11E5	6.74E4	0.50	0.0448	0.0465
12	6.74E4	4.09E4	0.50	0.0233	0.0248
13	4.09E4	2.55E4	0.47	0.0181	0.0157
14	2.55E4	1.99E4	0.25	0.0128	0.0113
18	5.53E3	3.35E3	0.50	4.3E-4	2.2E-4
23	1.23E3	7.48E2	0.50	3.0E-5	8.3E-6
29	1.37E1	1.13E0	2.50	2.4E-14	8.7E-15

The value of $\bar{E}$ is 442 kev for the ABR and 180 kev for the Phenix. Although the Phenix has its peak at a higher energy than the ABR, the Phenix exhibits a much broader spectrum, which lowers the flux averaged mean energy.

The relative group fluxes for the metal fuel actinide region and for the driver region are tabulated in Table V. The spectrum in the driver region is slightly harder than that in the actinide region. The flux is higher in the driver region for the first five groups (above 820 kev). The peaks for both spectra are in the seventh group, between 388 and 498 kev. Only a very small fraction of the spectra are below 40 kev.

The driven ABR has a harder spectrum than an undriven reactor fueled with just the actinide mix (Figure 4). The spectra indicate that more neutron capture would occur in an undriven reactor than in a driven reactor, thus resulting in the buildup of more trans-plutonics. It is not known how the spectrum of an undriven reactor would change as the fuel composition of future cycles changes, or even whether criticality could be maintained. The mean energy for an undriven reactor is 240 kev, well below the 442 kev of the driven system.

Figures 5 and 6 show the BOC1 spectra for carbide and oxide fuel, respectively. Figures 3, 5, and 6 reveal that the spectra are nearly independent of fuel type,

Figure 4

SPECTRUM OF METAL FUEL
IN EBR-II CONFIGURATION WITHOUT DRIVER

particularly at the important high energies. The large dip in the spectrum in the 500 to 800 kev range of oxide fuel is the result of a large elastic scattering cross section in oxygen. The cross section in that range is 9.67 barns, as compared to one to four barns at neighboring energies. The mean energies for carbide and oxygen fuel are 430 and 404 kev, respectively, slightly below the value for metal fuel.

The BOC10 spectrum for oxide fuel using the second reprocessing scheme was determined. The actinide region contained 93% U-238, a much higher percentage than the 56% of the first cycle. U-238 normally softens a spectrum through inelastic scattering. However, little difference existed in the spectrum from that of the first cycle due to the dominance of the driver region. The mean neutron energy for oxide fuel was 387 kev, down from 404 kev in the first cycle. This is not as much as might be expected with such large amounts of U-238.

Figure 7 shows the end of cycle (EOC) spectrum for oxide fuel at the two different power levels. A softening of the spectrum occurs because of the depletion of the actinides. This depletion allows lighter elements, with greater losses per collision, to compete better for the neutrons. Further softening is caused by U-238 (high inelastic scattering), which makes up a greater percentage

Figure 5

SPECTRUM FOR CARBIDE FUEL,
BOC, FIRST CYCLE

Figure 6

SPECTRUM FOR OXIDE FUEL,
BOC, FIRST CYCLE

of the actinides due to its relatively slow transmutation rate. A minor effect is from the buildup of fission products. These effects are obviously greater at the higher flux. The mean energy values of the EOC spectra are 363 and 349 kev, compared to 404 kev BOC. Nonetheless, the spectra are nearly constant at the higher energies over the three year burn. In actual reactors, rods are replaced every 12 to 18 months, resulting in an even more constant spectrum and flux. With this as a justification, several of the later cycles were simulated by inputting a representative spectrum and holding it constant through the three year burn. (This saves computer time and money.) The results of a constant spectrum run and a variable spectrum run are nearly identical (Table VI). Although the second cycle is shown in the example, the constant spectrum results were only used in the seventh and later cycles in order not to introduce any unnecessary errors early.

In all these calculations, the driver region was assumed to be bare: it did not have the surrounding fertile blanket intrinsic to breeder reactors. An NCINR8 run was performed that included 25 metric tonnes of U-238 as a blanket. Although the spectrum was altered towards the outer edge of the driver region, the actinide region was not significantly affected. The mean energy was 393 kev compared to 404 without a blanket, but no discernable

Figure 7

EOC SPECTRA FOR OXIDE FUEL
AT 135 MW AND AT 5.0×10^{15} FLUX

difference appeared in the burn rates. The results justify the exclusion of the blanket region for runs determining burn rates in the actinide region.

Collapsed Cross Sections

Because the spectrum was nearly constant over the entire burn, it was possible to collapse the isotopic 29 group cross-sections in this spectrum by the following equation:

$$\sigma_x = \frac{\sum_{i=1}^{29} \sigma_{x,i} \phi_i}{\sum_{i=1}^{29} \phi_i} \quad (\text{eq. 2})$$

The resulting collapsed cross-sections are listed in Table VII. Only three isotopes have a value of $1/\alpha$ less than one; U-236 (0.65), U-238 (0.57), and Am-241 (0.61). U-238 and Am-241, however, after one capture become Pu-239 and Am-242, isotopes with very high values of $1/\alpha$ (9.89 and 12.30, respectively). Am-242 has the highest fission cross-section in this spectrum (2.58 barns). Other isotopes with high fission to capture ratios are Cm-243 (26.15), Cm-245 (14.48), and Cm-247 (13.86). U-238, which dominates the actinide region if not removed during reprocessing, has a fission cross-section of only 0.083 barns. This is less than half that of the next lowest value. Because of its poor value as a fuel, the quantity

of U-238 should be kept to a minimum in the actinide region.

Radial Flux Profiles

Figures 8 and 9 show the radial, nine-coarse-group flux profiles for the actinide and driver regions of the ABR. The nine groups are those described in Table VIII. In Figure 9, the values for groups 5 and 6 have been doubled because their lethargy widths are half that of the other groups. The inequality in lethargy widths also results in Groups 1 and 9 being disproportionately high. In the higher energy groups (1-5), the group fluxes in the actinide region are less than in the bordering driver region. This is due to a smaller proportion of fissile material in the actinide region than in the 50% enriched driver region. The fissile isotopes act as a source for fast neutrons and absorb neutrons before many scatterings have occurred. This effect is accentuated using oxide and carbide fuel, which introduce elastic scatterers in competition for the neutrons. In the lower energy groups (6-9), the flux drops entering the driver region, as the highly enriched fuel removes the lower energy neutrons. The flux in group nine levels out at the boundary of the depleted uranium blanket.

TABLE VI: CONSTANT SPECTRUM RUN
COMPARED TO VARIABLE SPECTRUM RUN

<u>Isotope</u>	<u>BOC Quantity (grams)</u>	<u>EOC Quantity w/const. spectrum (grams)</u>	<u>EOC Quantity w/var. spectrum (grams)</u>
U-234	14.5	23.9	24.7
U-235	31.1	19.1	19.0
U-236	28.7	26.2	26.2
U-238	5892.	5447.	5448.
Np-237	3062.	2035.	2032.
Pu-238	32.4	487.	504.
Pu-239	15.3	222.	226.
Pu-240	2.26	29.7	30.3
Pu-241	0.191	0.618	0.621
Pu-242	0.136	0.245	0.247
Am-241	688.	439.	435.
Am-242	31.4	30.5	30.7
Am-243	513.	396.	394.
Cm-242	1.67	1.10	1.04
Cm-243	2.97	3.00	3.03
Cm-244	138.	113.	113.
Cm-245	19.5	20.4	20.7
Cm-246	1.90	2.39	2.43
Cm-247	0.133	0.118	0.118
Cm-248	7.47E-03	1.11E-02	1.12E-02
Bk-249	1.46E-05	2.53E-05	2.49E-05

TABLE VII: CROSS-SECTIONS
COLLAPSED IN CONSTANT SPECTRUM

<u>Isotope</u>	<u>σ_f</u>	<u>σ_c</u>	<u>σ_f/σ_c</u>
U-234	0.545	0.307	1.80
U-235	1.373	0.257	5.35
U-236	0.197	0.301	0.65
U-238	0.083	0.146	0.57
NP-237	0.609	0.602	1.01
Pu-238	1.331	0.130	10.27
Pu-239	1.643	0.166	9.89
Pu-240	0.628	1.989	3.16
Pu-241	1.807	0.212	8.53
Pu-242	0.527	0.132	4.00
Am-241	0.497	0.813	0.61
Am-242	2.578	0.210	12.30
Am-243	0.460	0.326	1.41
Cm-242	0.747	0.376	1.98
Cm-243	2.079	0.079	26.15
Cm-244	0.747	0.376	1.98
Cm-245	2.098	0.145	14.48
Cm-246	0.543	0.100	5.43
Cm-247	1.964	0.142	13.86
Cm-248	0.581	0.100	5.81
Bk-249	0.316	0.186	1.70
Cf-249	1.876	0.159	11.80

Figure 8

RADIAL FLUX PROFILES
FOR COARSE GROUPS 1-5

Figure 9

RADIAL FLUX PROFILES
FOR COARSE GROUPS 6-9

TABLE VIII: COARSE GROUP STRUCTURE

<u>Coarse Group</u>	<u>Collapsed From fine Group(s):</u>	<u>Lethargy Width</u>	<u>Upper Energy (ev)</u>	<u>Lower Energy (ev)</u>
1	1-3	1.5	1.0E7	2.2E6
2	4	0.5	2.2E6	1.4E6
3	5	0.5	1.4E6	8.2E5
4	6	0.5	8.2E5	5.0E5
5	7	0.25	5.0E5	3.9E5
6	8	0.25	3.9E5	3.0E5
7	9	0.5	3.0E5	1.8E5
8	10	0.5	1.8E5	1.1E5
9	11-29	12.5	1.1E5	1.1E0

Alternative Arrangements

Figure 10 shows the spectrum averaged over the entire driver region, as well as the spectrum on the outer boundary of the driver region. Even when averaged over the entire region, the driver region's spectrum is harder than that of the actinide region, as was shown in Table V. The values of $\bar{E}$ are 464 kev for the entire region and 400 kev at the boundary, compared to 442 kev in the actinide

region. The spectrum at the edge, although softer than that in the central region, still has high values of $1/\alpha$. Figures 8 and 9 show the flux diminishing near the edge. Nonetheless, the hardness of the spectrum there indicates that rods could be dispersed throughout the core and still achieve fission rather than capture. Because burn rates vary proportionately with flux levels, flux flattening techniques would have to be employed. Placing actinide rods near the edge would further soften the spectrum there.

As fuel burns out over the three years, the flux level must be increased to compensate for the loss of fuel. In the actinide region, the flux typically increased from 3.2 to 4.1×10^{15} n/cm²-sec at 135 MW. If not offset by the competing effect of a softening spectrum, this would result in an increased burn rate with time. Calculations showed, however, the EOC burn rate to be only marginally higher than that at BOC.

Nonetheless, it was decided to simulate a run using a 40% enriched driver region to determine whether the increased flux would more than compensate for the softer spectrum. The mean energy for the 40% enriched core was 367 kev, compared to 404 kev with a 50% enriched driver. The flux ranged from 4.0×10^{15} BOC to 5.3×10^{15} n/cm²-sec EOC. Higher burn rates were achieved at 40% enrichment,

Figure 10

SPECTRUM AVERAGED OVER DRIVER REGION AND
SPECTRUM AT OUTER EDGE OF DRIVER REGION

but not without drawbacks. A greater number of actinides built up in the driver region. The importance of this is discussed later. More importantly, the reactor was not critical. This could be overcome by the use of reflectors or a larger core, but brings up the question of optimization.

Although this paper primarily discusses burn rates, the amount of trans-plutonics fissioned per unit power is a better indication of efficiency. Perhaps the 40% enriched core could be made critical if a smaller actinide region were used, but this is self-defeating. It is better to burn 10% of 10,000 grams of actinides than 100% of 100 grams, even though a lower burn rate occurs.

The intent of this paper, however, is to describe the behavior of the actinide material through repeated cycling, not to optimize the core. For this reason, the 50% enriched core is used exclusively. By holding other parameters constant, insight is gained by examining the burn rates. It is recognized, however, that if metal, carbide, and oxide fuel all burn actinides at the same rate, and other design factors proved equal, the metal fuel would be superior because of its higher actinide number density. A diluent such as molybdenum would be counter-productive in a driven reactor.

An inherent flaw in the NCINR8 code is its inability

to account for axial variations in flux and spectra. Since the flux decreases and the spectrum softens near the top and bottom of the core, lower burn rates would be expected in those areas. Thus, the burn rates presented are representative of only the central portion of the core.

RESULTS AND DISCUSSION

Burn Rates in the ABR

The rate at which the major actinides decay is shown in Figure 11 and Table IX. Figure 11 shows the number of grams of the major non-plutonium isotopes remaining per kilogram of actinide mix after each five year cycle. Only results for oxide fuel are presented because the decay rates for the different fuel types are nearly identical. This is shown in Table X. The nearly identical burn rates are the result of the driver region determining similar spectra for each fuel type. The burn rates using the second reprocessing scheme were very similar to the results using the first reprocessing scheme. This will be discussed in detail later.

It can be seen from Figure 11 and Tables IX and X that a significant rate of burnup for the trans-plutronics does occur in the actinide region. After ten cycles, 1.1% of the original Am-241 (0.8 of an original 70.0 grams) and 7.4% of the original Am-243 remain (3.2 of 43.4 g). Although this reduction requires 30 reactor years time, the occurrence of a similar reduction through natural decay would require 2810 and 27,750 years, respectively. Np-237 and Pu-239 would remain as the by-products of the natural α -decay. The straight lines occurring in Figure 11 for some isotopes indicate that the same fraction is burning out each

Figure 11

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

TABLE IX: PERCENT OF MAJOR ACTINIDES
REMAINING AFTER TEN CYCLES

Isotope	$3.6 \times 10^{15} \text{ n/cm}^2\text{-sec}^{**}$		$5.0 \times 10^{15} \text{ n/cm}^2\text{-sec}$	
	Percent Destroyed per Cycle*	Percent Remaining After 10 Cycles	Percent Destroyed per Cycle*	Percent Remaining After 10 Cycles
U-234	---	868. %	---	339. %
U-235	---	4.1%	---	1.7%
U-236	---	23. %	---	17. %
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%	---	0.1%
Cm-243	---	31. %	---	8.9%
Cm-244	21. %	10. %	27%	4.4%
Cm-245	---	36. %	---	17. %
Cm-246	---	148. %	---	104. %

*If consistent

** Average flux at 135 MW

TABLE X: QUANTITY OF ACTINIDES
REMAINING FROM KG SAMPLE

<u>Isotope</u>	<u>CYCLE</u>			
	<u>0</u>	<u>5</u>	<u>10</u>	
Np-237	299.7	39.45	5.81	Oxide Carbide Metal
		39.55	5.97	
		40.72	6.27	
Am-241	70.04	7.42	0.784	
		7.55	0.824	
		7.74	0.880	
Am-242	0.400	0.732	0.0843	
		0.728	0.0862	
		0.721	0.0881	
Am-243	43.40	11.86	3.20	
		11.93	3.29	
		12.22	3.50	
Cm-242	0.439	0.0194	0.0021	
		0.0192	0.0020	
		0.0190	0.0021	
Cm-243	0.0322	0.0791	0.0101	
		0.0765	0.0095	
		0.0738	0.0095	
Cm-244	10.70	3.73	1.11	
		3.64	1.08	
		3.57	1.08	
Cm-245	0.804	0.885	0.291	
		0.847	0.278	
		0.805	0.266	

cycle. Specifically, 36% of the BOC Am-241 is transmuted each cycle, 23% of the BOC Am-243, 32% of the BOC Np-237, and 7.5% of the BOC U-238. It is not obvious that this should be the case because of the changing isotopic ratios in each cycle. For example, using the second reprocessing scheme, U-238 accounted for 93% of the actinide region in the tenth cycle as opposed to 56% in the first cycle. Thus, one might expect the buildup of Am-241 from U-238 to offset its burnup. Figure 11 shows that this is not the case. This will be discussed in depth later.

The transmutation rate of Cm-244 is not much higher than its rate of natural decay. With its half-life of 17.91 years, 17.6% of a given quantity of Cm-244 naturally decays to Pu-240 after five years. Only 14.4% remains after fifty years, the equivalent of ten five-year cycles. Table IX shows that at a power of 135 MW, 21% of the BOC Cm-244 is transmuted every cycle. After ten cycles, 10.3% remains. The small difference in the natural decay rate and the ABR's transmutation rate can be attributed to the buildup of Cm-244 from Am-243 in the ABR. Am-243 is initially four times as abundant as Cm-244 and is thus an ample source of Cm-244 through neutron capture. Am-243 has a fission-to-capture ratio of 1.41, meaning that for every 100 fissions that occur, 71 captures occur. These captures represent a significant proportion of the 23% of the Am-243 that is transmuted every cycle. At a flux of

5×10^{15} n/cm²-sec, the burn rate of the Cm-244 is 27% per cycle. Only 4.4% remains after ten cycles.

Table XI lists the in-core time period required to reduce the quantities of those isotopes with constant decay rates to half of their original value. This determines a system half-life of these isotopes. The system half-lives do not include the two year cooling period per cycle. Nonetheless, the reduction is quite significant from natural half-lives that include 7376 years for Am-241 and 4.5 billion years for U-238.

Figure 11 shows that Cm-245 and Am-242 build up through the first two cycles, but peak and diminish at nearly constant rates in later cycles. This can be explained by considering the rate of change. The major production and loss terms of Am-242 are represented in the following equation:

$$\frac{dN^{242}}{dt} = N^{241} \sigma_Y^{241} \phi - N^{242} \sigma_C^{242} \phi \quad (\text{eq. 3})$$

where the superscripts represent the atomic mass of the americium isotopes. Initially, the number density of Am-241 in oxide fuel is 1.54×10^{-3} atoms/b-cm compared to 8.77×10^{-6} atoms/b-cm for Am-242. The relatively large amount of Am-241 results in a production of Am-242 that

TABLE XI: SYSTEM HALF-LIVES OF MAJOR ISOTOPES

<u>$3.6 \times 10^{15} \text{ n/cm}^2\text{-sec}^{**}$</u>			<u>$5.0 \times 10^{15} \text{ n/cm}^2\text{-sec}$</u>	
<u>Isotope</u>	<u>Cycle</u>	<u>System Half-life (years*)</u>	<u>Cycle</u>	<u>System Half-life (years*)</u>
U-238	9	26.8	7	19.3
Np-237	2	5.2	2	4.0
Am-241	2	4.6	2	3.3
Am-243	3	8.0	2	5.8
Cm-244	4	9.0	3	6.7

*Does not include two year cooling time per cycle

** Average flux at 135 MW

exceeds the losses. Am-241 is one of only three isotopes with a fission-to-capture ratio less than one ($1/\alpha = 0.61$). However, the amount of Am-241, the only major source of Am-242, is greatly reduced after several cycles. The loss term then dominates the above equation, resulting in the subsequent decrease of Am-242. Thus the buildup of Am-242, as well as Cm-245, is not a concern. Figure 11 shows the system half-lives of these isotopes occurs in the ninth cycle, the equivalent of 24 to 27 years.

In one kilogram of original actinide material, Cm-244 naturally decays in five years from 10.71 grams to 8.82 grams. Thus 1.89 grams of Cm-244 naturally decay. Simultaneously, Pu-240 increases from 1.16 grams to 3.01 grams, the difference of 1.85 grams obviously coming from the α -decay of Cm-244. In the ABR, 1.73 grams of Cm-244 is transmuted per kilogram of actinide material each five year cycle. This quantity is 0.16 grams less than through natural decay due to the buildup of Cm-244 from the neutron capture by Am-243. The buildup from Am-243 masks the actual amount of Cm-244 that α -decays to Pu-240. In the ABR, Pu-240 increases to 3.24 grams/kg, as opposed to 3.01 grams/kg through natural decay. Since the majority of the Pu-240 comes from the α -decay of Cm-244, the buildup of Pu-240 from neutron capture by Pu-239 is minor.

Activity Levels of Trans-plutonics

Figure 12 plots against time the activity levels of the trans-plutonics and Np-237 (i.e., the isotopes other than U and Pu) if one metric tonne of original actinide material decayed for 1000 years. The plutonium activity is the result of the decay of americium and higher isotopes. For the first 20 years, the majority of the activity is due to Cm-244. From 100 to 1000 years, Am-241 accounts for over 80% of the total. Initially the total activity is 1.4 million curies per metric tonne (of Np plus

trans-plutonics), but decreases to 60,000 curies after 1000 years. These numbers are low because isotopes such as thorium and radium build up but are not included in this model. The specific radioactivities should not be compared to those cited in the first chapter, because the removal of the U and Pu invalidates such a comparison.

Figure 12 shows the contribution of Np-237 to the total activity to be minor. Because of its half-life of 2.1 million years, Np-237 is not a serious radiological hazard. It thus acts as an unnecessary diluent that occupies 30% of the BOC1 actinide mix. However, additional steps would be necessary to remove Np during reprocessing, as it does not chemically separate with the U and Pu using traditional reprocessing methods. For this reason, Np-237 remained with the actinide mix in this study. Np-237 forms Pu-238 through neutron capture, which is readily fissioned in the ABR.

Figures 13 and 14 show the curie levels from a metric tonne of trans-plutonics after it has been through five cycles and ten cycles in the ABR, respectively. Trends similar to those described in the previous paragraph still exist. However, after ten cycles the Am-241 never accounts for more than 50% of the total and is exceeded in activity by Am-243 after 1000 years. After ten cycles the 1000 year activity is 1500 curies, down by a factor of 40 from the original actinide material. This indicates that by

recycling the actinides the mass of material requiring long-term isolation is greatly reduced.

Comparison of Reprocessing schemes

Table XII shows the BOC composition of a kilogram of actinides according to the two reprocessing schemes. Table XIII, the quantities of trans-plutonics burned, and Table XII show that the second reprocessing scheme is impractical in terms of burning the actinides. If U-238 is not removed with each cycle, it soon dominates the actinide region to the point where the ABR becomes simply a breeder. By the tenth cycle, 93.5% of the actinide region is U-238. The U-238 continued to build up and an equilibrium level was never reached. U-238 consistently burned at 7.5% per cycle, a relatively slow rate for the ABR. Thus, after reprocessing and the removal of the fission products and plutonium, U-238 constituted a higher percentage of the remaining material each cycle. Although less U-238 actually exists, the fuel in each generation may be supplied by several reactors of the previous generation. Little room is left for the trans-plutonics, so only a small quantity is burned. Am-241 decreases from 7.0% to 0.4% of the actinide region from the first to the tenth cycle. Am-243 decreases from 4.3% to 1.4%, and Np-237 decreases from 30.0% to 2.8%. Only 6.2 grams of the

Figure 12

ACTIVITIES INITIATED
BY NP-237 AND TRANS-PLUTONICS
FROM ORIGINAL ACTINIDE MATERIAL

Figure 13 ACTIVITIES ORIGINATED BY NP-237
AND TRANS-PLUTONICS FROM ORIGINAL
ACTINIDE MATERIAL AFTER 5 CYCLES

Figure 14
ACTIVITIES ORIGINATED BY NP-237
AND TRANS-PLUTONICS FROM ORIGINAL
ACTINIDE MATERIAL AFTER 10 CYCLES

trans-plutonics burned in the tenth cycle, as opposed to 35.2 grams in the first cycle. This included 1.5 grams of Am-241 and 32 grams of Am-243, down from 25.3 grams and 10.0 grams, respectively. Since it is desirable to optimize the quantity of trans-plutonics burned per megawatt of power, uranium must be removed during reprocessing. In contrast, the quantities of trans-plutonics burned increases if the first reprocessing scheme is followed. In the tenth cycle, 53.0 grams of trans-plutonics are burned, including 27.6 grams of Am-243. Am-243 constitutes 11.8% of the actinide region at the beginning of the tenth cycle. Again, this does not imply that the quantity of Am-243 is increasing, but only that it constitutes a higher percentage after reprocessing. Cm-244 increases from 1.1% to 3.9%. Although Am-241 and Np-237 decrease to 3.5% and 21.5% respectively, these levels are not nearly as low as those when uranium is not removed. 60

All actinides are long-lived when their daughter isotopes are taken into consideration. Thus there is no diluting effect if an isotope with a relatively short half-life, such as Cm-244 (17.9 yrs.), constitutes a higher percentage of the actinides in each successive cycle. The efficiency of burning the long-lived isotopes is not reduced. However, uranium, with its extremely long-lived, nearly stable isotopes, does act as an unnecessary diluent and should be removed.

TABLE XII: BOC COMPOSITION OF 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	.073	3.95	3.73	5.08	2.68
U-235	4.71	1.16	1.53	1.0	.808
U-236	2.66	2.50	3.5	1.93	3.04
U-238	562.1	564.5	802.7	561.6	935.3
Np-237	299.7	270.0	115.1	215.3	27.91
Pu-238	.095	3.07	2.97	2.68	.769
Pu-239	2.86	1.49	3.28	1.63	4.19
Pu-240	1.16	.254	.308	.475	.265
Pu-241	.480	.005	.007	.008	.006
Pu-242	.207	.001	.001	.001	.001
Am-241	70.04	53.65	22.70	24.88	4.10
Am-242	.40	4.95	2.21	3.63	.439
Am-243	43.41	70.84	29.98	118.3	14.0
Cm-242	.439	.137	.059	.091	.01
Cm-243	.032	.518	.222	.408	.050
Cm-244	10.71	21.58	9.28	38.56	4.81
Cm-245	.804	4.82	2.07	9.82	1.24
Cm-246	.097	.889	.382	4.26	.530

TABLE XIII: QUANTITIES OF TRANS-PLUTONICS
BURNED PER KG OF MATERIAL (G)

<u>CYCLE</u> <u>REP.</u> <u>SCHEME</u>	<u>1</u>	<u>5</u>		<u>10</u>	
	<u>I & II</u>	<u>I</u>	<u>II</u>	<u>I</u>	<u>II</u>
Pu-241	-.21	+.071	.049	+.12	+.042
Am-241	-25.26	-19.38	-8.22	-12.70	-1.48
Am-242	+1.64	-1.61	-.69	-1.30	-.16
Am-243	-9.98	-16.20	-6.85	-27.57	-3.21
Cm-242	-.33	-.049	-.021	-.033	-.004
Cm-243	+.16	-.16	-.069	-.14	-.019
Cm-244	-1.73	-4.58	-1.91	-8.77	-1.07
Cm-245	+.46	-.83	-.35	-2.18	-.26
Cm-246	+.28	+.011	+.008	-.47	-.056
<hr/>					
NET	-35.2	-42.7	-18.1	-53.0	-6.2

The actinide region's power density, an indicator of the amount of material fissioning, changes along with the isotopic composition. The results of Table XIV verify that more actinide fissioning occurs if the first reprocessing scheme is used. Using the first method, power density remains constant even though the amount of trans-plutonics burned increases. This is attributed to the smaller amounts of U-235, Pu-239, and Pu-241 burned. U-238 dominates the actinide region using the second reprocessing scheme. The power density is thus driven down because of the very small fission cross-section of U-238 (0.083 b).

Breeding of Plutonium in the Actinide Region

The rate of burnup of Am-241 and other actinides has invariably been independent of the isotopic constitution of the actinide mix. Even with large quantities of U-238 present, the buildup of the heavier actinides from U-238 was not enough to affect their burn rates. To determine the fraction of U-238 that was converted to higher isotopes, one kilogram of pure U-238 was placed in a representative constant-spectrum flux of 3.6×10^{15} n/cm²-sec for three years. This produced 35.5 grams of Pu-239 (3.55% of the original 10 kg of U-238) and less than 0.001 grams of trans-plutonics. The spectrum is hard enough, and the fission-to-capture ratio high enough, that the buildup of trans-plutonics from U-238 is almost

non-existent.

One kilogram of Pu-239 was then exposed to the same flux. This is not to suggest that a kilogram of pure Pu-239 would be placed into a five centimeter radius in an actual reactor. It is the percentages that are of interest, and any amount of Pu-239 could have been used. One kilogram was used so that the buildup of transplutonics from plutonium could be directly compared to the uranium case. The results are shown in the second column of Table XV. Only 53.9% of the original Pu-239 remains. Again, mere traces of the trans-plutonics build up because of the high fission to capture ratio. It can be calculated from the numbers in Table VII that the probability of a Pu-239 atom undergoing two neutron captures instead of one fission is only 2.2%. This is the probability of a Pu-239 atom producing Pu-241, the parent isotope of Am-241. The probability of four captures occurring to produce Am-243 is one in 2160. The small buildup of trans-plutonics is a direct result of maintaining a hard spectrum.

Two nearly independent processes occur in the actinide region. The first is the conversion of U-238 to Pu-239 (breeding). The second is the burning of the trans-plutonics. The initial loading of uranium and plutonium has virtually no effect on the burn rates of americium, curium, and the higher actinides. This explains the constant burn rates that occur regardless of the BOC

TABLE XIV: POWER DENSITY FOR METAL FUEL AT 135 MW

	<u>Cycle</u>		
	<u>1</u>	<u>5</u>	<u>10</u>
	<u>P.D. (kw/l)</u>	<u>P.D. (kw/l)</u>	<u>P.D. (kw/l)</u>
Scheme 1	557	558	558
Scheme 2	557	337	224

TABLE XV: COMPOSITION OF ONE KG OF PU AFTER EXPOSURE TO FLUX FOR THREE YEARS

<u>Isotope</u>	<u>Amount Remaining Per kg Pu-239 (g)</u>	<u>Composition of Initial Pu mix (g)</u>	<u>Composition after Flux (g)</u>
U-234	.002	---	.50
U-235	.076	---	.06
U-236	.017	---	.11
Np-237	---	---	.06
Pu-238	.086	1.97	12.5
Pu-239	538.8	595.4	321.1
Pu-240	37.6	241.7	205.3
Pu-241	1.08	100.1	49.2
Pu-242	.032	43.1	39.4
Am-241	.159	---	12.7
Am-242	---	---	.2
Am-243	---	---	1.7
Cm-244	---	---	.08

compositions and reprocessing schemes. The removal of as much plutonium and uranium as possible during reprocessing is necessary only to allow the loading of more trans-plutonics into the core: it is not necessary to prevent the buildup of trans-plutonics from offsetting the burnup.

As for the actual breeding in the actinide region, 3.5% to 3.6% of the U-238 was converted to Pu-239 each three years. This was independent of the fuel type, of the cycle, and of the reprocessing scheme, as shown in Table XVI. The numerator in the last column represents the net amount of Pu-239 that builds up assuming that only 53.9% of the BOC PU-239 remains at the end of three years. The 3.5% to 3.6% conversion represents a large increase in the plutonium levels in the actinide region each cycle. This is true even when U-238 is removed during reprocessing. Table XVI shows that the levels of Pu-239 typically increase seven to nine fold in a five year cycle. In addition, other isotopes of plutonium build up significantly, particularly Pu-238, which results from neutron capture by Np-237. The total plutonium in the actinide region can increase as much as fifteen times, depending on the initial conditions.

Because the isotopes of plutonium increase at different rates, the isotopic proportions change with each cycle. Even though the BOC quantity of plutonium is

constant using the first reprocessing method, the fractions of the fissile isotopes Pu-239 and Pu-241 are changing. This is shown in Table XVII. The increase in Pu-238 after the first cycle is due to the buildup from Np-237. This restructuring would be important if the plutonium were used to fuel another type of reactor. The restructuring is less important in the ABR, where the power density of the actinide region was previously shown to remain constant each cycle.

A kilogram of plutonium of the isotopic ratios of the discharge from a LWR was exposed to the constant-spectrum flux. This was to determine the extent to which trans-plutonics build up when the mixture already contains higher isotopes of plutonium. This mixture has the same isotopic ratio as the plutonium at the beginning of the first cycle. The kilogram of Pu includes 100.0 grams of Pu-241 and 43.0 grams of Pu-242. The three year burn resulted in the buildup of 14.7 grams of trans-plutonics or 1.47% of the BOC Pu. The majority of the trans-plutonics is Am-241, which results from a neutron capture by Pu-240 and β -decay of Pu-241. In the first cycle, Am-241 accounted for 7.0% of the actinide mix. Total plutonium accounted for 0.48% of the actinide mix. Plutonium is only one-fourteenth the quantity of Am-241. A 1.47% conversion of this quantity to Am-241 is not of concern when compared to the 36% Am-241 burn rate.

TABLE XVI: BUILDUP OF PLUTONIUM IN VARIOUS CYCLES

A	B	C	D	E	F	G	H
<u>Fuel</u>	<u>Cycle</u>	<u>Rep. Scheme</u>	<u>Pu-239 BOC (g)</u>	<u>Pu-239 EOC (g)</u>	<u>U-238 BOC (g)</u>	<u>E/D</u>	<u>E-.539D F</u>
Oxide	1	--	29.93	229.2	5889	7.66	3.6%
Oxide	10	II	45.29	379.5	10100	8.38	3.5%
Oxide	10	I	28.12	221.9	5882	7.89	3.5%
Metal	1	--	55.81	411.8	10980	7.38	3.5%
Carbide	5	II	48.04	442.8	11920	9.22	3.5%

TABLE XVII: ISOTOPIC PLUTONIUM CONCENTRATION
USING FIRST REPROCESSING METHOD

	CYCLE			
<u>Isotope</u>	<u>0</u>	<u>1</u>	<u>5</u>	<u>10</u>
Pu-238	1.97	64.41	63.65	55.95
Pu-239	59.54	30.44	30.96	33.94
Pu-240	24.17	4.50	5.28	9.89
Pu-241	10.01	0.38	0.09	0.19
Pu-242	4.31	0.27	0.02	0.03

Actinide Buildup in the Driver Region

Burning actinides in the central region of the ABR would be of little value if actinides were building up in the driver region to an equal or greater extent. Table XVIII shows the net gain or loss of isotopes in the driver region, actinide region (oxide fuel, fifth cycle), and total core. The gain or loss in the actinide region (second column) varies with fuel type, cycle, and reprocessing scheme. The gain or loss in the driver region (first column) is nearly constant in all cases. Oxide fuel was chosen as a worst case example, and the second column could be adjusted for the other fuel types by multiplying by the ratios of the number densities (Table IV, col.3). The buildup of trans-plutonics in the driver region is negligible, despite the region's comparatively large volume. The ABR is still a net burner of trans-plutonics, even when using oxide fuel. The trans-plutonics created in the driver region have no effect on the calculations involving the actinide region when only meaningful digits are retained. The amount of Np-237 burned in the actinide region, however, is more than offset by that created in the driver region for oxide fuel. This is not the case with carbide or metal fuel. The amount of plutonium that builds up in the actinide region is insignificant compared to the 15,800 grams that build up in the driver region. The

importance of Table XVIII lies not in the numbers themselves, but in the relative magnitude of the numbers. It is conceivable that the quantities of trans-plutonics burned can be increased, but it is not conceivable that the amount that builds up from Pu and U will be of the same magnitude.

Table XVIII does not include the buildup of actinides in the blanket region. Preliminary results from the radial blanket, however, indicate that only a minor buildup of trans-plutonics does occur. This is due to the much lower flux levels in the blanket region, which averages only $5.3 \times 10^{13} \text{ n/cm}^2\text{-sec}$ over the three year burn. This flux level is two orders of magnitude less than that in the actinide region.

The incentive for the ABR thus far has been to create an alternative to disposing of the long lived radioactive waste by geological isolation. The fact is, however, that virtually no actinides build up in any region of the ABR. Exclusive use of very hard spectrum reactors would thus prevent the problem of actinide wastes from occurring in the first place. This is preferable to the present method of creating the waste and then attempting the politically difficult task of disposal. Although it would still be necessary to dispose of the waste that has already accumulated, a conversion from LWRs to hard spectrum reactors would prevent this problem from compounding.

TABLE XVIII: NET GAIN OR LOSS OF ACTINIDES IN ABR

<u>Isotope</u>	<u>Net Gain or Loss in Driver Region (g)</u>	<u>Net Gain or Loss in Act. 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	-32,470
Np-237	+1324	-948	+376
Pu-238	+63.1	+398	+461
Pu-239	+15,430	+207	+15,640
Pu-241	+3.41	+0.8	+4.2
Pu-242	+0.04	+0.11	+0.15
Am-241	+0.10	-202	-202
Am-242	+7.21E-4	-16.5	-16.5
Am-243	+2.38E-4	-170	-170
Cm-242	+1.77E-3	+17.3	+17.3
Cm-243	+2.54E-5	-1.5	-1.5
Cm-244	---	-34	-34
Cm-245	---	-8.8	-8.8
Cm-246	---	+0.12	+0.12

Buildup of the Higher Actinides

Figure 15 shows the buildup of the higher actinides for oxide fuel at 135 MW. Although most of the isotopes do build up, the levels that are obtained are orders of magnitude below the levels of the major actinides. From the original kilogram sample, the combined total of the isotopes reaches a peak at about 0.3 grams. The majority of this is Cm-243 and Cm-246. After several cycles, these isotopes diminish as their parent isotopes diminish. Although the levels of californium and berkelium still increase after ten cycles, they exhibit a tendency to level off, and never combine to more than 0.0001 grams. The buildup of the higher actinides, a major concern in designing the ABR, is virtually non-existent. The problem of building up heavier isotopes through neutron absorption can be completely disregarded due to the high fission-to-capture ratios in the ABR's spectrum.

Figure 15 shows only the buildup of the higher actinides using the first reprocessing method. To show a similar figure for the second scheme would be redundant, as the results are of the same magnitude, although not exactly the same. The point is that these levels are insignificant.

Burnup at the Higher Flux Level

Figure 16 and Table IX show the decay rates using the

Figure 15

BUILDUP OF MINOR ACTINIDES FROM INITIAL
KG SAMPLE, AT 135 MW DRIVER POWER

higher flux of 5.0×10^{15} n/cm²-sec. The percent of actinides destroyed per cycle increases at a rate almost proportional to the increase in flux, which averages a factor of 1.39 higher than the flux at 135 MW. This is shown in Table IX. The percent of actinides remaining from BOC1 after ten cycles decreases to 0.2% for Am-241, 2.7% for Am-243, 0.6% for Np-237, and 4.4% for C-244. Isotopes such as U-234 and Cm-246 do not build up as much using the higher flux. A comparison of the quantities remaining after ten cycles (oxide fuel) at the two flux levels is shown in Table XIX. A comparison of the quantities burned in the first cycle is also shown. The last column indicates that the quantities burned are slightly less than proportional to flux due to the softer spectrum at EOC.

Even at the higher flux, the burn rates of the trans-plutonics remain independent of the uranium and plutonium concentrations in the region. The buildup of trans-plutonics is still a very small fraction of the amount that burns out. Of the BOC U-238, 4.4% to 4.5% is now converted to Pu-239. This is 1.25 to 1.30 times the amount converted at 135 MW. The increase in conversion rate is slightly less than the increase in flux. As mentioned previously, less than .001 grams of trans-plutonics were formed from a kilogram of U-238 at 135 MW. Increasing this amount by a factor of 1.39 has no appreciable effect. Table XX shows the amount that the

Figure 16

DECAY OF MAJOR ACTINIDES FROM INITIAL
KG SAMPLE, AT 5.0×10^{15} FLUX

TABLE XIX: COMPARISON OF THE TWO POWER/FLUX LEVELS

A	<u>Amount Remaining After 10 Cycles</u>			<u>Quantities Burned In First Cycle</u>		
	B Original Amount (g/kg)	C 135 MW	D 5E15	E 135 MW	F 5E15	G Ratio F/E
<u>Isotope</u>						
Np-237	299.7	5.81	1.69	-100.3	-128.4	1.28
Am-241	70.0	.784	.150	-25.2	-32.0	1.27
Am-242	.40	.0843	.0162	+1.64	+1.75	1.07
Am-243	43.4	3.20	1.19	-9.98	-13.0	1.30
Cm-242	.439	.0021	.0005	-.330	-.319	0.97
Cm-243	.0322	.0101	.0028	+.161	+.257	1.60
Cm-244	10.7	1.11	.473	-1.73	-2.54	1.47
Cm-245	.804	.291	.138	+.464	+.572	1.23
Cm-246	.0965	.143	.100	+.0274	+.0421	1.54

higher actinides build up after five and ten cycles. Although a greater quantity builds up with the increased flux, the levels again do not warrant any serious concern. The levels of some isotopes peak and then decrease, as indicated in the last column.

Optimizing the ABR

This section has shown that significant decay rates of actinides can be achieved in an actinide burning reactor. It will now be shown that on a per megawatt basis, one ABR could handle the discharge of two or more LWR's. This can be achieved by distributing the actinides throughout the core and increasing the amount of actinide fuel. Improving reprocessing techniques could also increase the amount of trans-plutonics burned.

The total amount of actinides in the central region of the ABR is 19.5 kg for metal fuel, 14.6 kg for carbide fuel, and 10.5 kg for oxide fuel. A 1000 MWe LWR produces about 15 kg of Np, 4 kg of Am, and 1 kg of Cm every year (19). Thus, even using metal fuel, three ABRs of the type and size discussed would be required to handle one year's discharge from one LWR. The in-core time period in the ABR required to reduce the levels to those shown in Table IX is thirty years. This means that ninety reactor years in the ABR would be required to achieve those levels for one

TABLE XX: BUILDUP OF HIGHER ACTINIDES AT
135 MW* AND 5×10^{15} N/CM²-sec

(135 MW LEVELS LISTED FIRST)

Grams/Kg Original Material

Isotope	Cycle			Peak Cycle
	0	5	10	
Cm-247	1.23E0	8.76E-1	7.23E-1	1
		1.04E0	6.17E-1	1
Cm-248	8.04E-3	1.19E-1	1.52E-1	10
		1.58E-1	1.72E1	8
Bk-249	--	2.88E-4	3.90E-4	--
		4.97E-4	5.80E-4	8
Cf-249	--	4.99E-3	7.81E-3	--
		7.52E-3	1.01E-2	9
Cf-250	9.65E-5	2.56E-4	4.76E-4	--
		4.81E-4	7.95E-3	--
Cf-251	--	1.43E-5	3.43E-5	--
		3.05E-5	6.46E-5	--

*Avg.Flux = 3.6×10^{15} N/CM²-SEC

year's discharge. Clearly this will not suffice. Future work must include optimizing the ABR, to which end the following discussion will hopefully be of some value.

A greater quantity of actinides could be burned if the actinide region were enlarged. The 5 cm actinide region

occupies only 1.56% of the core. Increasing the radius to 10 cm quadruples the volume of the actinide region, and has little additional effect on the spectrum. The spectrum of the entire driver region is harder than that of the actinide region. It is thus feasible to have several actinide regions distributed throughout the core rather than one central region. Expanding on this idea, actinide rods could be distributed through the entire driver region, either uniformly or in increasing quantities towards the center. For example, each actinide rod could be in the center of a lattice of "driver rods", creating a localized version of the reactor used in this paper. A reactor of this nature would have to have enough driver rods to maintain criticality and a hard spectrum through the changing composition of the actinide rods. Table XXI shows the results of a simulated run in which every fifth rod was an actinide metal rod. The actinide rods were uniformly distributed throughout the core. The 20% actinide composition corresponds to 13 times the amount of actinides that were contained in the 5 cm central region. Out to a radius of 20 cm, the burn rates for Am-241 and Am-243 were only slightly less than the respective 36% and 23% burn rates achieved in the central actinide region. The reduced burn rates correspond to a flux that is proportionately less than the 3.6×10^{15} n/cm²-sec of the 5 cm central actinide region of the ABR. Contrary to the

TABLE XXI: PERCENT OF ACTINIDES DISTRIBUTED
THROUGHOUT CORE BURNED PER CYCLE

<u>Radial Segment (cm)</u>	<u>Avg Flux (x10E15)</u>	<u>Avg P.D. (kw/l)</u>	<u>Perc. Am-241 Burned</u>	<u>Perc. Am-243 Burned</u>	<u>Perc. Np-237 Burned</u>
0-10	3.43	1003	33%	21%	29%
10-20	3.18	969	33%	20%	28%
20-30	2.83	834	29%	18%	25%
30-40	1.31	414	15%	9%	13%

total core increase of Np-237 in an oxide fuel ABR, Np-237 burns out to 7.6% of its initial level out to 20 cm. After 20 cm, the flux drops significantly, as do the burn rates, but it must be recognized that no flux flattening techniques have been employed. The spectrum (Figure 17) still favors fission to neutron capture to such an extent that only 0.006 grams of Cf and Bk builds up in the entire core. The mean energy value for neutrons is 438 kev, slightly less than the 442 kev in the 5 cm metal actinide region.

The thermal output of the modeled ABR is an order of

Figure 17

SPECTRUM WITH ACTINIDES
DISTRIBUTED THROUGHOUT CORE

magnitude below planned LMFBRs. Superphenix, for example, is planned to have a thermal output of 3000 MW (16). An increase in the size and output of the ABR could be obtained by utilizing a parfait or a modular design. These designs alternate driver and blanket regions. If a ten-fold increase as described in the previous paragraph were combined with a twenty-fold increase corresponding to a larger reactor, the quantity of actinides burned would increase 200 times. One ABR could then handle the waste output of two 1000 MWe LWRs. These are crude approximations, but the point is that it is not inconceivable to design an LMFBR on the order of 1000 MWe that would burn the actinide waste of several LWRs. Beaman has shown that an equilibrium can be reached between one 1200 MWe LMFBR and three comparably sized LWRs (1). Beaman also states that an LMFBR of this size can accommodate 464 kg of recycled actinides without seriously affecting reactor performance (2).

Another method of increasing the quantity of actinides burned has already been briefly discussed. Calculations for the first cycle showed that a 40% enrichment in the driver region resulted in a higher burn rate of the actinides. The higher burn rates were due to achieving a higher flux. However, a larger core would have been necessary, and it is not certain whether the actinide burnup per megawatt was improved. If the low-enrichment

actinide rods were to be distributed throughout the core, a higher enrichment of the driver rods should perhaps be used for compensation. Since the 50% enrichment that was used in the ABR was in actuality a first guess, it would be unrealistic to expect that this enrichment results in the optimum burnup of actinides. Further studies should be directed towards reactor optimization. The 99.9% removal of uranium and plutonium during reprocessing used in this paper approaches today's economical limits. A higher level of extraction would result in an increase in actinides burned. The initial loading in the first cycle contained only 12.6% trans-plutonics and 30% neptunium, with the remainder being uranium and plutonium. Having the capability to completely separate the trans-plutonics and/or neptunium could increase the quantities of trans-plutonics burned several times.

To summarize this discussion, one should not think of the ABR utilized in this paper as the ultimate design for burning actinides. Rather, it is a vehicle for developing certain ideas. This computational model is used to help understand the behavior of the actinides in a hard spectrum, and provides a stepping stone in developing an optimal actinide burning reactor.

SUMMARY AND CONCLUSIONS

The results of continuously recycling actinides through a hard spectrum EBR-II type reactor, as described in this paper, are summarized below.

1. The reactor should be designed so that the spectrum is determined by the driver region and is as hard as possible. The spectrum for the ABR was nearly constant over a three year burn.
2. The rates of burnup are independent of fuel type, but the quantity burned is proportional to the number density of the actinides, making metal fuel superior in this respect.
3. As much uranium and plutonium as possible should be removed during reprocessing. This is so that a greater quantity of trans-plutonics can be burned, not because higher actinides build in. If not removed, U-238 occupies 93% of the actinide region by the tenth cycle.
4. The thermal limits are determined by the driver rods, which have a power density more than twice that of the actinide rods.
5. Two nearly independent processes are occurring in the actinide region - the conversion of U-238 to Pu-239 (breeding), and the destruction of the transplutonics. At constant power (135 MWth), 3.55% of the U-238 is

converted to Pu-239, but only a negligible amount converts to trans-plutonics. Simultaneously, 36% of The Am-241 and 23% of the Am-243 are transmuted each five year cycle regardless of the initial loading of U and Pu.

6. The buildup of the higher actinides (Cf and Bk) is insignificant, with less than 0.0001 grams of Cf and Bk forming per kilogram of actinide material after ten cycles at 135 MW. The amount that builds up in the driver and blanket regions is still negligible.

7. The 1000 year activity from the trans-plutonics is reduced by a factor of 40 after ten cycles.

8. A higher flux results in burn rates that increase almost proportionately with the flux. There is still a very minor buildup of the higher actinides.

9. An ABR with an electrical output of 1000 MW could handle the waste of at least two comparably-sized LWRs.

10. Recycling the actinide waste indefinitely as described in this paper would help eliminate long-term storage considerations for nuclear waste and simultaneously produce useful electrical power.

Although the emphasis in this paper has been to show that an ABR can be designed to destroy the long-lived discharge from a LWR, it should be pointed out that virtually no actinides build up in any region of the ABR.

This is an important point. Perhaps reactors should not be specifically built to destroy actinides, but rather all nuclear reactors should be designed with very hard spectra so that no actinide problem is created. This paper has shown that actinides do not build up regardless of the original composition of the actinide region. Rather than creating a problem and then striving to destroy it, why not design reactors so that the problem does not exist? This would be possible through the exclusive use of hard spectrum reactors.

REFERENCES

1. S.L. Beaman and E.A. Aitken, "Feasibility Studies of Actinide Recycle in LMFBRs as a Waste Management Alternative", Proceedings of Nuclear Regulatory Commission Workshop on the Management of Radioactive Waste: Waste Partitioning Workshop as an Alternative, NR-CONF-001, June 1976.
2. "Cooperative Nuclear Data and Methods Development", Liquid Metal Fast Breeder Reactor Program, Sixth Quarterly Report, GE-FBRD, GEFR-14074-6, April 1977.
3. A.H. Robinson, "Studies of Actinide Waste Burnup and Use in Fast Spectra, Quarterly Report, October 1, 1977 - December 31, 1977", document RLO/2227/T32-4 (1978).
4. A.H. Robinson, "Studies of Actinide Waste Burnup and Use in Fast Spectra, Quarterly Report, January 1, 1978 - March 31, 1978", document RLO/2227/T32-6 (1978).
5. J.B. Nelson, A.W. Prichard, P.E. Schofield, A.H. Robinson and B.I. Spinrad, "Studies on the Properties of Hard-Spectrum, Actinide Fissioning Reactors", submitted to the International Conference on Nuclear Waste Transmutation, Austin, Texas, July 1980.
6. Report to the President by the Interagency Review Group on Nuclear Waste Management, Washington, D.C., March 1979.
7. B.L. Cohen, "High Level Radioactive Waste from Light Water Reactors", Reviews of Modern Physics, Vol. 49, No. 1, pp. 1-20, 1977.
8. Significant Actinide Activities In The LWR And LMFBR Nuclear Fuel Cycles, U.S. Environmental Protection Agency, Wash, D.C., pp. A5-A14, Oct. 1974.
9. J.J. Cohen, D.G. Coves and L.D. Ramspott, "Lead Enapsulation of Nuclear Waste: A Risk Assessment", Trans. Am. Nucl. Soc., 34, pp. 350-351 (1980).
10. J.O. Blomeke and A.G. Croff, "An Overall Assessment of Actinide Partitioning and Transmutation", Trans. Am. Nucl. Soc., 34 pp. 417-419 (1980).
11. "Argonne News", Vol. 28, 5A, June 1978.

12. F. Girardi, "Overview of Partitioning and Transmutation Studies in the European Community", Trans. Am. Nucl. Soc., 34 p. 411 (1980)
13. A.G. Croff, C.W. Alexander and J.W. Wachter, "Transmutation and Fuel Cycle Impacts", Trans. Am. Nucl. Soc., 34 pp. 413-414 (1980)
14. B.I. Spinrad, "Plutonium Fueled Fast Reactors as Secure Fuel Factories: Synergism with Other Reactors and Fuel Cycles", OSU-NE-7705, pp. 16-20, Nov. 1977
15. G.W. Shirley and A.H. Robinson, "A Very Fast Spectrum Reactor for Actinide Burning", ANS Western Regional Student Conference (1977)
16. Liquid Metal Fast Breeder Reactor Program Plan, Volume 6, Core Design, prepared for the Division of Reactor Development & Technology, U.S. Atomic Energy Commission by LMFBR Program Office, Argonne National Laboratory, Aug. 1968
17. R.W. Benjamin et. al., "A Consistent Set of Heavy Actinide Multigroup Cross Sections", Savannah River, DP-1394.
18. R.W. Benjamin, "Status of Measured Neutron Cross Sections of Trans-actinium Isotopes for Thermal Reactors," Savannah River, DP-MS-75-87, Nov. 1975
19. F.M. Mann and R.E. Schenter, "Actinide Cross Sections Calculations and Evaluations", Trans. Am. Nucl. Soc., Toronto meeting, Nov. 1976
20. F.M. Mann and R.E. Schenter, "Calculated Neutron Capture Cross Sections to the Americium Ground and Isomeric States", Nucl. Sci. Eng., 63, p.242 (1977)
21. B.C. Finney and D.W. Tedder, "Actinide Partitioning Process for Fuel Cycle Wastes", Trans. Am. Nucl. Soc., 34 pp.412-413 (1980)
22. H. Kusters, M. Lalovic and H.W. Wiese, "Fuel Handling, Reprocessing, and Waste and Related Nuclear Data Aspects," submitted to International Conference on Neutron Physics and Nuclear Data For Reactors and Other Applied Purposes, Harwell, U.K., Sept. 1978

23. J. Leary and H. Kittle, Advanced LMFBR Fuels,
Topical Meeting Proceedings, Tuscon, Ariz., Oct.
1977
24. Neutron Cross-sections, BNL-325, NY (1958)

APPENDIX

APPENDIX - DESCRIPTION OF NCINR8

The NCINR8 (AP-1,AP-2) code was written specifically for the purpose of modeling fast reactors, similar to the FCC Fundamental Mode Fast Reactor Code (AP-3). It uses multigroup shielded cross-section data in the Bondarenko format to determine spectra, perform one-dimensional diffusion calculations, and simulate isotopic burnup for an inputted interval of reactor time. In addition to cataloging the core inventory throughout the burn, inventories for the 25 isotopes are also followed for a 1000 year period following the burn. The 29 group cross-section data, a preliminary version of ENDF/B-V, was provided by R.E. Schenter (AP-4 - AP-7).

Input variables to NCINR8 include core size, reactor power and geometry, number and size of different regions, unit cell geometry, fuel pin radius, gap thickness, clad thickness, and array pitch. Sodium and helium are possible coolants, and structural material includes iron, chromium, manganese, nickel, and molybdenum. Molybdenum can also be used as a diluent, as can oxygen and carbon.

NCINR8 starts the sequence of operations by reading the cross-section library and calculating the regional number density of each isotope. Each region is initially treated as infinite and has a fine group spectrum calculated by iterating on the elastic down-scattering

source. Converging the difference in successive fluxes in each group to less than .0001 typically requires 10 to 20 iterations. An option exists to by-pass these calculations and input a fixed spectrum.

After k_{inf} is calculated for each region, the 29 fine group fluxes and cross-sections are collapsed to an inputted number of coarse groups. In this thesis, nine coarse groups were used. A one-dimensional diffusion calculation is then performed to determine the spatial flux distribution for the coarse groups. A neutron balance is performed to determine leakage from each coarse group and region. A new spectrum is then calculated for each region, again by iterating on the elastic down-scattering source. If one region is dominated by another, as the actinide region is by the driver region in the ABR, it may take up to 35 iterations to converge. These new spectra are then used to recalculate the collapsed cross-sections, which in turn determine new coarse group spatial flux distributions. This process is repeated an inputted number of times.

The spectra and flux distribution are outputted for each region, and the burnup calculations commence. The basic burnup equations for actinides have the form:

$$\frac{dN_i}{dt} = -\lambda_i N_i - \sigma_i^a \phi_t N_i + \lambda_k N_k + \sigma_j^c \phi_t N_j$$

where N_i = density of nuclide i

σ_i^a = spectrum averaged absorption cross-section
for nuclide i

ϕ_t = total flux

It is possible in this model for a nuclide to be formed from the decay or capture of more than one nuclide, due to short β -decay half-lives. An example of this is Cm-244, formed from the neutron capture of both Am-243 and Cm-243.

A fourth order Runge-Kutta technique was used in calculating the burnup equations. Regional inventories are outputted at given time periods. Spectra and flux profiles are updated after an inputted interval of time, and the burnup equations continue. At the completion of the entire burn, a summary of the BOC and EOC inventory for the core is outputted. A decay of up to 1000 years is then performed, either for the entire core or for a specific region, again using the Runge-Kutta technique. With each inventory, during the actual burn or subsequent decay, a hazard factor or activity level is computed per isotope, depending on the version of NCINR8 library used.

The NCINR8 library contains the necessary group structures, fission neutron fractions per group, and parent - daughter relationships through decay and capture schemes. For each of the 35 isotopes, the library contains the atomic mass number, theoretical density, decay constants, and shielding factors. Each group of each isotope has data for average neutrons per fission within that group, average loss of the scattering angle, and average lethargy gain per

collision, as well as fission, capture, total, and elastic and inelastic scattering cross-sections. Also provided are inelastic scattering cross-sections from group to group.

REFERENCES FOR APPENDIX

- AP-1 A.H. Robinson, "Studies of Actinide Waste Burnup and Use in Fast Spectra, Quarterly Report, October 1, 1977 - December 31, 1977", document RLO/2227/T32-4 (1978).
- AP-2 G.W. Shirley and A.H. Robinson, "A Very Fast Spectrum Reactor for Actinide Burning", ANS Western Regional Student Conference (1977)
- AP-3 W.W Little, Jr. and R.W. Hardie, "FCC-IV, A Revised Version of the FCC Fundamental Mode Fast Reactor Code," BNWL-450, Aug. 1967
- AP-4 R.W. Benjamin et. al., "A Consistent Set of Heavy Actinide Multigroup Cross Sections", Savannah River, DP-1394.
- AP-5 R.W. Benjamin, "Status of Measured Neutron Cross Sections of Trans-actinium Isotopes for Thermal Reactors," Savannah River, DP-MS-75-87, Nov. 1975
- AP-6 F.M. Mann and R.E. Schenter, "Actinide Cross Sections Calculations and Evaluations", Trans. Am. Nucl. Soc., Toronto meeting, Nov. 1976
- AP-7 F.M. Mann and R.E. Schenter, "Calculated Neutron Capture Cross Sections to the Americium Ground and Isomeric States", Nucl. Sci. Eng., 63, p.242 (1977)