THE PRODUCTION OF ^{238}Pu IN COMMERCIAL LIGHT WATER REACTORS

December 1969

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THE PRODUCTION OF ^{238}Pu
IN COMMERCIAL LIGHT WATER REACTORS

by

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I. INTRODUCTION

Dr. Glenn T. Seaborg has stated* that 900 kilograms of Pu-238 will have to be provided to satisfy the estimated needs for this isotope by 1980. The present production rate of Pu-238 will not satisfy this estimated requirement. Commercial reactors may prove to be the only economic irradiation facilities of adequate size available to meet this need. Thus, based on prior work, the contractor proposed this study to evaluate the technical and economic feasibility of producing Pu-238 in commercial water reactors.

In any reactor that utilizes U-235 as the initial fuel, Pu-238 is produced in significant quantities. However, since the major portion of the fuel in water reactors of current design is U-238, most of the plutonium produced during the irradiation process is Pu-239 and the higher isotopes of plutonium. One can see that, although a significant quantity of Pu-238 (approximately 1% of the total) is contained in the spent fuel, an expensive isotopic separation process would be necessary to recover the Pu-238.

Pu-238 can be produced in a relatively pure form in commercial water reactors by irradiating target capsules containing Np-237. Up to the time

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*USAEC News Release No. S-43-69, Remarks by Dr. Glenn T. Seaborg, Chairman, U. S. Atomic Energy Commission, American Nuclear Society Winter Meeting, Special Session on Criticality Problems of Synthetic Actinide Elements, San Francisco, California, Dec. 2, 1969 - The Synthetic Actinides - From Discovery to Manufacture.

water reactor technology had progressed to the point that burnups of approximately 30,000 Mwd/mtU could be attained, the addition of Np-237 would have been somewhat impractical. As technology has improved, design fuel burnups have increased — attended by the addition of soluble poison to the moderator to control excess reactivity. As the design burnup of water reactor cores increased through the use of soluble poison in the moderator, the poison concentration required to control the initial excess reactivity became great enough to cause the moderator temperature coefficient of reactivity to be positive. Reluctance on the part of the AEC Regulatory Staff to accept reactor designs having positive moderator temperature coefficients caused the designers to substitute burnable poison rods, separately encapsulated from the fuel, for part of the reactivity control; that is, the soluble poison concentration is reduced to the point where the moderator temperature coefficient of reactivity is either zero or negative and the additional excess reactivity is controlled by fixed burnable poison rods. In a typical pressurized water reactor, these burnable poison rods are in the form of Pyrex (0.0866 grams natural boron per cm³) glass tubes clad in stainless steel. The rods look very much like the ordinary fuel rods which make up the fuel assemblies for the core. In a typical design, there are over 1000 burnable poison rods grouped in clusters and distributed throughout the core in vacant rod-cluster-control guide tubes. Spaces are available for additional burnable poison rods.

The study described here has been concerned with replacing some of the burnable poison rods in a typical PWR core with rods containing Np-237 in amounts sufficient to give an initial core reactivity the same as the design value. If the Np-237 behaved identically with the burnable poison material during fuel burnup, then neutrons normally captured in the burnable poison would convert the Np-237 to Pu-238, and at the same time serve the safety function of keeping the moderator temperature coefficient of reactivity either zero or negative. Unfortunately, Np-237 and the Pu-238 which accumulates as burnup progresses do not behave identically with a typical burnable poison. If some of the burnable poison rods are removed and replaced by rods containing Np-237, those neutrons normally captured in the portion of the burnable poison now removed will be utilized to produce Pu-238. The non-positive temperature coefficient requirement is also satisfied. In other words, more neutrons will be captured in the Np-237 rods than would have been captured in the burnable poison rods, resulting in some reduction in burnup. However, some of these neutrons, namely those which would have been captured in the burnable poison, are captured in Np-237 to form the useful product Pu-238.

A one-dimensional model has been used to calculate a base for the reactivity-time behavior of a conventional PWR as designed. Cross-section measurements for the isotopes in the Pu-238 production chain were reviewed extensively and a set incorporated in the model to allow calculations of both

Np-237 and Pu-238 masses in target capsules as a function of burnup. A parametric study having the number of Np-237 rods substituted, the amount of Np-237 per rod, and the position of the Np-237 rods in the reactor as variables was performed. Results of the study indicate that this method of irradiating Np-237 in commercial reactors to produce Pu-238 is, indeed, practical. The Np-237 rods can be arranged in the reactor so that the power distribution as a function of burnup is not affected significantly. The study indicates that the lightest loading of Np-237 per rod gives the highest production rate of Pu-238 and also the lowest value of the undesirable impurity Pu-236. The calculations further indicate that approximately 1 kilogram of Pu-238 can be produced by this method for each full-power day lost as a result of irradiating the Np-237. In at least one practical case, replacing approximately 200 of the burnable poison rods with sufficient Np-237 to balance the initial reactivity yielded, in one year, approximately 15 kilograms of Pu-238. Pu-236 impurities in the final product are calculated to be 4 to 5 ppm. It is believed that the calculated Pu-236 concentration is quite conservative, since comparison with measured values in Yankee fuel shows the calculations to be significantly higher than the observed values.

Estimates of costs of producing Pu-238 by this process were made with the assistance of information gathered by visits to reactor fuel fabricators, fuel reprocessors, and electrical utilities having operational reactors.

Although considerable uncertainty is still associated with many of the estimates obtained, it appears possible to produce Pu-238 by this process for somewhere between \$134 and \$282 per gram.

The total Pu-238 production capability by the method studied has been estimated to be between one and two thousand kilograms over the ten-year period beginning in 1972. Although the estimates described in Section VIII of this report are conservative, they demonstrate that the process is commercially attractive.

II. DESCRIPTION OF THE REFERENCE REACTOR AND THE NEPTUNIUM-BEARING ROD

A typical PWR of current design was chosen as a basis for the study. Design parameters from a published¹ Final Safety Analysis Report were used. It should be made clear that independent reactor physics calculations were performed without regard for disagreement with the reactor vendor's physics calculations. The reference reactor was chosen simply because it is typical of PWR's being designed and built in the United States at the present time. Further, since the actual burnup of any commercial reactor core is generally confidential information, a burnup period of 80% of a full-power year was arbitrarily chosen to represent the first-core burnup. Fission-product cross sections were arbitrarily adjusted to yield this burnup for the reference reactor.

The core loading arrangement for the reference reactor is given in Fig. 1. In the calculational model, the core was represented as four concentric cylinders having areas equal to the actual area of each zone shown in the figure. The numbers shown on the figure indicate the number of burnable poison rods in a particular fuel assembly. The reactor was assumed to operate for 0.8 years at a constant power level of 2.758×10^9 watts.

A sketch of the neptunium rod that replaces burnable poison in the reference reactor is given in Fig. 2. The sintered pellet is a uniform

mixture of neptunium oxide and, to reduce self-shielding, some diluent such as aluminum oxide. The geometry of the rod was chosen such that the cladding is of standard fuel-rod material and size. The rod will fit in the existing guide tube with ample space for coolant flow. Using the standard fuel rod design also leaves open the possibility of replacing some of the fuel rods with neptunium rods should the need develop to irradiate more neptunium than can be accommodated by the vacant guide tubes.

III. METHODS USED FOR CALCULATIONS

A. Slowing Down Model

The space-independent, lethargy-dependent, steady-state diffusion equation for epithermal neutrons (> 0.625 ev) of lethargy u in a homogeneous medium was integrated over the epithermal lethargy range.

Thirty-seven groups, most being one-third of the lethargy unit wide, were used for the integration. The slowing down density was assumed to have three components: one from hydrogen scattering, one from inelastic scattering, and one from elastic scattering by heavy isotopes. Details of the integration of the slowing down equation are given in Appendix A.

B. Resonance Capture in U-238

For every resonance in U-238 where measured resonance parameters are available, the Bright-Wigner formula was integrated to obtain the infinite dilute resonance integral for that peak. The infinite dilute resonance integral was then multiplied by a factor which is a function of scattering per atom and temperature to obtain the homogeneous value of the resonance integral. An equivalence relation first suggested by Levine was used to compute an effective scattering per atom which takes account of lump-size and Dancoff effects. Fission cross sections and (n, γ) cross sections at high energies for U-238 were integrated numerically. The $1/v$

portion of the resonance integral was calculated from the 2200 m/sec cross section, and the unresolved portion of the resonance integral was estimated from the average reduced neutron width and the average gamma width for resolved resonances. Details of the resonance integral methods are given in Appendix B.

C. Thermal Spectrum

The standard TEMPEST program was used to calculate the thermal spectrum in the homogeneous medium. Details of this calculational method are presented in Ref. 2.

D. Spatial Distribution of the Thermal Flux

A formula that compares in simplicity to that of elementary diffusion theory was used to calculate the thermal flux distribution within a unit cell. Although the formula is simple, it gives results comparable to S_n and spherical harmonics methods. The formula is based on the work of Amouyal and gives the one-group, one-dimensional ratio of the flux in the moderator to that in the fuel for an infinite cell. A description of the method can be found in Ref. 3.

E. Self-Shielding Factors

Calculation of the effective self-shielding factors for the isotopes of interest in this study (i. e., Pu-240, Pu-238, and Np-237) was accomplished

using a modified version of the CNROD^{*} program. Once the effective self-shielding factors were obtained for various arbitrary concentrations, they were fitted to fourth degree polynomials in the concentration. The coefficients of the polynomial fits were used as input data in the burnup program. Consistent with the slowing down program's energy-group structure, self-shielding factors were computed separately for the thermal neutron energy group (0-0.625 ev) and for the epithermal energy group (i.e., those neutrons of energy greater than 0.625 ev). In the thermal group, self-shielding factors exhibit principally a spatial dependence due to the group flux depression effects in the vicinity of the absorber, while in the epithermal group, the self-shielding factors are more sensitive to the neutron energy because of the very marked resonance structure of the absorption cross sections. Appendix C presents details of the analytical technique, an assessment of cross section information, and a comparison with the available experimental data on self-shielding factors.

F. One-Dimensional Diffusion Model

Methods used for the gross radial flux and power distributions and for reactivity calculations were taken from the FEVER program. Details can be found in Ref. 8.

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* CNROD is a computer program written in Fortran IV and originally developed by Southern Nuclear Engineering to estimate the effective absorption of strongly self-shielded lumped absorbers (i.e., control rods) in slab, annular, and cylindrical geometry.

G. Burnup Equations

Single cross sections for each of the isotopes in each radial region considered in the calculations were averaged over the calculated neutron spectrum from 0 to 10 Mev. These cross sections were then used in analytical solutions to the differential equations describing the rate of change of concentrations of materials with burnup. The differential equations for the isotopes of interest in the Pu-238 chain, and their solutions, are listed in Appendix D.

IV. CROSS-SECTION EVALUATION

Cross sections used in the calculations are summarized in Table I, and each of the isotopes is discussed in a following paragraph.

Table I CROSS-SECTION VALUES USED
IN CALCULATIONS (barns)

<u>Material</u>	<u>Effective 2200 m/sec value for 1/v dependence</u>	<u>Infinite dilute resonance integral</u>
Pu-240	(calc. in TEMPEST)*	8041
Np-237 (absorption)	(calc. in TEMPEST)*	943
Np-238	1500	603.5
Pu-238	469	162
Pu-236	354	740

Pu-240. Thermal cross sections for this isotope were calculated by the TEMPEST program. Self-shielding in the thermal group is negligible; it occurs only in the epithermal group and is largely attributable to the very strong resonance peak at 1.056 ev. Calculation of the infinite-dilute resonance integral from reported resonance parameters of 9 resolved resonances⁵ (and including 100 barns estimated for the contribution of the unresolved resonances) yields a value of 8041 barns, not including a 1/v contribution

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*See TEMPEST library for values used.

of approximately 116 barns. The calculated resonance integral of 8041 barns is in reasonable agreement with reported experimental values,⁵ which range from 7900 barns to an upper limit of 15,000 barns, with most of the measurements in the region of 8000-9000 barns. Nichols⁶ has reported values of self-shielding for Pu-240 at several atom concentrations, normalized to a calculated infinite-dilute resonance integral of 8493 barns (non-1/v). Comparison of the calculated self-shielding factors with the measured values of Nichols will be discussed in a subsequent section of this report.

Np-237. As in the case of Pu-240, effective thermal group cross sections were provided by the TEMPEST program utilizing the neutron spectrum calculated by TEMPEST. The assumption is made that the thermal group spectrum "seen" by the neptunium target material is determined by the surrounding fuel region. Resonance parameter values reported^{5,7} in the literature yield a calculated infinite-dilute resonance integral (64 resolved resonances) of 621 barns, including a 134-barn contribution from the unresolved resonances and a 73-barn contribution from the 1/v cross section. This value of 621 barns is considerably less than the experimental value of 943 ± 130 barns,⁵ thus indicating the need for additional investigation of Np-237 cross-section values.* For analytical

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*Two later measured values, 905 barns¹⁰ and 807 barns¹¹ have been reported (both by Idaho Nuclear Corp.), but the reports are in abstract form and cannot be readily evaluated or compared.

purposes in this study, the experimental value of 943 barns was used for the infinite-dilute resonance integral. However, calculations of the self-shielding factors were based on the reported resonance parameters, and the resulting self-shielding factor applied to the measured infinite-dilute resonance integral to obtain effective values of the shielded resonance integral. Brookhaven National Laboratory was kind enough to do an independent analysis of the n, 2n cross section of Np-237. This resulted in the values given in Table II.⁸

Table II. THE n, 2n CROSS SECTION OF Np-237

<u>Energy, Mev</u>	<u>n, 2n cross section, millibarns</u>
10.0	292
9.7	282
9.3	264
8.59	213
8.28	180
7.99	145
7.71	107
7.46	69.21
7.22	35.35
7.07	17.22
6.93	4.714
6.789	0.0

It was interesting to note that the fission spectrum average of the cross sections given in Table II agrees quite well with the value of 1.3 millibarns reported in Ref. 9.

Np-238. A 2200 m/sec value⁵ of 1500 barns was assumed in order to give the thermal absorption cross section. The resonance integral was estimated to be 603 barns. Since Np-238 is never present in large quantities, the calculated neutron spectrum was assumed to be independent of Np-238.

Pu-238. Early values measured for the Pu-238 thermal capture cross section ranged from 454 to 588 barns. References 12 and 13 give the most recent and most complete assessments of the Pu-238 cross sections from 0.008 to 6500 ev, including the derived resonance parameters for 14 resolved resonances up to 192 ev and two proposed negative resonance levels. The shape of the absorption cross-section curve in the thermal energy range indicates a marked deviation from $1/v$ behavior. However, Young^{12, 13} provided results of an analytical evaluation of the "effective" $1/v$ cross section in a Maxwellian spectrum at several temperatures. The effective $1/v$ cross section at 300°C, i. e., 469 barns, was used in the present study. This value multiplied by the unit absorption factor from the TEMPEST program yields the effective average thermal group cross section for Pu-238.

Individual resonance parameters (14 resolved resonances up to 192 ev) and the estimated contribution from higher-energy unresolved resonances result in a calculated infinite-dilute resonance integral of 162 barns (including a $1/v$ contribution of about 2.8 barns). Earlier Canadian measurements gave an experimental value of 3260 ± 280 barns for the infinite-dilute resonance integral, which is in serious disagreement with the calculated value. However, the measured value was derived from activation experiments in a heavy-water reactor (low epithermal flux relative to the thermal neutron flux), correcting for the thermal absorption on the basis of a 2200 m/sec cross section of 489 barns. Young's measurement of 559 barns at 2200 m/sec would be consistent with a resonance integral of about 160 barns for the Canadian experiments. A later report¹⁴ indicated a measured value for the infinite-dilute resonance integral of 160 barns; this agrees well with the calculated value. For the purposes of the calculations reported here, the resonance parameters reported by Young were adopted, yielding an infinite-dilute resonance integral of 162 barns.

Pu-236. The only cross section given in the literature for this isotope is a 2200 m/sec fission cross section of about 170 barns. The cross section group at Brookhaven National Laboratory made estimates⁸ of the 2200 m/sec capture cross section and the total absorption resonance integral. These

values are given in Table I. Although the uncertainty in the values might be as great as $\pm 50\%$, they are the best estimates obtainable at the present time.

Comparison with Experimental Data

The validity of the method used for calculating self-shielding factors for absorber rods, whose absorption is predominately in the thermal group (e.g., boron), is shown in Ref. 15. Comparison of calculated values with those observed in a series of critical experiments, with boron rods in cylindrical and annular geometry, showed excellent agreement (as measured by the effect on reactivity). Although experimental data on self-shielding factors for neptunium rods are not available, the ability to predict accurately the effect in boron-loaded rods gives assurance that the same technique would be applicable to neptunium rods, at least in the thermal neutron group.

In the epithermal group, since neptunium does not show the same $1/v$ dependence as boron, a different method was necessary to account for the resonance structure of neptunium absorption. Unfortunately, the only experimental data available on self-shielding in concentrations of interest for materials with marked resonance absorption is that of Nichols for Pu-240. For rods of the geometry described by Nichols,⁶ the CNROD program was used to calculate self-shielding factors at the various

experimental Pu-240 concentrations. A comparison of the experimental and calculated values is shown in Fig. 3, which indicates that agreement well within the experimental uncertainty was observed throughout the concentration range. In addition, the self-shielding for U-238 in a typical fuel element was calculated by the same analytical technique despite the fact that the U-238 loading (and hence the self-shielding factor) is considerably beyond the range normally considered for neptunium rods. In the case of U-238, the calculated self-shielding factor (0.08) agrees reasonably well with the value (0.0805) obtained from the standard Hellstrand experimental correlation for a single isolated rod.

V. EXPERIMENTAL BASIS FOR MODEL

A. Reactivity

The scope of the contract did not permit an extensive analysis of critical experiments to verify that the model gave exactly the right initial reactivity. However, one experiment was analyzed which contained fuel similar to that in the base reactor and which also contained a very heavy concentration of soluble poison in the moderator. Data on the experiment analyzed was taken from Ref. 16. A photograph of the experiment, also taken from Ref. 16 is presented as Fig. 4. The fuel material in the critical experiment was UO_2 enriched to approximately 4% and swaged in stainless steel tubes. The experiment contained 4904 of these tubes. The tubes had an active fuel height of 66.7 in., a fuel density of 9.46 grams per cubic centimeter, an outside diameter of 0.4755 in., and a stainless steel wall thickness of 0.0160 in. Moderator for the experiment was light water at room temperature. The moderator contained 3390 ppm of boron. The total metal-to-water ratio for the lattice was approximately 1.0.

The one-dimensional methods described in Section III of this report give a value of k_{eff} for this lattice of 1.016. Since the experiment was a large one, and since the boron concentration in the moderator is several times that expected for a PWR of current design, it was felt that the

comparison of this experiment with calculations was a good test of the model for computing initial reactivity.

B. Isotopic Concentrations in the Pu-238 Chain

Experimental measurements of Np-237 content and Pu-238 content of neptunium targets as a function of burnup in neutron spectrums typical of those found in pressurized water reactors are not available. However, extensive measurements¹⁷ have been made on spent fuel from the Yankee Rowe reactor. The neptunium concentration in reactor fuel is an increasing function of time, whereas neptunium concentration in a separately-encapsulated target is a decreasing function of time. Thus, it was felt that the ability of the model to predict the U-236, Np-237, and Pu-238 concentration as a function of time in the fuel itself would be a good test. These calculations have been performed with the methods used in this study. Results are presented in Figs. 5, 6, and 7. One can see from the figures that the U-236, Np-237, and Pu-238 contents are predicted extremely well by the model. The ability of the model to predict the Pu-236 to Pu-238 mass ratio as a function of burnup in the spent fuel from Yankee is quite another story. As can be seen in Fig. 8, the measured points fall considerably below the calculated values. Although the reason for this discrepancy is not known at the present time, the model predicts too high a value for the Pu-236 to Pu-238 mass ratio in the fuel material and one would expect that calculations

for the neptunium target irradiations would yield values of this ratio considerably higher than the actual values. A program to irradiate neptunium targets in a commercial PWR has been proposed to the USAEC. Results of such a program will verify the actual Pu-236 to Pu-238 ratio.

VI. PARAMETRIC STUDY

A. Reference Reactor

Burnup calculations were performed for the reference reactor depicted in Fig. 1. Reactivity behavior with burnup is described in terms of the required moderator boric acid concentration required for criticality, as a function of time. Figure 9 gives the boron concentration as a function of time for the reference reactor, as well as the results of other one-dimensional calculations discussed later. One can see from the illustration that the boron concentration for the reference reactor goes to zero at approximately 80% of a full power year (~ 290 days).

B. Description of One-Dimensional Problems

When reading the following descriptions of one-dimensional problems, the reader should use Fig. 1 as a guide to regions.

- Problem 1   -   Reference reactor
- Problem 2   -   192 burnable poison rods replaced with 192 Np-237 rods containing 535 grams Np-237 per rod in region 2.
- Problem 3   -   192 boron rods replaced with 192 Np-237 rods containing 535 grams Np-237 per rod in region 3.
- Problem 4   -   192 burnable poison rods replaced with 192 Np-237 rods containing 535 grams Np-237 per rod over

the entire reactor with the ratio of the number of Np-237 rods to the total number of poison rods held constant for each region.

Problem 5 - 192 burnable poison rods replaced with 192 Np-237 rods containing 535 grams Np-237 per rod in region 4.

Problem 6 - This problem was used to test the computer programs and no results are reported here.

Problem 7 - All burnable poison rods in the reactor design replaced with Np-237 rods at 535 grams Np-237 per rod.

Problem 8 - 192 burnable poison rods replaced with 384 Np-237 rods at 267.5 grams Np-237 per rod. The Np-237 rods are spread over the entire core such that the Np-237 content in each region is identical with that of problem 4.

Problem 9 - 192 burnable poison rods replaced with 768 Np-237 rods at 133.75 grams Np-237 per rod. The Np-237 rods are spread over the entire core such that the Np-237 content in each region of the reactor is identical with that of problem 4.

Problem 10 - 100 burnable poison rods replaced with 100 Np-237 rods at 535 grams Np-237 per rod over the entire core with the ratio of the number of Np-237 rods to the total number of poison rods held constant for each region.

Problem 11 - 300 burnable poison rods replaced with 300 Np-237 rods at 535 grams Np-237 per rod over the entire core with the ratio of the number of Np-237 rods to the total number of poison rods held constant for each region.

C. Discussion of One-Dimensional Problems

Boron concentrations as a function of time for the problems described in Section B are seen in Fig. 9. The figure indicates that the number of full power days lost as a result of irradiating neptunium can range from 8 days to 78 days, depending on the quantity of Np-237 irradiated, its position in the reactor, and the Np-237 loading per target rod. The problems can be grouped to show the following effects on Np-237 consumption and Pu-238 production; the position of the neptunium in the reactor, the number of burnable poison rods replaced, and the neptunium loading per rod. Problems 2, 3, 4, and 5 show the effect of irradiating neptunium at various positions in the reactor. All of these problems have 192 Np-237 rods,

each containing 535 grams of Np-237 per rod in different positions in the reactor. Problems 4, 7, 10, and 11 show the effect of the number of burnable poison rods replaced. In these problems, the number of rods varied but the loading of neptunium per rod and the spatial distribution of the neptunium were constant. Problems 4, 8, and 9 show the effect of the neptunium loading per rod. These problems all had the same initial amount and spatial distribution of neptunium but the number of rods containing the neptunium was varied. Figure 10 shows the net Np-237 and Pu-238 as a function of time for problems 2, 3, 4, and 5. Figure 11 gives the net Np-237 and Pu-238 contents as a function of time for problems 4, 7, 10, and 11. Figure 12 shows the Np-237 and Pu-238 contents as a function of time for problems 4, 8, and 9. The radial power distributions as a function of time for problems 1, 2, 3, 4, 5, 7, 8, 9, 10, and 11 are given in Figs. 13 through 22, respectively. Note that the small numbers on the power distributions indicate full power days.

The effect of irradiating neptunium at various positions in the reactor can be seen in Fig. 10. This figure shows the net Np-237 and Pu-238 as a function of time for the case in which 192 burnable poison rods at different locations in the reference reactor were replaced with the neptunium rods. The neptunium rods contained 535 grams of neptunium per rod. The curves labeled "Problems 2, 3, and 5" correspond to the 192 burnable poison

rods being replaced with neptunium in radial regions 2, 3, and 4, respectively. The curves labeled "Problem 4" are for a situation where the 192 burnable poison rods replaced are distributed over the entire reactor core in a homogeneous fashion such that the ratio of the number of neptunium rods to the total number of poison rods is held constant for each region. In all of these problems, except Problem 4, the power distributions as a function of time are somewhat distorted when compared with the power distribution as a function of time for the reference reactor. This effect is most pronounced in Problem 2, where the 192 neptunium rods are all inserted in radial region 2. The power distribution for Problem 2 can be seen in Fig. 14. When the power distribution as a function of time for Problem 4 (the neptunium rods spread over the entire core), as shown in Fig. 16, is compared with the power distribution for the reference reactor, as shown in Fig. 13, one can see that little perturbation to the power distribution is observed. The Pu-238 yield for the homogeneous problem (Problem 4) was almost as great as that of the problem in this series that gave the maximum yield of Pu-238. The power distribution as a function of time was not perturbed for the homogeneous problem, as it was in the other problems where the rods were irradiated in discrete regions in the reactor. Therefore, an acceptable way to do the irradiations space-wise within the reactor is in a homogeneous fashion

similar to that represented by Problem 4. It should be emphasized that, with the exception of Problems 2, 3, and 5, the rest of the problems discussed in this report are based on homogeneous distribution of neptunium within the reactor core.

Figure 11 shows the net Pu-238 and Np-237 as a function of time for situations where the neptunium loading per rod was held constant but the actual number of burnable poison rods replaced with neptunium rods varied from 100 rods replaced to all 1160 burnable rods replaced. In order to get all of the lines on the graph, it was necessary to multiply some of the values by 10^{-1} . The figure shows that up to 70-odd kilograms of Pu-238 can be made, depending on the number of burnable poison rods replaced. Figure 9, giving the time in full power days where the boron concentration in the moderator goes to zero, shows that the number of full power days lost as a result of irradiating the neptunium can range from a few days, for one hundred rods replaced, up to approximately 78 days for all of the burnable poison rods replaced. Some perturbations in the power distribution, as compared to that of the reference reactor, are observed as the amount of neptunium approaches 600 kilograms. This effect is, of course, most pronounced in the problem where all of the burnable poison rods are replaced with neptunium. The power distribution for this problem is given in Fig. 18. Note that the power peak at the core-reflector interface is pushed up to unacceptable limits toward

the end of lifetime. The series of calculations indicates that the optimum number of burnable poison rods to replace falls between 100 and 200.

Figure 12 shows the net Pu-238 and Np-237 as a function of time for problems having the same initial amount of neptunium, but having various amounts of neptunium per rod; that is, one burnable poison rod might be replaced by one rod containing Np-237 or by several rods containing lower loadings of Np-237. The figure indicates that the production rate of Pu-238 is significantly increased as the neptunium loading per rod is decreased. Power distributions as a function of time for the series of problems listed on Fig. 12 showed little perturbation when compared with the power distribution as a function of time for the reference reactor. This can be seen by studying Figs. 15, 19, and 20, which give the power distributions as functions of time for this series of problems.

Several cell-type calculations were performed to study the effect of the neptunium loading per rod on the production rate of Pu-238 and on the ratio of Pu-236 to Pu-238 as a function of time. In effect, the cell calculations are based on the central region of the reference reactor, with assumed boundary conditions that give a flat flux distribution over the region. Results of this series of calculations are shown in Fig. 23. Note that the production rate of Pu-238 is maximized and the Pu-236 to Pu-238 mass ratio is minimized as the neptunium loading per rod is decreased. It should be noted that the same initial amount of Np-237 was used in all

of the cell calculations. Figure 24 shows the results of this same series of calculations but at a specific fuel burnup of 9500 Mwd/mtU, roughly an average figure for the burnup in a PWR core region after the first cycle.

D. Practical Problem

Results of the parametric study were analyzed extensively in order to specify a practical situation that could be applied to the reference reactor for neptunium irradiation. First, the total number of spaces available for neptunium target rods, after 192 burnable poison rods were removed, was determined. Then, neptunium target rods were distributed among the available spaces in such a way that the ratio of the number of neptunium rods to the total number of poison rods could be nearly constant for each region in the reactor. The practical situation came out as follows:

	<u>Number of burnable poison rods removed</u>	<u>Number of neptunium rods inserted</u>
Region 1	11	27
Region 2	42	109
Region 3	82	213
Region 4	57	148

As the above listing shows, a total of 192 burnable poison rods was removed; these rods were replaced with 497 neptunium rods. The neptunium concentration per rod was then adjusted such that the initial reactivity of the reference reactor and that of the practical problem were the same.

The resulting mass of Np-237 per rod was 147.8 grams. A complete burnup analysis of the configuration was then performed. Figure 25 shows the net Np-237 and Pu-238 masses as functions of time for the practical problem. Note that approximately $73\frac{1}{2}$ kilograms of neptunium are required for the initial target rods and that the neptunium's mass is reduced to approximately 56 kilograms during the irradiation. At the same time, approximately 15 kilograms of Pu-238 are produced. The Pu-236 impurity in the Pu-238 for the practical problem was calculated to be slightly greater than 4 ppm. However, the previous comparison between calculated and experimental results indicates that the actual Pu-236 impurity in the target rods will be considerably less than that calculated, probably of the order of 2 ppm. The actual amount of Pu-236 impurity will not be known until an irradiation test is performed on this type target rod in a commercial pressurized water reactor. The gross radial power distribution as a function of time for the practical problem is seen in Fig. 26. Although slightly distorted when compared with the reference problem, the gross radial power distribution would seem to be acceptable since the power peak does not exceed that of the reference problem and fairly uniform irradiation is obtained in all four regions of the reactor.

E. Neutron Economics

It is important to show how many neutrons normally captured in burnable poison can be used to irradiate Np-237 by the process reported

here. To do this, a comparison was made between the neutron absorptions in burnable poison in a region of the reference reactor and the combined absorptions in burnable poison, Np-237, and Pu-238 in that same region of the reactor where some of the burnable poison rods were replaced with neptunium rods. Figure 27 gives the absorption rate in burnable B-10 in region 4 of the reference reactor as well as absorption rates in Np-237, B-10, and Pu-238 in region 4 of Problem 5. In Problem 5, 192 of the burnable poison rods in region 4 were replaced with neptunium rods containing 535 grams of neptunium per rod. Numerical integrations of the lines on the graph from 0 to 240 days (both systems were critical at 240 days) give numbers which are proportional to the number of neutrons absorbed in the various materials for the time period indicated. One can calculate from these integrations the percentage of neutrons absorbed in Np-237 that would normally be captured in B-10 in the reference reactor. This number is calculated to be 68%. In other words, 68% of the neutrons captured in the neptunium rods would normally have been captured in the reference reactor by boron anyway. Some of the reactor's first core life-time is lost by irradiating Np-237; Fig. 9 indicates that this loss is about one full power day for each kilogram of Pu-238 produced. For the practical problem, the figure is about 1.1 kilograms of Pu-238 produced per full power day lost. The energy loss is due to extra absorption by Np-237 (the other 32%).

VII. ECONOMICS

The great advantage of the irradiation technique discussed in this report is that it employs facilities which are used primarily for other purposes, and Pu-238 is produced strictly as a by-product. The neptunium target rods replace burnable poison rods and the reactor has similar, though not identical, reactor physics characteristics. While there is an impact on the reactor's fuel cycle and operation, it is minimal. Neutron economy in the reactor is affected but little. A majority of the neutrons captured in the neptunium target rod would be captured in burnable poison anyway. Therefore, the commercial power reactors under construction or planned represent a potential for economically producing large quantities of Pu-238. The estimated cost of Pu-238 production by this method is of considerable interest even though there are a number of uncertainties in the cost figures. A preliminary cost analysis was performed as described below. Because of the magnitude of the uncertainties, both a "high" and a "low" cost estimate were made to show a range rather than a single cost figure.

A. Cost of Fabrication for the Target Rods

Discussions were held with two commercial fuel fabricators. At this preliminary stage, it was impossible to define fully the requirements and specifications for the target rods, but it was possible to make certain

clarifying assumptions. The major technical area of doubt from the fabricators' standpoint concerns the lack of background information on manufacturing neptunium-containing pellets. A search of the literature revealed that there is little or no useful information on the compatibility of neptunium oxide with aluminum oxide^{*} and the behavior of such mixtures in pellet fabrication. The fabricators seemed to consider that these oxides should be compatible and no unusual problems would be expected. However, a research and development program would be required to investigate the compatibility of the mixed oxides and to develop process parameters and methods for commercial pellet production. One fabricator estimated that such an R&D program might cost \$85,000, with this price including the fabrication of six prototype rods.

To obtain a cost estimate, the following assumptions were made:

1. A short research and development program has been carried out. The program demonstrated the compatibility of neptunium oxide with aluminum oxide, and no difficult fabrication problems were encountered.
2. The rods to be produced are of a design suitable for use in the latest generation of large power reactors — i.e., the Indian Point 2 type.

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^{*}Aluminum oxide is apparently slightly preferable to zirconium and magnesium oxides, but these oxides would be satisfactory if difficulties should develop with Al_2O_3 .

3. The necessary quality control is at the level required for present commercial fuel elements rather than for Navy fuel elements or plutonium recycle elements. It was also assumed that the neptunium oxide will not tend to segregate in the pellets to an unacceptable level.
4. The neptunium as received has attained protactinium equilibrium so that shielding would be provided for this maximum level of radiation.
5. The cost for accountability, licensing, and administration would be no greater than similar present costs for uranium.
6. At least 192 target rods will be produced each year.

It was considered that fabrication could best be carried out in an existing plutonium fabrication line and that it would not be necessary or economically advisable to build a special facility for the limited production envisioned. If production were increased noticeably above the 192 rods per year level, unit costs would be expected to go down.

On the basis of these assumptions, and on previous experience with conventional fuel and with plutonium recycle elements, one fabricator considered fabrication costs to be bracketed between a low cost of \$330 per rod and a high cost of \$600 per rod. It should be emphasized, of course, that these figures are budgetary or "ball park" estimates but do represent

informed opinions. It is believed that the fabrication time required would be from one month to a maximum of two months after the receipt of the neptunium.

B. Reprocessing Costs

The techniques for recovering plutonium and neptunium from the irradiated target rods are fully developed and present no technical problems. Either the solvent-extraction or ion-exchange technique could be employed with little difference in total cost. Discussions with a commercial fuel reprocessor centered around the problem of the quantities to be processed and the size of the unit required. The major difficulty is that the cost of a plant of this nature is not sensitive to size — a large plant is not very much more expensive than a small one. The minimum-size recovery plant considered would be able to process 500 kilograms of Pu-238 per year. This amount of plutonium, while large, is not out of line with some of the predicted requirements and, by the late 1970's, the amount of Np-237 available would be adequate to produce it if the recovery of Np-237 is started in the near future and if this neptunium is kept in the plutonium production cycle. For estimating purposes, the recovery unit was considered to be a separate plant with its own staff and laboratories. Plant operations would be done remotely, but the shielding necessary would be relatively light. The plant was considered to have a 15-year life. The total staff

for independent operation would be relatively large. Including all materials, supplies, capital charges, and operating costs, the plant was estimated to operate at \$1,500,000 per year. If 500 kilograms of plutonium were produced, the recovery cost would be \$3 per gram of Pu-238. However, all the costs would remain quite nearly constant, so that, at a 50 kilogram yearly production rate, the cost would approach \$30 per gram. If it were necessary to operate the plant at below its nominal capacity, savings might be realized if the unit were operated as part of a larger reprocessing plant system by running the plutonium recovery in "campaigns" and borrowing the operating crews from other units when circumstances permitted. This type of operation would only be possible with a combination of fortunate circumstances. The production cost for a 50 kilogram per year production rate would be somewhat less than the \$30 given above under these circumstances. Capital charges would continue, and it would be necessary to provide certain maintenance, monitoring, and other services regardless of the production level at which the plant was operated.

The cost of Pu-238 recovery would certainly warrant a far more complete study when plutonium requirements are more clearly established. It does appear from this preliminary study that, when sufficient capacities are involved, recovery operations can be carried out reasonably economically.

C. Irradiation Costs in Commercial Power Reactors

The primary cost of irradiation of neptunium in a commercial power reactor arises from the small difference in reactivity behavior of the core, as compared to the core without neptunium. The neptunium requires extra neutrons, and the first-cycle lifetime is therefore shortened by about two weeks for the practical case. In our low cost estimate, we have assumed that only the first batch (one-third of the core, which is removed at the first refueling as are the neptunium target rods) is affected. This assumption is justifiable because there is flexibility inherent in the successive fuel cycles which allows for minor perturbations, allowing target burnups to be reached by all fuel except the first batch.

For a reactor such as the one under consideration, about 45% of the total fuel costs incurred in the first cycle are attributable to the first batch removed. The total fuel cost is about 2 mills/kw-hr, or, for a 1000 Mw(e) plant, \$2000/hr. The first cycle is shortened by about 15 days, or 360 hours, and the energy lost from the first batch of fuel is valued at about \$320,000. However, there is a compensation. The enrichment of the first-batch fuel will be higher than if it had achieved the full burnup, and this will offset about one-half the cost, leaving a net cost of \$160,000. Adding \$40,000 to cover additional expenses in fuel cycle analysis, licensing, extra handling, and contingencies, the total irradiation cost was estimated to be \$200,000.

For the high cost estimate, no credit was taken for the increased value of the spent first-batch fuel. Recognizing the possibility of some delayed impact on the fuel cycle (i.e., costs occurring after the irradiation is completed) and the possibility of extra reactor down time, an additional \$140,000 was assumed for the high cost estimate. The licensing, etc. was still estimated to be \$40,000. The total irradiation cost is then \$500,000 for the high estimate.

D. Neptunium Cost Estimates

The cost of neptunium is an extremely important factor. The cost has been estimated to be between \$40 and \$75 per gram. Although this is a broad range, its use is consistent with the numbers obtained for fabrication and reprocessing. As discussed above, it was not feasible to attempt a rigorous cost analysis.

E. Capital Charges

For capital charges (or cost of money), a rate of 15% per year was used; this percentage corresponds to the cost of money to normal, investor-owned utilities. The rate would be applicable to a typical utility and covers all the normal costs of doing business, including federal, state, and local taxes and return on investment. We have assumed utility ownership of the target rods, including the contained neptunium.

The total costs as we have estimated them are shown in Table III for both "high" and "low" costs. Omitted from these estimates are credits for burnable poison rods which were replaced by the neptunium target rods. A small cost reduction would result if this credit were taken.

The estimate is based on the practical problem discussed in Section VI of this report. In the practical problem, 192 burnable poison rods were removed and were replaced with 497 rods containing a total of 73,463 grams of neptunium; 14,849 grams of Pu-238 were produced. Following the irradiation, 55,993 grams of neptunium exist in the target rods. An 18-month period was assumed for the entire cycle, including a two-month fabrication time before irradiation, a 12-month cycle in the reactor, and a four-month handling and reprocessing period afterward. Nonrecoverable losses of 2% in both fabrication and reprocessing were assumed.

As shown in Table III, Pu-238 produced by the method of replacing burnable poison with neptunium will cost between \$134 and \$282 per gram.

Table III COST ESTIMATES FOR Pu-238 PRODUCTION
IN COMMERCIAL REACTORS

		<u>Low Cost Estimate</u>		<u>High Cost Estimate</u>	
			Value, thousands of \$		Value, thousands of \$
	<u>Unit</u>			<u>Unit</u>	
1.	Neptunium in 73,463 g x 1.02 = 74,932 g	\$40/g	2,997	\$75/g	5,620
2.	Fabricate 497 rods	\$330/rod	164	\$600/rod	298
3.	Capital charges on (items) 1 & 2 @ 15%/yr for 18 months		711		1,332
4.	Handling and shipping		25		25
5.	Additional fuel cycle costs		200		500
6.	Reprocessing & recovery 14.850 kg Pu-238	\$3/g Pu-238	45	\$30/g Pu-238	446
7.	Neptunium recovered 55.99 x 0.98 = 54.87 kg	\$40/g	(2,195)	\$75/g	(4,115)
TOTAL COST			1,947		4,106
Plutonium recovered 14,849 g x 0.98 = 14,552 g			\$134/g		\$282/g

VIII. TEN-YEAR PROJECTIONS

In light of the difference between currently quoted prices of Pu-238 and estimated production costs in commercial pressurized water reactors, the amount of Pu-238 that could be produced in these reactors is of interest. An estimate of Pu-238 production capability in commercial PWR's in the United States has been projected on a year-to-year basis over the decade starting in 1972. Figure 28 shows an estimated total production capability of about 1400 kilograms over this period. This amount of Pu-238 is consistent with the requirements indicated by Dr. Seaborg (see footnote on page 1). If commercial boiling water reactors were also used, and if their conversion characteristics are assumed to be the same as those of PWR's, the total production capability would increase to approximately 2300 kilograms.

In accordance with Section VI, D, where a practical substitution of neptunium rods in a typical PWR was considered, it was assumed that a PWR rated at 800 Mw(e) would produce 15 kilograms of Pu-238 during its first operating cycle. All commercial PWR's scheduled to begin commercial operation in 1972 or later (42 total) were included in the projection presented here. It is not expected that commercial-scale production could commence any sooner than 1972, since the earliest

possible experimental results will not be available before 1971. The latest information on presently-announced nuclear plants was used to obtain the figures through the year 1975. One year was added to the announced date of commercial operation to allow for the first core cycle. Production figures were adjusted for design power levels significantly above or below 800 Mw(e).

Nuclear plants not yet on order will be operating after 1975. Therefore, beginning with 1976, the estimates of first-year PWR capacity are based on forecasts of anticipated nuclear capacity. The low side of the 1967 AEC forecast was used as a basis for the extrapolation. A constant rate of increase, rather than the generally-expected accelerating growth rate, was used as an additional conservatism. Published estimates¹⁸ of growth in nuclear power production in the latter 70's have ranged up to 16,000 Mw(e)/yr and higher. The projection described is based on a growth rate of 12,000 Mw(e)/yr. The Arthur D. Little report¹⁹ on the nuclear power market shows cumulative PWR orders at approximately 60% of the total nuclear Mw(e); events since the publication of that report have shown that the percentage for PWR's has tended to be confirmed. This percentage of the 12,000 Mw(e)/yr growth rate, coupled with the 15 kg/800 Mw(e) assumption, yielded the 135 kg/yr production estimate for the 1978-81 period. Due to the recent slack period in nuclear power plant orders, production figures for the interim years of slower nuclear-capacity growth (i.e., 1976 and 1977) were adjusted downward.

Although not included in the estimates, it is expected that additional PWR-produced Pu-238 could be provided by irradiation in core cycles subsequent to the first. In fact, there is a definite possibility that burnable poison rods will be required in some PWR cores beyond the first cycle. In these cores, Pu-238 production by the proposed method could continue beyond the first year of operation, possibly over the life of the plant. In other cores not requiring burnable poison, there is still a possibility for Pu-238 production, with direct, but acceptable, economic penalty from reduced core lifetime or higher enrichment.

The total estimated potential for producing Pu-238 by the methods outlined in this study demonstrates that commercially-attractive quantities can be produced over the ten-year period starting in 1972. Additional production potential exists at higher costs, with virtually no limitation on meeting future requirements for Pu-238.

IX. CONCLUSIONS AND RECOMMENDATIONS

The study described here has shown that it is feasible to produce Pu-238 in significant quantities in commercial water reactors. The amount of Pu-238 produced in a first-core cycle can range up to 70 kilograms. Production of Pu-238 by the methods discussed in this report has the advantage of putting into effective use neutrons which otherwise would be captured in boron or other burnable poisons. The methods discussed would also permit large commercial power reactors now operating, or approaching operating status, to be used for Pu-238 production. Should a large demand for Pu-238 develop, these commercial reactors may prove to be the only economic irradiation facilities of adequate size available. The cost of Pu-238 produced in commercial water reactors has been estimated to be between \$134 and \$282 per gram.

In addition to uncertainties associated with the cost calculations, this study had two serious limitations: a lack of experimental data on neptunium targets irradiated in a neutron spectrum characteristic of light-water reactors, and the one-dimensional limitation of the model used for the study. Irradiation of neptunium targets in a water-reactor spectrum, and further calculations using two- and possibly three-dimensional models, are strongly recommended. The irradiation of neptunium targets will experimentally

confirm and demonstrate the feasibility of Pu-238 production by irradiation of Np-237 target material in light-water spectra. It will also establish the level of Pu-236 contamination and serve as a valid checkpoint for neutron cross sections and analytical methods. Multi-dimensional calculations are necessary to describe explicitly the neptunium target rod geometries in relation to the fuel within an assembly, to describe more accurately self-shielding (flux depression) effects, and to evaluate the effects of target rods on local power distributions as a function of burnup.

Although they will require additional assessments of the market potential for the isotope and verification of the production process and its economics (through irradiation tests and further analyses), the utilities are enthusiastic about the process and quite willing to produce Pu-238 as a by-product.

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APPENDIX A

NEUTRON SLOWING DOWN MODEL

The space independent, lethargy dependent, steady state diffusion equation for epithermal neutrons of lethargy u in a homogeneous medium is:

$$D(u)B^2\phi(u) + \sum^A(u)\phi(u) = S(u) - \frac{\partial q(u)}{\partial u} \quad A-1$$

The lethargy dependent variables are:

- D , the diffusion coefficient
- ϕ , the neutron flux per unit lethargy
- $\sum^A$ the absorption cross section
- S , the source spectrum
- q , the slowing down density

Equation A-1 is numerically integrated over the epithermal lethargy range. Thirty-seven groups, most groups being $1/3$ of a lethargy unit wide, are considered for the integration. Nuclear cross sections for each of the groups are averaged over a spectrum which is considered to vary as the reciprocal of the neutron energy within the group. The slowing down density is assumed to have three components: one from hydrogen, one from inelastic scattering events, and one from elastic scattering by heavy isotopes. These are denoted by $q_h(u)$, $q_I(u)$, and $q_{o.h.}(u)$, respectively.

Integrating Eq. A-1 from lethargy u_i to lethargy u_{i+1} , the group width being $\Delta u_i = u_{i+1} - u_i$, one gets:

$$D_i B^2 \phi_i + \sum_i^A \phi_i = S_i - q_{u_{i+1}} + q_{u_i} \quad A-2$$

where D_i and $\sum_i^A$ are average quantities for the group, i.e.,

$$D_i = \frac{\int_{u_i}^{u_{i+1}} D(u) \phi(u) du}{\int_{u_i}^{u_{i+1}} \phi(u) du}$$

$$\sum_i^A = \frac{\int_{u_i}^{u_{i+1}} \sum^A(u) \phi(u) du}{\int_{u_i}^{u_{i+1}} \phi(u) du} \quad A-4$$

The hydrogen component of the slowing down density is that found in Glasstone and Edlund,¹ which in terms of lethargy is:

$$q_h(u) = \int_0^u e^{u'-u} \sum_H^S(u') \phi(u') du' \quad A-5$$

where $\sum_H^S$ is the total scattering cross section for hydrogen.

¹S. Glasstone and M. C. Edlund, The Elements of Nuclear Reactor Theory, (Fourth Printing), D. Van Nostrand Co., Inc., Princeton, New Jersey, November 1955.

As a first approximation, the contribution to the slowing down density from inelastic scattering by all inelastic scatterers is based on the following: The distribution in lethargy of a neutron which has suffered an inelastic collision is uniform between the lethargy at which the collision occurred and a fixed cutoff lethargy U_m . This approximation yields the following for the inelastic component of the slowing down density.

$$q_I(u) = \int_0^u \Sigma^I(u') \phi(u') \frac{(U_m - u)}{(U_m - u')} du' \quad A-6$$

The component of the slowing down density from elastic scattering by heavy isotopes is obtained from Fermi age theory, i.e.,

$$q_{o.h.}(u) = \xi \Sigma^S(u) \phi(u) \quad A-7$$

where $\xi \Sigma^S$ is the slowing down power for the homogeneous medium not considering the hydrogen.

The slowing down density is the sum of Eqs. A-5, A-6, and A-7, i.e.,

$$q(u) = q_h(u) + q_I(u) + q_{o.h.}(u) \quad A-8$$

The group-wise integration of Eqs. A-5, A-6, and A-7 is performed as follows:

For hydrogen

$$q_h(u) = e^{-u} \int_0^u e^{u'} \Sigma_H^S(u') \phi(u') du' \quad A-9$$

with the group structure:

$$\frac{\Delta u_0}{\Delta u_1} = \frac{u_0}{u_1} = 0$$

and

$$\Sigma_{H_i}^s = \frac{\int_{u_i}^{u_{i+1}} \Sigma_H^s(u) \phi(u) du}{\int_{u_i}^{u_{i+1}} \phi(u) du} \quad A-10$$

$$\bar{\phi}(u)_i = \frac{\int_{u_i}^{u_{i+1}} \phi(u) du}{\int_{u_i}^{u_{i+1}} du} = \frac{\phi_i}{\Delta u_i} \quad A-11$$

one gets for the slowing down density at u_1

$$q_h(u_1) = e^{-u_1} \int_0^{u_1} e^{u'} \Sigma_H^s(u') \phi(u') du' = \Sigma_{H_0}^s \phi_0 \left[\frac{1 - e^{-\Delta u_0}}{\Delta u_0} \right] \quad A-12$$

and for the slowing down density at u_2

$$q_h(u_2) = e^{-u_2} \int_0^{u_2} e^{u'} \Sigma_H^s(u') \phi(u') du' \quad A-13$$

$$q_h(u_2) = e^{-u_2} \left[\int_0^{u_1} e^{u'} \Sigma_H^s(u') \phi(u') du' + \int_{u_1}^{u_2} e^{u'} \Sigma_H^s(u') \phi(u') du' \right] \quad A-14$$

using Eq. A-12

$$q_h(u_2) = e^{-u_2} \times e^{+u_1} q_h(u_1) + e^{-u_2} \int_{u_1}^{u_2} e^{u'} \sum_H^S(u') \phi(u') du' \quad A-15$$

$$q_h(u_2) = q_h(u_1) e^{-\Delta u_1} + \sum_{H_1}^S \phi_1 \left[\frac{1 - e^{-\Delta u_1}}{\Delta u_1} \right] \quad A-16$$

so that in general

$$q_h(u_{i+1}) = q_h(u_i) e^{-\Delta u_i} + \sum_{H_i}^S \phi_i \left[\frac{1 - e^{-\Delta u_i}}{\Delta u_i} \right] \quad A-17$$

For inelastic scattering

$$q_I(u) = \int_0^u \sum^I(u') \phi(u') \frac{u_m - u}{u_m - u'} du' \quad u_m > u \quad A-18$$

With the same group structure one gets for the slowing down density at u_1

$$q_I(u_1) = (u_m - u_1) \int_0^{u_1} \sum^I(u') \phi(u') \frac{1}{(u_m - u')} du' \quad A-19$$

$$\sum_i^I = \frac{\int_{u_i}^{u_{i+1}} \sum^I(u) \phi(u) du}{\int_{u_i}^{u_{i+1}} \phi(u) du} \quad A-20$$

$$q_I(u_1) = (u_m - u_1) \sum_0^I \frac{\phi_0}{\Delta u_0} \int_0^{u_1} \frac{du'}{(u_m - u')} \quad A-21$$

$$q_I(u_1) = \frac{(u_m - u_1)}{\Delta u_0} \log \left[\frac{u_m - u_0}{u_m - u_1} \right] \sum_0^I \phi_0 \quad A-22$$

and for the slowing down density at u_2

$$q_I(u_2) = (u_m - u_2) \int_0^{u_2} \sum^I(u') \phi(u') \frac{1}{u_m - u'} du' \quad A-23$$

$$q_I(u_2) = (u_m - u_2) \left[\int_0^{u_1} \sum^I(u') \phi(u') \frac{1}{(u_m - u')} du' \right. \\ \left. + \int_{u_1}^{u_2} \sum^I(u') \phi(u') \frac{1}{(u_m - u')} du' \right] \quad A-24$$

Using Eq. A-19, one gets

$$q_I(u_2) = (u_m - u_2) \left[\frac{1}{(u_m - u_1)} q_I(u_1) + \int_{u_1}^{u_2} \sum^I(u') \phi(u') \frac{1}{(u_m - u')} du' \right] \quad A-25$$

$$q_I(u_2) = \frac{(u_m - u_2)}{(u_m - u_1)} q_I(u_1) + \frac{(u_m - u_2)}{\Delta u_1} \sum_1^I \phi_1 \int_{u_1}^{u_2} \frac{du'}{(u_m - u')} \quad A-26$$

$$q_I(u_2) = \frac{(u_m - u_2)}{(u_m - u_1)} q_I(u_1) + \frac{(u_m - u_2)}{\Delta u_1} \log \frac{(u_m - u_2)}{(u_m - u_1)} \sum_1^I \phi_1 \quad A-27$$

so that in general

$$q_I(u_{i+1}) = \frac{(u_m - u_{i+1})}{(u_m - u_i)} q_I(u_i) + \frac{(u_m - u_{i+1})}{\Delta u_i} \log \frac{(u_m - u_i)}{(u_m - u_{i+1})} \sum_i^I \phi_i \quad A-28$$

Equation A-28 is approximated

$$q_I(u_{i+1}) = \frac{(u_m - u_{i+1})}{(u_m - u_i)} q_I(u_i) + \sum_i^I \phi_i \quad A-29$$

$$\text{Since } \log \frac{(u_m - u_i)}{(u_m - u_{i+1})} \simeq \frac{\Delta u_i}{(u_m - u_{i+1})} \quad A-30$$

Equation A-30 is exact for two terms of an exponential expansion.

Finally, for heavy isotopes:

$$q_{o.h.}(u) = \xi \sum^{So.h.} (u) \phi(u) \quad A-31$$

when $\xi \sum^{So.h.}$ is the slowing down power for isotopes other than hydrogen.

Equation A-31 is approximated by

$$q_{o.h.}(u_i) = \xi \sum_{i-1}^{So.h.} \phi(u_i) \quad A-32$$

$$q_{o.h.}(u_{i+1}) = \xi \sum_i^{So.h.} \phi(u_{i+1}) \quad A-33$$

where

$$\phi(u_{i+1}) = \frac{2\phi_i}{\Delta u_i} - \phi(u_i) \quad A-34$$

Rearranging Eq. A-2

$$D_i B^2 \phi_i + \sum_i^A \phi_i + q(u_{i+1}) = S_i + q(u_i) \quad A-35$$

$$\begin{aligned}
\text{or } D_i B^2 \phi_i + \sum_i^A \phi_i + q_h(u_i) e^{-\Delta u_i} + \sum_{h_i}^S \phi_i \left[\frac{1 - e^{-\Delta u_i}}{\Delta u_i} \right] \\
+ q_I(u_i) \frac{(u_m - u_{i+1})}{(u_m - u_i)} + \sum_i^I \phi_i + \xi \sum_i^{\text{So.h.}} \phi_i \frac{2}{\Delta u_i} - \xi \sum_i^{\text{So.h.}} \phi(u_i) = \\
S_i + q_h(u_i) + q_I(u_i) + \xi \sum_{i+1}^{\text{So.h.}} \phi(u_i)
\end{aligned} \tag{A-36}$$

Using Eqs. A-17, A-29, A-33, and A-34

$$\begin{aligned}
D_i B^2 \phi_i + \sum_i^A \phi_i + q_h(u_{i+1}) + q_I(u_{i+1}) + q_{o.h.}(u_{i+1}) = \\
S_i + q_h(u_i) + q_I(u_i) + q_{o.h.}(u_i)
\end{aligned} \tag{A-37}$$

Solving Eq. A-37 for ϕ_i

$$\phi_i = \frac{S_i + q_h(u_i) + q_I(u_i) + (\xi \sum_{i-1}^{\text{So.h.}} + \xi \sum_i^{\text{So.h.}}) \phi(u_i) - q_h(u_i) e^{-\Delta u_i} - q_I(u_i) \frac{(u_m - u_{i+1})}{(u_m - u_i)}}{D_i B^2 + \sum_i^A + \sum_{h_i}^S \left[\frac{1 - e^{-\Delta u_i}}{\Delta u_i} \right] + \sum_i^I + \frac{2}{\Delta u_i} \xi \sum_i^{\text{So.h.}}} \tag{A-38}$$

if $q_h(u_0) = q_I(u_0) = \phi(u_0) = q(u_0) = 0$, Eq. A-38 can be solved as a recursion relation making use of Eqs. A-17, A-29, A-33, and A-34.

The thermal flux may be obtained assuming the source at the lower lethargy of the thermal group equal to the sum of absorptions and leakage in that group, i.e.,

$$D_{th} B^2 \phi_{th} + \sum_{th}^A \phi_{th} = q(u_{37}) \tag{A-39}$$

or

$$\phi_{th} = \frac{q(u_{37})}{D_{th} B^2 + \Sigma_{th}^A} \quad A-40$$

where D_{th} and Σ_{th}^A are, respectively, the appropriate diffusion coefficient and absorption cross section for the thermal group.

The neutron spectrum ϕ may be used to obtain many of the reactor parameters for the medium under consideration. These include group coefficients for use in a few-group spatial calculation of the flux, the Fermi Age for the medium, various reaction rates, the infinite multiplication factor for the medium, and the effective multiplication factor for the one-region bare reactor whose leakage is characterized by the buckling, B^2 .

APPENDIX B

RESONANCE CAPTURE IN URANIUM-238

The Breit-Wigner¹ formula for the cross section for a reaction r ,
in the neighborhood of a resonance peak is:

$$\sigma_r(E) = \frac{\pi h^2}{P_i^2} g \left[\frac{\Gamma_n \Gamma_r}{\frac{\Gamma^2}{4} + (E - E_R)^2} \right] \quad \text{B-1}$$

where h

is Planck's Constant $(6.625 \times 10^{-27} \text{ erg.-sec.})$,

$$h = \frac{h}{2\pi} = 1.054 \times 10^{-27} \text{ erg.-sec.}$$

$P_i \equiv$ neutron momentum before collision

$$\sqrt{2mE} = 1.83 \times 10^{-12} \sqrt{E} \left(\frac{\text{gm.cm.}}{\text{sec.}} \right)$$

m is neutron mass and E its energy before collision

$g \equiv$ spin factor

$E_R \equiv$ energy at peak of resonance

$\Gamma_n \equiv$ neutron width

$\Gamma_r \equiv$ reaction width

$\Gamma \equiv \Gamma_n + \sum_r \Gamma_r$ (sum over all reactions)

The maximum cross section for the reaction r is obtained from Eq. B-1

where $E = E_R$

$$\sigma_r^{\text{max}} = \frac{\pi h^2 g}{2mE_R} \frac{\Gamma_n \Gamma_r}{\frac{1}{4} \Gamma^2} \quad \text{B-2}$$

The maximum cross section for all reactions is

$$\sigma^{\max} = \frac{2\pi \lambda^2 g \Gamma_n}{m E_R \Gamma} = \frac{2.6 \times 10^6 g \Gamma_n}{E_R \Gamma} \quad \text{B-3}$$

The energy dependent cross section is, in terms of $\sigma^{\max}$

$$\sigma^r(E) = \frac{\sigma^{\max} \Gamma \Gamma_r E_R}{[\Gamma^2 + 4(E - E_R)^2] E} \quad \text{B-4}$$

The infinite dilute resonance integral, for the reaction r , due to a single peak is

$$R_{\infty} = \int \sigma_r(E) \frac{dE}{E} \quad \text{B-5}$$

Assuming that the momentum of the neutron at resonance and the flux at resonance to be constant and performing this integration from $-\infty$ to $+\infty$, one gets

$$R_{\infty} = \frac{\pi \sigma^{\max} \Gamma_r}{2 E_R} \quad \text{B-6}$$

For a homogeneous mixture of a resonance absorber and a scattering material, the effective resonance integral for the absorber in the mixture is given by²

$$R_H = R_{\infty} f_H(\theta, \zeta) \quad \text{B-7}$$

where

$$\theta = \frac{4E_R K T}{A \Gamma^2} \quad \text{and} \quad \zeta = \frac{\sigma_m}{\sigma_p}$$

$$\sigma_P = \frac{\sum s}{N_A} \text{ (the scattering per atom of absorber in the mixture).}$$

The factor $f_H(\theta, \zeta)$ as given by Roe² is

$$f_H(\theta, \zeta) = \frac{\beta + \sqrt{1 + \zeta \alpha}}{\sqrt{1 + \zeta \alpha} \left[\zeta + (1 + \beta)^2 + \frac{2\beta}{\alpha} [\sqrt{1 + \zeta \alpha} - 1] \right]^{\frac{1}{2}}} \quad \text{B-8}$$

$$\alpha(\theta) = M(2\theta)$$

where

$$M(\theta) = \int_0^\infty e^{-V - \left(\frac{\theta}{2}\right) V^2} dV = \sqrt{\frac{\pi}{2\theta}} e^{\frac{1}{2\theta}} \left\{ 1 - \operatorname{erf} \left[\frac{1}{\sqrt{2\theta}} \right] \right\} \quad \text{B-9}$$

$$\beta = \frac{1 - M(\theta)}{M(\theta) - \alpha(\theta)} \quad \text{B-10}$$

$M(\theta)$ and $M(2\theta)$ are evaluated using Hastings'³ approximation for the erf.

$$\eta = \frac{1}{1 + \frac{P}{\sqrt{2\theta}}} \quad , \quad \lambda = \frac{1}{1 + \frac{P}{\sqrt{4\theta}}} \quad \text{B-11}$$

Then

$$M(\theta) = (a_1 \eta + a_2 \eta^2 + a_3 \eta^3) \frac{\sqrt{2}}{\sqrt{\theta}} \quad \text{B-12}$$

and

$$M(2\theta) = \alpha(\theta) = (a_1 \lambda + a_2 \lambda^2 + a_3 \lambda^3) \frac{1}{\sqrt{\theta}} \quad \text{B-13}$$

The constants a_1 , a_2 , a_3 , and P are found in Ref. 3. Thus one can compute the homogeneous resonance integral for a single peak as a function of σ_p and the temperature.

Levine^{4,5} has produced an equivalence relation which allows the correct calculation of the resonance integral by use of an effective scattering per atom. For a lumped absorber in a moderating medium, his effective scattering per atom, σ_p^* , for resolved resonances is given by

$$\sigma_p^* = \frac{f \sigma_p^m}{1.27} + \frac{S}{4N_A V} \frac{\gamma}{(1.1 - .1\gamma)} \quad \text{B-14}$$

and for unresolved resonances

$$\sigma_p^{eq} = \frac{\sigma_p^a + \sigma_p^m}{1.27} + \frac{S}{4N_A V} \frac{\gamma}{(1.1 - .1\gamma)} \quad \text{B-15}$$

In Eqs. B-14 and B-15, f is the fraction of scatterings which are effective in a resonance (0.7 for oxygen), σ_p^m is the potential scattering of the moderator atoms in the fuel lump per atom of absorber, S/V is the surface to volume ratio for the lump, N_A is the number of absorber atoms per cm^3 of the lump, γ is the Dancoff factor, and σ_p^a is the potential scattering per atom for the absorber.

The unresolved contribution to the resonance integral is obtained from a two point fit to the expression

$$R_{ur}(E) = AE^B \quad \text{B-16}$$

where $R_{ur}(E)$ is the resonance integral, E is the neutron energy, and A and B are dependent on the effective scattering per atom and temperature for the unresolved resonance region. The fit is obtained from resonance parameters given at two energies, namely 2000 ev and 8000 ev. The neutron width Γ_n is computed

$$\Gamma_n = \Gamma_n^0 \sqrt{E} \quad B-17$$

where Γ_n^0 is the average reduced neutron width and the gamma width Γ_γ is taken as the average value for the resolved resonances. The two values of $R_{ur}(E)$ are calculated using Eq. B-7 with the appropriate scattering per atom and temperature. Once A and B are found, the contribution to the resonance integral is

$$R_{ur} = \frac{1}{d} \int R(E) dE \quad B-18$$

where d is the average energy spacing between lines.

Integration of Eq. B-18 yields

$$R_{ur} = \frac{1}{d} \left[\frac{A}{B+1} \right] \left[E_{\text{upper limit}}^{B+1} - E_{\text{lower limit}}^{B+1} \right] \quad B-19$$

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3. Cecil Hastings, Jr., Approximations for Digital Computers, Princeton University Press, 1955.
4. M. M. Levine, General Resonance Integral Representation, Nuclear Science & Engineering, 8(4): 363 (October 1960).
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APPENDIX C

SELF-SHIELDING FACTORS

Analytical Methods

The highly absorptive nature of the Np (and the boron burnable poison) rods and the strong perturbation of the neutron flux and energy distribution near the rods poses severe problems in the accurate treatment of the self-shielding. The direct application of multi-group transport theory, while correct in principle, is far too complex to be useful for practical applications. Methods of strong absorber analysis (i.e., control rods) frequently utilize diffusion theory in one way or another, but supplement it by imposing a boundary condition at the surface of the control rod which in effect will force diffusion theory to yield nearly the same values as the more exact transport analysis for the reactivity, the neutron current into the rod, and the neutron flux at some distance from the rod surface. In CNROD, the transport boundary condition is imposed by the concept of the linear extrapolation distance (d), which requires that the ratio of neutron flux at the surface of the rod to the total neutron current into the rod from the surrounding diffusion medium be that required by transport theory within the control rod, i.e.,

$$\left. \frac{D \nabla \phi}{\phi} \right]_{\text{rod surface}} = \frac{D}{d} \quad \text{C-1}$$

where D is the diffusion coefficient of the surrounding medium, and d is the linear extrapolation distance obtained by transport theory.

The linear extrapolation distance (d) can be obtained from the following relationship in slab geometry

$$d = 0.7104 \lambda_{tr} \left[\frac{1+p_1}{1+p_0} \right] \quad C-2$$

by normalizing to the known transport extrapolation distance ($0.7104 \lambda_{tr}$) for a black absorber (p_0 and p_1 equal zero); p_0 and p_1 are the probabilities of a neutron escaping from the rod without being captured, for the isotropic and anisotropic components of the flux, respectively; and λ_{tr} is the transport mean free path of the surrounding diffusion medium. The approximation for the linear extrapolation distance (d) in cylindrical geometry is as follows:

$$d = \lambda_{tr} \left[0.7104 C + \frac{1.333 p_0}{1-p_0} \right] \quad C-3$$

in which the second-order terms, distinguishing between p_0 and p_1 , are neglected. The factor C accounts for the curvature of the outer surface of the rod and may be obtained from the Reactor Physics Constants Handbook.¹ An approximate expression for C in terms of the radius

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¹ Reactor Physics Constants, ANL-5800, Second Edition, U.S. Atomic Energy Commission, July 1963.

of the rod, R, has been given by Pomraning² as follows:

$$C = \left[\frac{1 + \frac{0.7603795}{R/\lambda_{tr}}}{\frac{0.40515635}{R/\lambda_{tr}}} \right] \quad C-4$$

The escape probability terms, p_0 and p_1 , may be evaluated by the following expressions.

Slab of thickness t and absorption cross-section Σ

$$p_0(\Sigma t) = 2 \int_0^1 e^{-\Sigma t/x} x dx = 2 E_3(\Sigma t) \quad C-5$$

$$p_1(\Sigma t) = 3 \int_0^1 e^{-\Sigma t/x} x^2 dx = 3 E_4(\Sigma t) \quad C-6$$

Cylinder of radius r and absorption cross-section Σ

$$p_0(\Sigma r) = 1 - \frac{4}{3}(\Sigma r)^2 \left\{ 2 \left[\Sigma r I_0(\Sigma r) K_0(\Sigma r) + \Sigma r I_1(\Sigma r) K_1(\Sigma r) - 1 \right] \right. \\ \left. + K_1(\Sigma r) I_0(\Sigma r) - I_1(\Sigma r) K_0(\Sigma r) + \frac{I_1(\Sigma r) K_1(\Sigma r)}{\Sigma r} \right\} \quad C-7$$

In the thermal neutron group, the effective absorption cross-section, Σ_a , was evaluated on the assumption that the neutron spectrum

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²G. C. Pomraning, Control Worth of B₄C Rods, Report GEAP-3800, General Electric Co., August 1961.

(i.e., thermal group < 0.625 ev) was determined primarily by the spectrum in the fuel region surrounding the absorber. Since the absorber would "see" this spectrum, the effective cross section of the absorber was determined by averaging over the spectrum calculated by the TEMPEST code in the main line program.

For neutrons of energy greater than 0.625 ev, the variation in number density with energy and the marked dependence on resonance absorption structure made it necessary to use two different analytical techniques. For materials of smoothly varying absorption cross section (i.e., boron with a $1/v$ dependence), the effective escape probability was determined by averaging over a $1/E$ spectrum above 0.625 ev, i.e.,

$$\bar{P} = \frac{\int_{0.625}^{E_{\max}} p(E) \frac{dE}{E}}{\int_{0.625}^{E_{\max}} \frac{dE}{E}} \quad \text{C-8}$$

In practice, the integration was performed numerically using 300 energy points of approximately equal lethargy interval.

For materials with a marked resonance absorption structure, integration over the > 0.625 ev neutron flux would be excessively time consuming. Consequently, an effective absorption cross section was determined from the reported resonance parameters using the equations for calculating the infinite-dilute resonance integral and a semi-empirical

expression for determining the self-shielding for each resonance peak.

The infinite-dilute resonance integral in a $1/E$ spectrum is given by

$$RI = 4.084 \times 10^3 \frac{\Gamma_n \Gamma_\gamma g}{\Gamma E_r^2} \quad C-9$$

where Γ_n, Γ_γ and Γ are the neutron, gamma, and total reaction widths, respectively; g is the statistical weighting factor; and E_r is the energy of the resonance.

Effective self-shielding factors for each resonance peak were calculated by the following approximation.

$$ss = e^{-2N\sigma_p r} (I_0(2N\sigma_p r) + I_1(2N\sigma_p r)) \quad C-10$$

where N is the atom density of the absorber rod, r is the radius of the rod, and σ_p is the Doppler-broadened peak absorption cross section of the resonance. Using Amster's expressions for the peak absorption cross section (Doppler-broadened) and the Hastings approximation for the probability function (or error integral), the value of σ_p was estimated as follows:

$$\sigma_p = \frac{4.608 \times 10^6 \Gamma_n g}{E_r \Gamma} \frac{\gamma}{\sqrt{T}} \{ .34802 z - .09587 z^2 + .74785 z^2 \} \quad C-11$$

where T is the absorber temperature ($^{\circ}K$), $\gamma = \Gamma/4(E_r k/A)^{\frac{1}{2}}$ (k is the Boltzman constant, 8.6138×10^{-5} , and A is the atomic weight), and $z = 1/(1 + .47078 \gamma/\sqrt{T})$.

The total effective resonance integral is then the sum of the contributions from each of the shielded resonances and the effective smeared group cross section is the effective resonance integral divided by the total lethargy interval, Δu (i.e., $= \ln(E_{\max}/.625)$). The value of the effective absorption cross section is then used in Eq. C-7 to obtain the average escape probability.

Equation C-1 above effectively imposes a transport boundary condition on the surface of the rod in terms of the extrapolation distance, d , i.e., $\frac{\Delta \phi}{\phi} = \frac{1}{d}$. Conventional diffusion theory (using cylindrical rod geometry for example) leads to the following equation for the ratio of the derivative of the flux to the flux at the surface of the absorber.

$$\frac{\Delta \phi}{\phi} = k \frac{I_1(kr)}{I_0(kr)} \quad , \text{ where } k \equiv \sqrt{\Sigma_a/D} \quad \text{C-12}$$

Equating the transport boundary condition (Eq. C-1) and the diffusion theory result (Eq. C-12), the following expressions can be obtained.

Slab geometry

$$d = \frac{1}{k} \left\{ \frac{e^{kt} + e^{-kt}}{e^{kt} - e^{-kt}} \right\} \quad \text{C-13a}$$

(t is the slab half-thickness)

Cylindrical geometry

$$d = \frac{1}{k} \left\{ \frac{I_0(kr)}{I_1(kr)} \right\} \quad \text{C-13b}$$

Annular geometry

$$d = \frac{1}{k} \left\{ \frac{I_0(kr_2) k_1(kr_1) + I_1(kr_1) k_0(kr_2)}{I_1(kr_2) k_1(kr_1) - I_1(kr_1) k_1(kr_2)} \right\} \quad \text{C-13c}$$

(r_2 and r are the outer and inner radii, respectively)

Knowing the value of d from Eqs. C-2 or C-3, Eq. 13 may be solved for the value of k , and hence a fictitious value of $\Sigma_a (= D k^2)$ is obtained which, when used in diffusion theory, will impose a logarithmic boundary condition at the surface of the absorber.

A special subroutine was written and added to the CNROD code to utilize these equivalent diffusion theory constants in a simple one-group cylindrical calculation of the average flux in the absorber rod to that in the fuel of a relatively large volume surrounding the absorber rod. The ratio of the two fluxes was defined as the over-all self-shielding factor for fitting with a polynomial and subsequent use in the main-line program.

Calculated self-shielding factors are shown in Figs. C-1 and C-2.

Figure C-1 Thermal group self-shielding factors as a function of concentration for annular and rod-type burnable poison elements.

Figure C-2 Epithermal group self-shielding factors as a function of concentration for B-10, Np-237, and Pu-238.

APPENDIX D

BURNUP MODEL FOR NEPTUNIUM TARGETS

A nuclear diagram showing reactions considered for Np-237 targets in pressurized water reactors is given below.

One-group cross sections, averaged over the calculated neutron spectrum for each region in the reactor, and at each time step in the burnup calculations, describing the reactions indicated in the diagram, are defined below.

σ_{37}^A - n, γ cross section for ${}^{237}_{93}\text{Np}$

$\sigma_{37}^{n,2n}$ - $n,2n$ cross section for ${}^{237}_{93}\text{Np}$

σ_{46}^A - Total absorption cross section for ${}^{236}_{94}\text{Pu}$

σ_{38}^A - Total absorption cross section for ${}_{93}\text{Np}^{238}$

σ_{48}^A - Total absorption cross section for ${}_{94}\text{Pu}^{238}$

The differential equations describing the reactions indicated in the diagram are:

$$\frac{dN_{37}}{dt} = -N_{37}(t) \sigma_{37}^A \quad \text{D-1}$$

$$\frac{dN_{38}}{dt} = N_{37}(t) \sigma_{37}^A \phi - \lambda_{38} N_{38}(t) - \sigma_{38}^A \phi N_{38}(t) \quad \text{D-2}$$

$$\frac{dN_{48}}{dt} = \lambda_{38} N_{38}(t) - \sigma_{48}^A \phi N_{48}(t) - \lambda_{48} N_{48}(t) \quad \text{D-3}$$

$$\frac{dN_{46}}{dt} = 0.33 \times N_{37}(t) \sigma_{37}^{n,2n} \phi - \lambda_{46} N_{46}(t) - \sigma_{46}^A \phi N_{46}(t) \quad \text{D-4}$$

where the subscripts indicate the last digit in the atomic number and mass number, respectively, of the isotope in question, e.g., 37 is ${}_{93}\text{Np}^{237}$.

The λ 's in the above equations are decay constants, ϕ is the power normalized neutron flux in neutrons/cm² sec, σ is the cross section in cm² and N is the concentration in atoms/cm³; t is the time variable.

Solutions to Eqs. D-1 through D-4 are:

$$N_{37}(t) = N_{37s} \cdot e^{-\sigma_{37}^A \phi \Delta t} \quad \text{D-5}$$

$$\frac{\lambda_{38}}{\sigma_{38}^A} = \frac{\lambda_{38}}{\phi} + \sigma_{38}^A \quad \text{D-6}$$

$$A = \frac{\sigma_{37}^A N_{37s}}{(\sigma_{37}^A - \bar{\sigma}_{38}^A)} \quad D-7$$

$$B = N_{38s} + \frac{\sigma_{37}^A N_{37s}}{(\sigma_{37}^A - \bar{\sigma}_{38}^A)} \quad D-8$$

$$N_{38}(t) = -A \cdot e^{-\sigma_{37}^A \phi \Delta t} + B \cdot e^{-\bar{\sigma}_{38}^A \phi \Delta t} \quad D-9$$

$$\bar{\sigma}_{48}^A = \frac{\lambda_{48}}{\phi} + \sigma_{48}^A \quad D-10$$

$$D = \frac{\lambda_{38} \cdot A}{(\sigma_{37}^A - \bar{\sigma}_{48}^A) \phi} \quad D-11$$

$$E = \frac{\lambda_{38} \cdot B}{(\bar{\sigma}_{38}^A - \bar{\sigma}_{48}^A) \phi} \quad D-12$$

$$F = N_{48s} - D + E \quad D-13$$

$$N_{48}(t) = D \cdot e^{-\sigma_{37}^A \phi \Delta t} - E \cdot e^{-\bar{\sigma}_{38}^A \phi \Delta t} + F \cdot e^{-\sigma_{48}^A \phi \Delta t} \quad D-14$$

$$\bar{\sigma}_{46}^A = \frac{\lambda_{46}}{\phi} + \sigma_{46}^A \quad D-15$$

$$G = 0.33 \left(\frac{N_{37s} \cdot \overset{n,2n}{\sigma_{37}^A}}{\sigma_{37}^A - \bar{\sigma}_{46}^A} \right) \quad D-16$$

$$H = G + N_{46s} \quad D-17$$

$$N_{46}(t) = -G \cdot e^{-\sigma_{37}^A \phi \Delta t} + H \cdot e^{-\sigma_{46}^A \phi \Delta t}$$

D-18

The subscripts, s, on atomic concentrations, appearing in the above solutions, indicates the value at the beginning of the time interval, Δt .

Equations similar to those for isotopes of importance in the neptunium targets were derived for the isotopes of importance in the reactor fuel and in the burnable poison material. These included: U-235, U-236, U-237, Np-237, Np-238, Pu-238, Pu-236, U-238, Pu-239, Pu-240, Pu-241, Pu-242, I-135, Xe-135, Pm-149, Sm-149, fission products from U-235 fission, fission products from U-238 fission, fission products from Pu-239 fission, fission products from Pu-241 fission, and B-10. Although the decay of Cm-242 contributes to the production of Pu-238 in reactor fuel, this contribution was assumed to be small and was not included. The analytic solutions for all concentrations indicated above were incorporated in the burnup routine used in the one-dimensional model.

Figure 1 Sketch of the reference pressurized water reactor. (Numbers indicate the number of burnable poison rods in a particular assembly.)

- Poison uniformly distributed over full active core height
- Zircaloy cladding tube same as used for fuel pins
- Guide tube existing

Figure 2 Np-237 rod, reference design.

Figure 3 Pu-240 self-shielding factors, 0.8255 cm radius rod.

Figure 4 Photograph of the critical experiment used to test the model for reactivity calculations.

Figure 5 Net U -236 as a function of exposure in Yankee spent fuel.

Figure 6 Net Np-237 as a function of exposure in Yankee spent fuel.

Figure 7 Net Pu-238 as a function of exposure in Yankee spent fuel.

Figure 8 Pu-236 to Pu-238 mass ratio in Yankee spent fuel as a function of exposure.

Figure 9 Soluble poison concentration required for criticality as a function of burnup.

Figure 10 Net Pu-238 and Np-237 as a function of time for target rods (535 grams Np-237/rod) in various spatial arrangements within the reactor.

Figure 11 Net Pu-238 and Np-237 as a function of time for varying numbers of target rods in the same geometric configuration (ratio of the number of Np rods to total burnable poison rods constant in each region).

Figure 12 Net Pu-238 and Np-237 as a function of time for constant initial amount of Np-237 in varying numbers of rods (the variable is the Np loading per rod).

Figure 13 Power distribution as a function of burnup for the reference reactor Prob. 1.

FIGURE 14 POWER DISTRIBUTION AS A FUNCTION OF BURNUP FOR PROBLEM 2

FIGURE 15 POWER DISTRIBUTION AS A FUNCTION OF BURNUP FOR PROBLEM 3

FIGURE 16 POWER DISTRIBUTION AS A FUNCTION OF BURNUP FOR PROBLEM 4

FIGURE 17 POWER DISTRIBUTION AS A FUNCTION OF BURNUP FOR PROBLEM 5

FIGURE 18 POWER DISTRIBUTION AS A FUNCTION OF BURNUP FOR PROBLEM 7

FIGURE 19 POWER DISTRIBUTION AS A FUNCTION OF BURNUP FOR PROBLEM 8

FIGURE 20 POWER DISTRIBUTION AS A FUNCTION OF BURNUP FOR PROBLEM 9

FIGURE 21 POWER DISTRIBUTION AS A FUNCTION OF BURNUP FOR PROBLEM 10

FIGURE 22 POWER DISTRIBUTION AS A FUNCTION OF BURNUP FOR PROBLEM 11

Figure 23 Net Pu-238 and the mass ratio of Pu-236 to Pu-238 as a function of burnup for different Np-237 loadings per rod.

Figure 24 Net Pu-238 and Pu-236 to Pu-238 mass ratio @ 9,500 Mwd/mtU as a function of Np-237 loading per rod. Total initial Np loading, 1.368 kg/mtU.

Figure 25 Net Pu-238 and Np-237 as a function of time for the practical problem.

Figure 26 Power distribution for practical problem.

Figure 27 Absorption rates in B-10 in region 4 for the reference problem, and in Np-237, Pu-238, and B-10 for region 4 of Problem 5 as a function of time.

Figure 28 Projected Pu-238 production capability in U.S. PWR's 1972-1981.