1962

Some effects of Pu-240, Pu-241, and Pu-242 on the Pu-239 critical mass of a fast reactor

Frank Peter Mertes Jr.
Iowa State University

SOME EFFECTS OF PU-240, PU-241, AND PU-242 ON THE PU-239 CRITICAL MASS OF A FAST REACTOR

by

Frank Peter Mertes, Jr.

A Dissertation Submitted to the
Graduate Faculty in Partial Fulfillment of
The Requirements for the Degree of
DOCTOR OF PHILOSOPHY

Major Subject: Nuclear Engineering

Approved:

Signature was redacted for privacy.
In Charge of Major Work

Signature was redacted for privacy.
Head of Major Department

Signature was redacted for privacy.
Dean of Graduate College

Iowa State University
Of Science and Technology
Ames, Iowa

1962

TABLE OF CONTENTS

I. LIST OF SYMBOLS

b	A variable in the difference equations defined in Equation 18	
B_c^2	Buckling in the core	$cm.^{-2}$
c_n	A variable in the difference equations defined in Equation 19	neutrons/$cm.^3$-sec.
D	Diffusion coefficient	cm.
D_c	Diffusion coefficient in the core	cm.
D_r	Diffusion coefficient in the reflector	cm.
D'	Diffusion coefficient in the blanket	cm.
E	Neutron energy	MEV
H_n	The solution to the homogeneous difference equation	
L_r	Attenuation length	cm.
m	A subscript indicating core blanket boundary in the difference equations	
n	A subscript indicating radius position in the difference equations	
N_{xy}	Concentration of nuclide	atoms/$cm.^3$
p	Resonance escape probability	
P_1	Fission to resonance non-leakage probability	
P_n	The particular solution to the difference equation	
R	Feed enrichment ratio	
R_c	Core radius	cm.
S	Neutron source strength	neutrons/$cm.^3$-sec.
t	Time	sec.
T'	Reflector thickness	cm.
α	Ratio of capture cross section to fission cross section	
α_n	A variable in the difference equations defined in Equation 30	

β_n A variable in the difference equation defined in Equation 32.

$\bar{\alpha}$ Excess reactivity

ϵ Fast fission factor

η Fission neutron produced per thermal neutron absorbed

θ Flux-time

ν Average number of neutrons per fission

σ Microscopic cross section $cm.^2$/atom

Σ_a Absorption cross section $cm.^{-1}$

Σ_{er} Cross section for neutrons removed to the next lowest energy group by elastic scattering $cm.^{-1}$

Σ_f Macroscopic fission cross section $cm.^{-1}$

$\Sigma_{n,n\ j\rightarrow j+k}$ Cross section for neutrons transferred from group j to j + k by inelastic scattering $cm.^{-1}$

Σ_{tr} Macroscopic transport cross section $cm.^{-1}$

φ Neutron flux neutrons/$cm.^2$-sec.

$\bar{\Phi}$ Neutron flux divided by radius neutrons/$cm.^3$-sec.

II. SUMMARY

This study considered the effects of variation in the isotopic composition of plutonium used as a fast reactor fuel material. When the plutonium fuel material is produced in a thermal reactor, the primary factor affecting composition is the flux time of irradiation θ, $10^{21} cm^{-2}$. Flux times of $\theta = 1$, $\theta = 5$, and $\theta = 10$ were studied. It was found that for a flux time of $\theta = 10$, the concentration of Pu-239 which yielded a critical mass was reduced to a concentration of 7.42% of the fuel from a concentration of 14.1% for the pure Pu-239 case. In all cases studied the spherical core consisted of 35% fuel, 15% Fe, and 50% Na by volume and had a radius of 31 centimeters. The core was surrounded by a blanket of 70% U, 10% Fe, and 20% Na by volume which extended to a radius of 75 centimeters.

The critical masses of the fast reactor were calculated by a one dimensional, two region, 16 neutron energy group computer code written in IBM FORTRANSIT. This code also was used to tabulate the neutron flux in 16 energy groups at each of 75 radius points. No significant change in the flux spectra resulted from the changes in plutonium isotopic composition. The peak flux was in the energy range $0.3 \text{ MEV} < E < 0.5 \text{ MEV}$. While the computer code was written in the FORTRANSIT coding system for the IBM 650, the program is compatible with the FORTRAN coding system and could be employed on any machine using FORTRAN. The program could be used to compute the critical mass of any spherical fast reactor with one or two regions. The choice of fuel, coolant, and structural materials in the reactor is in no way restricted by the program instructions.

For the particular reactor studied, a one neutron energy group diffusion theory calculation predicted approximately the same changes in critical mass as were obtained from the computer solution.

The plutonium fuel was assumed to be produced in a thermal plutonium recycle reactor with an average enrichment of 1%. The set of differential equations detailing the buildup of Pu-239, Pu-240, Pu-241, and Pu-242 in the thermal reactor was solved by use of an electronic analog computer.

III. INTRODUCTION

It has been stated that the breeder reactor is probably the closest approach to a perpetual motion machine that our technology has yet devised. The fast breeder reactor not only produces power but also produces fissionable plutonium in excess of its own needs for fuel. While this intrinsic fuel saving is attractive, today it is the thermal reactor --not the fast reactor--that has been most intensively developed.

One of the main difficulties in the development of the fast breeder reactor has been the very heavy investment in fissionable material needed for this type of power reactor. Some conceptual designs have considered cores containing upwards of 500 kilograms of Pu-239. When Pu-239 is valued at $12 per gram, the fissionable material costs are $6 million.

The existing thermal reactors, that already have reached a comparatively high level of development, would seem to be the most logical source of the plutonium needed for the fast breeder reactors. While any thermal reactor containing U-238 will produce plutonium, the quantity and the isotopic composition of the plutonium produced will vary widely. The quantity of plutonium produced is dependent upon the amount of U-238 present. This varies widely; at one extreme are the highly enriched reactors containing only a few percent of U-238, and at the other extreme are the natural uranium reactors containing 99.3% U-238. The composition of the plutonium also will change, since plutonium produced in a reactor is a mixture of isotopes having varying nuclear properties. The primary factor affecting composition is the flux-time of irradiation; that is, as flux-times of irradiation increase, the plutonium isotopes of higher

atomic weight tend to build up. The extreme example of high irradiation flux-times is that of the plutonium recycle reactor. Here the plutonium is recycled through several fuel loadings, resulting in very large flux-times of irradiation. The calculation of the buildup of these higher isotopes will tend to be complicated by the fact that three fissionable nuclides, U-235, Pu-239, and Pu-241, serve as sources of neutrons in the thermal reactor. Pu-240 and Pu-242 are not fissionable by neutrons of thermal energy.

Since these variations exist in the plutonium produced by the thermal reactors, it may be expected that the design of a fast breeder reactor could be influenced by the composition of the plutonium used as fuel. In the fast reactor both Pu-240 and Pu-242 are fissionable in neutron fluxes with energies greater than 0.1 MEV, and thus fission is possible for all of the plutonium isotopes present. The description of a fast reactor is inherently much more difficult to handle mathematically than the description of the thermal reactors. Since a wide variation in neutron energies exists and the properties of the reactor materials are a function of this energy, the neutron flux at any point in the reactor must be subdivided into neutron fluxes within various ranges of neutron energy. Account must be taken of the production, capture, fission, the elastic scattering, and the inelastic scattering for each of these energy ranges Thus a computer solution of the problem is strongly suggested.

From the above discussion it can be seen that when plutonium produced in a thermal reactor is used as a fast reactor fuel, variations in the isotopic composition of the plutonium can possibly affect the fuel investment problem in fast reactors. A study of the effect of this variation in plutonium isotopic composition is the purpose of this research.

IV. LITERATURE REVIEW

An extensive development of fuel cycle analysis has been published by Benedict and Pigford (2), but the work was restricted to fuel cycles in thermal reactors. Neutron balance equations were written for a "just critical" reactor with the initial loading of fuel assumed to be present. Xenon and samarium were the only fission products assumed to be present, and these were considered to be at equilibrium concentrations dependent upon the designed power level. A second neutron balance equation was written for the reactor at an arbitrary flux time θ. When these two equations were combined, a feed-enrichment ratio R was obtained, which was defined as the ratio of U-235 to U-238 in the actual reactor feed relative to that in the reference design. This feed enrichment ratio was then related to the flux time of reactor operation. The analysis was carried out for several cases, including completely mixed fuel with no plutonium recycle, unmixed fuel with no plutonium recycle, and unmixed fuel with plutonium recycle. A set of differential equations was derived which detailed the change in fuel composition with time for the unmixed fuel case. Values for the concentrations of U-235, U-236, Pu-239, Pu-240, Pu-241, and Pu-242 were calculated for the case of the unmixed feed, plutonium recycle reactor.

A further extension of this work by Shanstrom, et al. (17) resulted in the development of the computer code FUELCYC, usable for calculating the effect on the fuel cycle of different methods of scheduling replacement of fuel and movement of control poisons. These included batch, inout, outin, and graded movement of the fuel elements. The computer code

FUELCYC was two dimensional and used two groups of neutron energies. Outin fuel movement was found to be the economical method of fuel scheduling for a reactor similar to Yankee. Use of computer code FUELCYC resulted in a calculation for the average burnup which was 33% higher than the calculation of the average burnup under the assumption of a time-invariant chopped-cos, chopped Jo thermal flux distribution used in a simpler code. This was attributed to the fact that the flux and power density in an irradiated core were grossly different from the flux and power density in a uniform core.

Starr (18) discussed a general approach to enriched plutonium recycle reactors which did not consider such reactor details as homogeneous versus heterogeneous cores, solid versus fluid cores, or fixed versus circulating cores. The condition assumed for the plutonium feeder-converter system was that the enriched reactor plants were to be supported from plutonium produced in a natural reactor plant. The complete fuel cycle of such combined plants were considered to operate with natural uranium feed. A rather elaborate cost comparison, including projected reprocessing costs, was calculated for the feeder-converter complex. This was considered to include a natural uranium reactor power plant (feeder reactor) which discharged fuel for plutonium removal after approximately 3,000 megawatt days per ton irradiation. The plutonium which was removed from the fuel was assumed to be fed into the enriched reactor plants, and the uranium remaining in the fuel was assumed to be discarded. The enriched reactor plants, which were considered to burn mostly plutonium and convert U-238 to plutonium, were designated as plutonium converters in a plutonium feeder-converter system. Some typical values assumed for

the feeder reactor were 100 megawatt thermal power with an initial conversion ratio of 0.8. For 3,000 megawatt days per ton fuel irradiation and 100% load factor, the reactor would produce 25.8 kilograms of plutonium with isotopic content of 79% Pu-239, 16% Pu-240, and 5% Pu-241. The plutonium converter reactor system considered was of a type capable of operating with 2% U-235 enriched uranium and an initial conversion ratio of 0.7. It was considered necessary to discard some of the plutonium recycle stream to stop the buildup of higher isotopes in the plutonium converter reactor which irradiated the fuel for 10,000 megawatt days per ton and utilized the plutonium from the feeder reactor as a feed material. When 17.8 kilograms of plutonium per year were considered to be discarded, the isotopic plutonium recycle concentration was calculated to be 47.2% Pu-239, 20.5% Pu-240, 11.4% Pu-241, and 20.9% Pu-242.

Greebler, et al. (6) considered the recycle of plutonium in light water reactors. They emphasized that the calculation of isotopic compositions and reactivities in reactors with close-packed lattices—and especially in light water reactors—is sufficiently complex so that simplified models of reactor spectra cannot be used. The resonance effect in close-packed lattices was considered to necessitate the calculation of the geometric shielding and the shifts in the thermal neutron spectra with changes in isotopic composition. A model was developed which considered the details of resonance effects, spectrum shifts, geometric effects, changes in flux with composition, and practical partial batch refueling schedules. The uncertainties caused by variations in nuclear parameters were calculated. For example, a variation of 10% in the value of α for Pu-241 affected the calculated value of the reactivity of the

pseudo-steady state by about 2%. Consideration was also given to the startup transient which was traversed in the approach to the pseudo-steady state conditions. Startup cycles were developed which converged to the desired asymptotic state. The relation of the startup cycle cost to the cost of the pseudo-steady state and to the lifetime average fuel cost was determined. A calculation for the required inventory was developed. The normal inventory considered to be required for the reactor and its supply line was 1.5 core loads of enriched fuel plus an additional inventory of 0.5 core loads of enriched fuel for the first 15 years of operation and a further addition of 1.0 core loads of enriched fuel for the second 15 years of operation. Operation on natural uranium makeup alone required design for maximum reactivity and short cycle lives, resulting in fuel cycle costs which were about double the cost of optimum cycles with slight enrichment.

Several attempts have been made to determine the quantities of materials required for the long-term development of national nuclear programs. Andriot and Gaussens (1) described the French requirements for a long-term nuclear power buildup. Comparisons were made on the basis of required uranium production for varying approaches, including the natural uranium and graphite approach, the natural uranium and heavy water approach, and the enriched uranium and heavy water approach. Requirements were considered in terms of desired power production and doubling times.

Dayal, et al. (4) considered a three-phase approach to the problem of India's power requirements through use of natural uranium reactors, Pu-Th converters, and U-233-Th breeders. This three-phase approach was considered to permit the buildup of a nuclear electrical capacity based

only on natural uranium and thorium feed supplies. This was stated to be of particular interest because of the relatively scarce uranium and very abundant supplies of thorium in India.

Work on fuel cycles for plutonium fueled fast breeder reactors was reported by Hall (7). The breeding ratios and critical masses were computed by a 10-group S_n method in a two-region spherical geometry. The outer region was in all cases considered to be a breeding blanket of infinite thickness of the composition 70% U, 10% Fe, and 20% Na. The iron and sodium were assumed arbitrarily as provisions for structural materials and coolant. Various core configurations were considered, including Pu-U, PuO_2-UO_2, Pu-Al, Pu-Fe, and Pu-U-Fe. The minimum mass of plutonium required for a critical core was found to vary from 25 to 150 kilograms, and the breeding ratio was found to vary from 1.4 to 2.2 for the above cores. A special core with equilibrium Pu in U, varying from 0% to 10% for a continuous transition from blanket to core, was calculated to have a core mass of 2,400 kilograms and a breeding ratio of 1.5. In this case natural uranium was assumed to be introduced at the edge of the blanket region and to be continuously moved toward the center as the plutonium built up and was eventually discharged from the center. Thus it was possible to utilize 25% of the total energy content of the uranium. However, radiation damage was not considered as limiting over this large burnup.

Several authors discussed alternative methods of calculating fast reactors and the advantages of various methods in attaining agreement with critical assembly data. Loewenstein and Okrent (11) discussed the conceptual design of fast neutron breeder reactor systems in terms of

existing critical assembly data. They considered the S_n method to be the best approximation to the solution of the Boltzman equation at the present time. This S_n method and the diffusion theory method were employed for specific calculations. They found that normalized one-dimensional diffusion theory calculations predicted criticality rather well for cores which were uniformly reflected with a high density uranium blanket. The accuracy of the diffusion theory approximation decreased as the reflector density and/or the reflector uniformity decreased.

Carlson and Bell (3) discussed the solution of the transport equation by the S_n method, including fast reactor calculations. Use of a three-group spectrum of 0-0.4 MEV, 0.4-1.4 MEV, and 1.4 MEV-infinity enabled computation of critical radii to an accuracy of approximately 2%. By use of a six-group spectrum, 0-0.1-0.4-0.9-1.4-3.0 MEV-infinity, critical radii of experimental systems were predicted with an accuracy of within 1%.

Roach (15) discussed a computational survey of idealized fast breeder reactors. He used a S_4 approximation with 16 energy groups of neutrons which led to a 5% uncertainty in radius due to cross section data uncertainties. The reactors considered were assumed to consist of U-235 and depleted U, Pu-239 and depleted U, and U-233 and Th. Calculated radii were compared with critical assembly data.

Long, et al. (12) discussed the methods of fast neutron power reactor calculations as exemplified by the work on critical assembly ZPR-III. They described the use of multi-group constants for the neutron spectrum determination and critical mass determination for non-spherical systems. Specific results at given energy levels were reported and variation of results due to the various computational schemes were noted.

Hansen (8) discussed the calculation of elementary fast-neutron critical assemblies when a six-group cross section spectrum was used. These were then compared to constants obtained from a critical assembly.

Kiehn (10) discussed internal versus external breeding. Some results of calculation on blanket behavior were as follows: 1) The predominant effect of increasing tantalum capture reaction in the core was to reduce the number of blanket injection neutrons per core fuel absorption; 2) The addition of U-238 to the core reduced the number of available blanket injection neutrons; and 3) The loss of external production ratio was compensated by the increase in internal production ratio. Also, gains in production ratios were noted for blanket enrichment.

Okrent (14) discussed the sensitivity of the breeding ratio in fast reactors to uncertainties in the cross sections. He indicated that variations in the experimentally determined values of α did not result in large changes in the calculated breeding ratio. These variations caused changes from 1.52 to 1.62 in the breeding ratio for a metal system. The systems considered were of simple spherical geometry and were assumed to contain U and Pu-239 plus a coolant (sodium) and a structural material (steel). The blanket was in all cases assumed to be 60% U, 20% steel, and 20% Na. Various cores which contained oxide and carbide fuels were studied. The critical mass, breeding ratio, and the average α of the plutonium were calculated as a function of core size, e.g., 800, 1,500 or 2,500 liters. When reactivity changes for a sudden coolant removal were calculated, the metal system showed a positive reactivity change. The changes in the breeding ratio and the critical mass resulting from substitution of molybdenum structures and tantalum structures for the iron

structures were studied. The breeding ratio was severely decreased to a value below 1.0 when tantalum structures were substituted for the iron structures. Finally, the effects of an increase in fission products on the reactivity worth of a void at the core center were considered.

Kazachkovsky (9) calculated the fuel costs of power produced by fast reactors. Specific consideration was given to an economic breeding ratio, which he defined as the difference between the amount of fuel newly formed and that which had disappeared through radiative capture, divided by the amount of spent fuel. From this he determined a fuel cost based on the immediate expenses of fuel and chemical processing plus the interest on the fuel investment.

V. MATHEMATICAL FORMULATION OF PROBLEM

A. Buildup of Plutonium Isotopes in a Thermal Reactor

Benedict and Pigford (2, p. 65) solved the problem of calculation of buildup of plutonium isotopes in a thermal reactor for two special cases. The first case was that of a reactor with relatively low total irradiation of the fuel and no plutonium recycle. The U-238 concentration was considered to be constant over the period of irradiation, the buildup of Pu-242 was neglected, and the production of Pu-239 from fission of Pu-241 was ignored since the Pu-241 concentration was low. The second case considered was that of steady-state operation with plutonium recycle. For any one fuel loading, the U-238 concentration was assumed to be constant and the Pu-241 concentration was assumed to be a function only of the total irradiation time and was constant during the irradiation. The calculation of buildup of plutonium isotopes for irradiation times such that neither set of assumptions can legitimately be made led to solutions which were exceedingly complex algebraically. Since these irradiation times were of considerable interest to the problem at hand, the solutions were obtained through the use of an electronic analog computer.

The following set of differential equations were obtained by following the general approach of Benedict and Pigford (2, p. 91) but without introducing the limiting assumptions stated above. To simplify the equations, the following notation scheme was used. N_{xy} represented the atoms per cubic centimeter of the isotope in question, x was equal to the atomic number minus 90, and y was equal to the last digit of the mass number.

Thus N_{28} referred to U-238, N_{49} to Pu-239, etc.

The first differential equation in the set was that describing the buildup of U-235, as given in Equation 1.

$$\frac{dN_{25}}{dt} = -N_{25}\ \sigma_{25}\varphi . \qquad (1)$$

The next equation for Pu-239 buildup was somewhat more complicated. The production of Pu-239 was due to four terms: The thermal absorption in U-238, and the resonance absorption of fast neutrons produced by fission of U-235, of Pu-239, and of Pu-241. Removal was by means of thermal and resonance absorption in Pu-239. Also, since the half-life of the Pu-239 precursor was short compared to reactor fuel life, the Pu-239 was treated as if formed directly from U-238. Thus the change in Pu-239 was described by Equation 2.

$$\frac{dN_{49}}{dt} = N_{28}\ \sigma_{28}\varphi + \eta_{25}\ \sigma_{25}\ N_{25}\ \varphi\ \varepsilon\ P_1(1-p) +$$

$$\eta_{49}\ \sigma_{49}\ N_{49}\ \varphi\ \varepsilon\ P_1\ (1-p) + \eta_{41}\ \sigma_{41}\ N_{41}\ \varphi\ \varepsilon\ P_1(1-p) -$$

$$N_{49}\ \sigma_{49}\ \varphi . \qquad (2)$$

In order to simplify the equations, the nomenclature of Benedict and Pigford (2, p. 70) was followed, and the effective thermal absorption cross sections for the plutonium isotopes were increased to allow for resonance absorption of neutrons still in the slowing-down process. This was the σ_{49} used in Equation 2.

The third differential equation in the set described the buildup of

Pu-240. Pu-240 was produced only by capture in Pu-239, as shown in Equation 3.

$$\frac{dN_{40}}{dt} = \frac{N_{49}\ \sigma_{49}\ \alpha_{49}\ \varphi}{1 + \alpha_{49}} - N_{40}\ \sigma_{40}\ \varphi\ . \qquad (3)$$

Pu-241 is produced by capture in Pu-240, as given in Equation 4.

$$\frac{dN_{41}}{dt} = N_{40}\ \sigma_{40}\ \varphi - N_{41}\ \sigma_{41}\ \varphi\ . \qquad (4)$$

Finally, Pu-242 is produced by capture in Pu-241, as shown in Equation 5.

$$\frac{dN_{42}}{dt} = \frac{N_{41}\ \sigma_{41}\ \alpha_{41}\ \varphi}{1 + \alpha_{41}} - N_{42}\ \sigma_{42}\ \varphi\ . \qquad (5)$$

B. Analysis of a Fast Reactor by Single Energy Group - Two Region Diffusion Theory

As a rough guide to the expected behavior of the fast reactor fueled with a mixture of plutonium isotopes, a single group - two region diffusion calculation was useful. This was, of course, a gross simplification of the physical situation. All neutrons were assumed to be born in the energy group 0.3 MEV E 0.5 MEV and not to lose energy by collision. For this one group model Equation 6 was used, which was the critical equation for a reflected sphere as given by Murray (13, p. 68).

$$B_c\ R_c\ \cot B_c\ R_c = 1 - \frac{D_r}{D_c}\left(1 + \frac{R_c}{L_r}\right). \qquad (6)$$

For a particular isotopic composition of plutonium, the total

Pu/U-238 ratio was varied until criticality resulted. This particular Pu-239 critical mass was then considered to be characteristic of the plutonium composition studied.

C. Analysis of a Fast Reactor by Multigroup - Two Region Diffusion Theory

For any neutron energy group, the neutron balance equation may be written using the terms shown in Equation 7.

$$\begin{aligned} &\text{Production - Leakage - Absorption} = \partial N / \partial t \\ &\text{Neutron Leakage per unit volume per second} = -D \nabla^2 \varphi \\ &\text{Absorption} = \Sigma_a \varphi \\ &\text{Production or Source} = S. \end{aligned} \tag{7}$$

Insertion of the above terms in the neutron balance yields Equation 8.

$$S + D \nabla^2 \varphi - \Sigma_a \varphi = \partial N / \partial t. \tag{8}$$

For a spherical geometry, the value of ∇^2 is given in Equation 9.

$$\nabla^2 = \frac{d}{dr^2} + \frac{2d}{rdr}. \tag{9}$$

Substituting Equation 9 into Equation 8 gives Equation 10 for the steady state.

$$D \left[\frac{d^2\varphi}{dr^2} + \frac{2d\varphi}{rdr} \right] - \Sigma_a \varphi + S = 0. \tag{10}$$

If $\bar{\varphi}$ is defined as equal to φr, Equation 10 can be written as Equation 11.

or Equation 12.

$$\frac{d^2 \bar{\Phi}}{r dr^2} - \frac{\Sigma_a \bar{\Phi}}{D r} + \frac{S}{D} = 0. \qquad (11)$$

$$\frac{d^2 \bar{\Phi}}{dr^2} - \frac{\Sigma_a \bar{\Phi}}{D} + \frac{Sr}{D} = 0. \qquad (12)$$

In order to make the equations amenable to a digital computer solution, the differential equations were converted to difference equations. The reactor was considered to be divided into a series of n spherical shells with an equal incremental width Δr. According to the theory of difference equations as stated by Scarborough (16, p. 310), the derivative of a function $\bar{\Phi}(r)$ with respect to r can be approximated by a forward first difference quotient as in Equation 13.

$$\left[\frac{d \bar{\Phi}(r)}{dr}\right]_f \cong \frac{\bar{\Phi}(r + \Delta r) - \bar{\Phi}(r)}{\Delta r} = \frac{\bar{\Phi}_{n+1} - \bar{\Phi}_n}{\Delta r}. \qquad (13)$$

This can be expressed equally well by the backward first difference quotient given in Equation 14.

$$\left[\frac{d \bar{\Phi}(r)}{dr}\right]_b \cong \frac{\bar{\Phi}(r) - \bar{\Phi}(r - \Delta r)}{\Delta r} = \frac{\bar{\Phi}_n - \bar{\Phi}_{n-1}}{\Delta r}. \qquad (14)$$

The second derivative is approximated by Equation 15.

$$\frac{d^2 \bar{\Phi}(r)}{dr^2} \cong \frac{\left[\frac{d \bar{\Phi}(r)}{dr}\right]_f - \left[\frac{d \bar{\Phi}(r)}{dr}\right]_b}{r} =$$

$$\frac{\bar{\Phi}_{n+1} - 2\bar{\Phi}_n + \bar{\Phi}_{n-1}}{(\Delta r)^2}. \qquad (15)$$

In the limit $\Delta r = 0$, the formulas become exact. This indicated that there should appear in the solution of difference equations the counterparts of the familiar complementary and particular integrals. Using the second derivative given in Equation 15, Equation 12 for a space point n then becomes Equation 16.

$$\frac{\Phi_{n+1} - 2\Phi_n + \Phi_{n-1}}{(\Delta r)^2} - \frac{\Sigma_a \Phi_n}{D} + \frac{n S_n \Delta r}{D} = 0. \qquad (16)$$

If Equation 16 is rearranged, it becomes Equation 17 where the constants used are given by Equation 18 and Equation 19.

$$\Phi_{n+1} = b\Phi_n - \Phi_{n-1} - c_n . \qquad (17)$$

$$b = \left[2 + \frac{\Sigma_a (\Delta r)^2}{D}\right]. \qquad (18)$$

$$c_n = \frac{n S_n (\Delta r)^3}{D} . \qquad (19)$$

The complete solution of difference Equation 17 consisted of two parts: H_n, the solution of the homogeneous equation, with $c_n = 0$; and P_n, a particular solution, and is shown in Equation 20.

$$\Phi_n = X H_n + P_n . \qquad (20)$$

Here X is an arbitrary coefficient designed to make Φ_n equal zero at the reactor boundary.

The homogeneous form of Equation 17 is shown in Equation 21.

$$H_{n+1} = b H_n - H_{n-1} . \qquad (21)$$

At this point Ehrlich and Hurwitz (5) suggest the technique of multiplication of Equation 17 by H_n and Equation 21 by Φ_n. This gives Equation 22 and Equation 23.

$$H_n \Phi_{n+1} = b \Phi_n H_n - \Phi_{n-1} H_n - c_n H_n . \tag{22}$$

$$\Phi_n H_{n+1} = b \Phi_n H_n - \Phi_n H_{n-1} . \tag{23}$$

Subtraction of Equation 22 from Equation 23 results in Equation 24.

$$H_{n+1} \Phi_n - H_n \Phi_{n+1} = H_n \Phi_{n-1} - H_{n-1} \Phi_n + H_n c_n . \tag{24}$$

Evaluation of Equation 24 with n = 1 and n = 2 yields Equation 25 and Equation 26.

$$H_2 \Phi_1 - H_1 \Phi_2 = H_1 \Phi_0 - H_0 \Phi_1 + H_1 c_1 . \tag{25}$$

$$H_3 \Phi_2 - H_2 \Phi_3 = H_2 \Phi_1 - H_1 \Phi_2 + H_2 c_2 . \tag{26}$$

Equation 25 is inserted into Equation 26 to give Equation 27.

$$H_3 \Phi_2 - H_2 \Phi_3 = H_1 \Phi_0 - H_0 \Phi_1 + H_1 c_1 + H_2 c_2 . \tag{27}$$

Repeated application of this procedure results in Equation 28.

$$H_{n+1} \Phi_n - H_n \Phi_{n+1} = H_1 \Phi_0 - H_0 \Phi_1 + \sum_{j=1}^{n} H_j c_j . \tag{28}$$

At this point a boundary condition requiring that φ be finite at the center of the reactor was invoked. To prevent an indeterminate solution, Φ_0 must equal 0 since φ equals Φ/r. Since Φ_0 equals 0, H_0 equals 0 also. When these restrictions are put into Equation 28, Equation 29 results.

$$\Phi_n = \frac{H_n}{H_{n+1}} \left(\Phi_{n+1} + \sum_{j=1}^{n} H_j c_j \right). \tag{29}$$

Now a new variable α_n is defined in Equation 30.

$$\alpha_n = \frac{H_n}{H_{n-1}} . \tag{30}$$

Insertion of α_n in Equation 21 yields Equation 31.

$$\alpha_{n+1} = b - \frac{1}{\alpha_n} . \tag{31}$$

An additional new variable β_n is defined in Equation 32.

$$\beta_n = \frac{H_1 c_1}{H_n} + \frac{H_2 c_2}{H_n} + \cdots\cdots\cdots \frac{H_{n-1} c_{n-1}}{H_n} + c_n . \tag{32}$$

H_{n-1}/H_n is factored out, which results in Equation 33 or Equation 34.

$$\beta_n = \frac{H_{n-1}}{H_n} \left(\frac{H_1 c_1}{H_{n-1}} + \frac{H_2 c_2}{H_{n-1}} + \cdots\cdots \right.$$

$$\left. \cdots\cdots c_{n-1} + \frac{c_n H_n}{H_{n-1}} \right) . \tag{33}$$

$$\beta_n = \frac{\beta_{n-1}}{\alpha_n} + c_n . \tag{34}$$

Equation 34 and Equation 31 are inserted into Equation 29 to give Equation 35.

$$\Phi_n = \frac{1}{\alpha_{n+1}} \left[\Phi_{n+1} + \beta_n \right]. \qquad (35)$$

Equation 31, 34, and 35 are used to evaluate the neutron flux for the reactor. Equation 31 is evaluated first. Since Φ_0 must equal zero for a finite flux φ at the center of the reactor, it can be seen from Equation 35 that α_1 equals ∞. From Equation 31, α_2 is seen to be equal to b. Subsequent values of α follow directly from Equation 31 until the outer reactor boundary is reached. Equation 34 is evaluated next. β_0 is not needed for purposes of calculation, and β_1 equals c_1 since α_1 equals ∞. Subsequent values of β follow directly from Equation 34 and the values of α previously calculated. Evaluation of β proceeds to the outer boundary of the reactor. Finally, Equation 35 is evaluated working inwards from the outer boundary of the reactor. Since φ equals zero at the outer boundary and Φ equals φ r, Φ at the outer boundary is zero. Subsequent values of Φ are evaluated directly from Equation 35 and the previously evaluated values of α and β.

For the case of the two region reactor, the additional boundary conditions of continuous neutron flux and current at the boundary were invoked. Flux is continuous from the nature of the calculation of Equation 35. The neutron current is made continuous by putting restrictions on values of α and β at the core blanket boundary. The variables b, S_n, and c_n are all material dependent and change value at the core blanket boundary. The neutron current is defined in Equation 36.

$$\text{Current} = -D \frac{d\varphi}{dr} . \tag{36}$$

In difference equation formulation, this can be approximated by Equation 37.

$$\frac{d\varphi}{dr} \cong \frac{\Delta d\varphi}{\Delta r} . \tag{37}$$

The derivative within the core is given in Equation 38 where m is the space point on the core blanket boundary.

$$\frac{d\varphi}{dr} \cong \frac{\varphi_m - \varphi_{m-1}}{r_m - r_{m-1}} . \tag{38}$$

In the blanket the derivative is approximated as shown in Equation 39.

$$\frac{d\varphi}{dr} = \frac{\varphi_{m+1} - \varphi_m}{r_{m+1} - r_m} . \tag{39}$$

When the currents were equated, Equation 40 resulted. The prime indicates the blanket

$$-D \frac{(\varphi_m - \varphi_{m-1})}{(r_m - r_{m-1})} = -D' \frac{(\varphi_{m+1} - \varphi_m)}{(r_{m+1} - r_m)} . \tag{40}$$

For equal incremental distances in the core and blanket, the equality of Equation 41 holds so that Equation 42 results from Equation 40.

$$r_m - r_{m-1} = \Delta r = r_{m+1} - r_m . \tag{41}$$

$$D(\varphi_m - \varphi_{m-1}) = D'(\varphi_{m+1} - \varphi_m) . \tag{42}$$

Φ_m is equal to $\varphi_m r_m$, and thus Equation 42 may be modified to give

Equation 43.

$$D\left(\frac{\Phi_m}{r_m} - \frac{\Phi_{m-1}}{r_{m-1}}\right) = D'\left(\frac{\Phi_{m+1}}{r_{m+1}} - \frac{\Phi_m}{r_m}\right). \qquad (43)$$

Equation 35 was rewritten as shown in Equation 44 and Equation 45.

$$\Phi_{m+1} = (\Phi_m)(\alpha_{m+1}) - \beta_m . \qquad (44)$$

$$\Phi_{m-1} = \frac{\Phi_m + \beta_{m-1}}{\alpha_m} . \qquad (45)$$

Equation 44 and 45 were next used in Equation 43, yielding Equation 46.

$$\Phi_m \left[\frac{D}{r_m} - \frac{D}{(r_{m-1})(\alpha_m)} + \frac{D'}{r_m} - \frac{D'(\alpha_{m+1})}{r_{m+1}}\right]$$

$$= \frac{D'}{r_{m+1}}(-\beta_m) + \frac{D(\beta_{m-1})}{(r_{m-1})(\alpha_m)} . \qquad (46)$$

The term in brackets on the left is set equal to zero, which solves for α_{m+1} in terms of α_m.

$$\alpha_{m+1} = \frac{D}{D'}\left(\frac{r_{m+1}}{r_m} - \frac{r_{m+1}}{r_{m-1}\alpha_m}\right) + \frac{r_{m+1}}{r_m} . \qquad (47)$$

With α_{m+1} thus fixed in terms of α_m, Equation 46 was solved for β_m in terms of β_{m-1}.

$$\beta_m = \frac{D(\beta_{m-1})(r_{m+1})}{D'\alpha_m (r_{m-1})} . \qquad (48)$$

To summarize, the three equations used for evaluation of the neutron flux were the following.

$$\Phi_n = \frac{1}{\alpha_{n+1}} \left[\Phi_{n+1} + \beta_n \right]. \tag{49}$$

$$\beta_n = \frac{\beta_{n-1}}{\alpha_n} + c_n . \tag{50}$$

$$\alpha_{n+1} = b - \frac{1}{\alpha_n} . \tag{51}$$

The following four equations resulted from the boundary conditions of the problem.

$$\alpha_1 = \infty . \tag{52}$$

$$\Phi = 0 \text{ at the reactor boundary.} \tag{53}$$

$$\alpha_{m+1} = \frac{D}{D'} \left[\frac{r_{m+1}}{r_m} - \frac{r_{m+1}}{\alpha_m (r_{m-1})} \right] + \frac{r_{m+1}}{r_m} . \tag{54}$$

$$\beta_m = \frac{D (\beta_{m-1})(r_m + 1)}{D' \alpha_m (r_{m-1})} . \tag{55}$$

D. Effects of Higher Isotopes on Thermal Reactor Fuel Loadings

Although the primary emphasis of this investigation was on the effects of the higher isotopes of plutonium on a fast reactor, a short analysis of effects on thermal reactors proved useful for purposes of comparison. A thermal reactor normally operates with considerable excess reactivity to allow for xenon override, fuel burnup, fission product poisoning, temperature control, etc. Benedict and Pigford (2, p. 100) calculated this excess reactivity in terms of poisons, higher plutonium isotopes, and

absorption of fission product poisoning. Their equation can be considerably simplified for the case of initial excess reactivity if the uranium concentration is assumed equal to the reference design. Of course, no fission products are assumed to be initially present. Subject to these restrictions, the excess reactivity was calculated using Equation 56.

$$\bar{\Delta} = \frac{N_{49}\,\sigma_{49}\,(\eta_{49}\,\varepsilon\,p_{th}\,p-1)}{N_{25^*}\,\sigma_{25^*}} + \frac{N_{41}\,\sigma_{41}\,(\eta_{41}\,\varepsilon\,p_{th}\,p-1)}{N_{25^*}\,\sigma_{25^*}}$$

$$- \frac{N_{40}\,\sigma_{40}}{N_{25^*}\sigma_{25^*}} - \frac{N_{42}\,\sigma_{42}}{N_{25^*}\sigma_{25^*}} \quad . \qquad (56)$$

The initial reference design was considered to have 1% enrichment and no plutonium. For this case the concentrations of the mixture of plutonium isotopes corresponding to a particular burnup were inserted into the equations and an excess reactivity was determined. Next the amount of pure Pu-239 that would give that same excess reactivity was determined. The ratio of Pu-239 concentrations indicated the effect of the higher isotopes on the Pu-239 required in the thermal reactor.

VI. COMPUTER SOLUTION OF PROBLEM

A. Analog Computer Solution

The set of equations describing the buildup of plutonium isotopes within the thermal reactor can be solved in a straight forward manner for the case of no plutonium recycle. However, the change of concentrations of the various isotopes becomes a discontinuous function of time at the time of refueling. This causes difficulties with respect to electronic analog solution. The following method was therefore employed.

A transformation of variables governed by the definition $\theta = (\varphi t/10^{21})$ was utilized. The values for σ_{28}, η_{25}, σ_{25}, ϵ, P_1, P, η_{49}, σ_{49}, η_{41}, σ_{41}, α_{49}, σ_{40}, σ_{41}, α_{41}, and σ_{42} given for the example of Benedict and Pigford (2, p. 76) were substituted in Equation 2, 3, 4, and 5, resulting in the equations shown below.

$$\frac{dN_{49}}{d\theta} = 1.64 \times 10^{-3} N_{28} + 1.56 \times 10^{-1} N_{25} + 5.98 \times 10^{-1} N_{41} - 1.06 N_{49}. \quad (57)$$

$$\frac{dN_{40}}{d\theta} = 6.05 \times 10^{-1} N_{49} - 2.25 N_{40}. \quad (58)$$

$$\frac{dN_{41}}{d\theta} = 2.25 N_{40} - 1.41 N_{41}. \quad (59)$$

$$\frac{dN_{42}}{d\theta} = 3.62 \times 10^{-1} N_{41} - 1.97 \times 10^{-2} N_{42}. \quad (60)$$

At this point the problem of discontinuity in the concentration

variations was resolved by assuming that the concentration of the plutonium isotopes per unit volume of fuel remained constant through refueling. However, due to the depletion of U-235 and to a lesser extent the depletion of U-238, the uranium concentrations have to be changed at refueling to insure reactivity. Thus an average (N_{25}/N_{28}) ratio over a single irradiation was calculated as shown in Equation 61.

$$\left(\frac{N_{25}}{N_{28}}\right)_{avg.} = \frac{1}{2}\left[\left(\frac{N_{25}}{N_{28}}\right)_{initial} + \left(\frac{N_{25}}{N_{28}}\right)_{final}\right] . \qquad (61)$$

The value of $(N_{25}/N_{28})_{final}$ depends, of course, upon the length of time and flux levels between refueling. For a low enrichment reactor, the buildup of plutonium isotopes was relatively independent of the exact value of $(N_{25}/N_{28})_{avg.}$ chosen. Thus the type of average chosen did not significantly affect the results. This technique permitted the calculation of buildup of plutonium isotopes over several refuelings to be treated as continuous. The N_{28} concentration was arbitrarily scaled at 1,000 volts, which resulted in convenient voltages to represent the plutonium isotopic concentration. The coefficients were set by means of self-compensating potentiometers on the Heathkit electronic analog computer. The wiring diagram employed is shown in Figure 1.

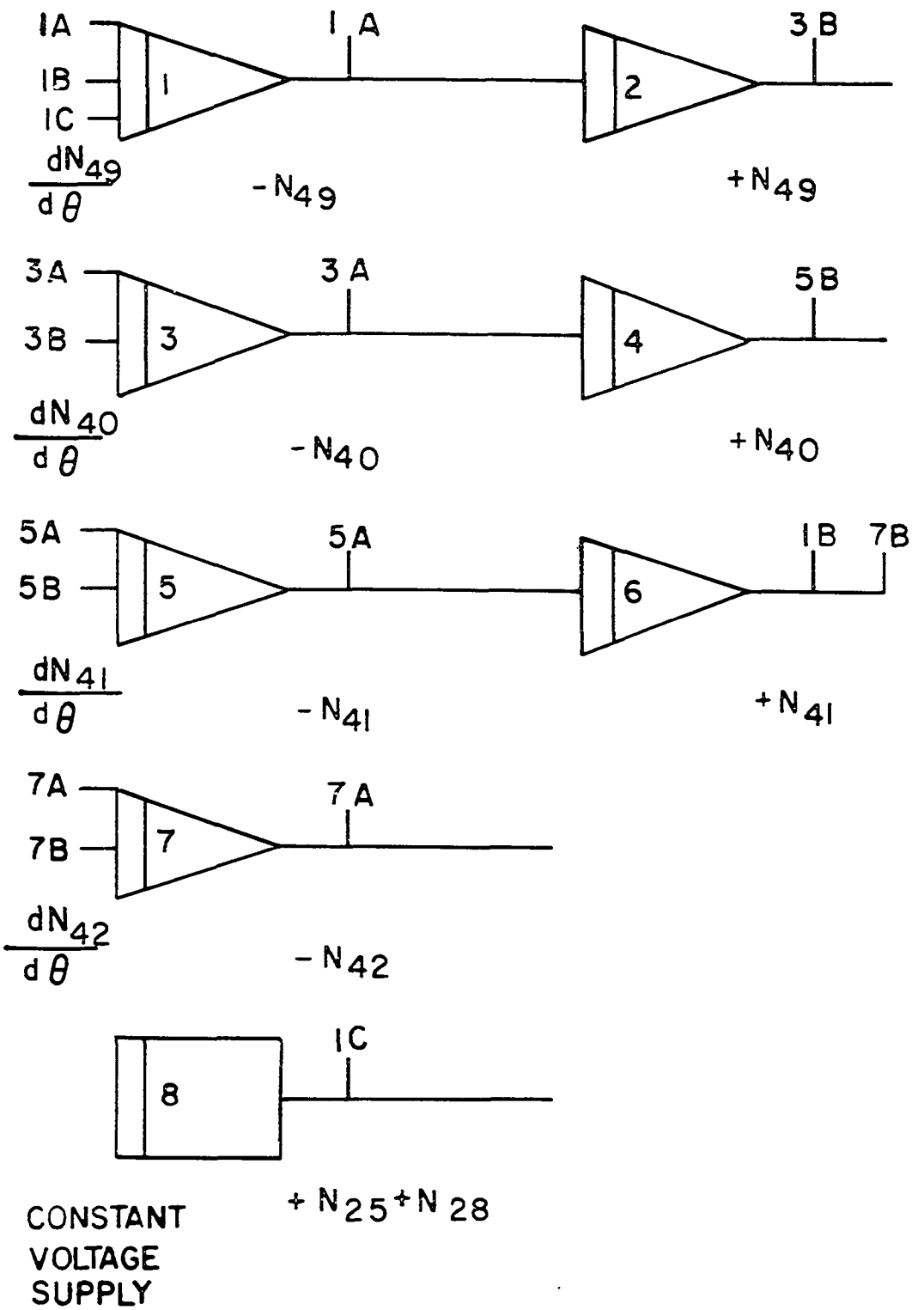

Figure 1. Wiring diagram for the analog computer

B. Digital Computer Solution

The problem was programmed for an IBM 650 Data Processing System by the use of the FORTRANSIT automatic coding system. The system restricted the data storage to less than one-half of the total 4,000 words of storage. Of these 2,000 words, 1,788 were used for data storage. The second 2,000 words of storage capacity contained the 1,033 words of program instruction plus 425 words of instruction from the program deck which contained the standard initial instructions for the object program. The FORTRANSIT program which is given in the Appendix, is in general compatible with any IBM FORTRAN System.

The computer program was designed to calculate the reactivity of a fast reactor system in a spherical geometry given the dimensions and fuel loadings. The fast reactor used as a model was similar to that of Hall (7). Cross section data used was that of Yiftah et al. (19). The reactor was of spherical geometry with two regions. The core had a radius of 31 centimeters and consisted in all cases of 15% Fe, 50% Na, and 35% fuel, which was a mixture of U-238 and plutonium. The blanket consisted of 70% U-238, 10% Fe, and 20% Na for all cases studied. The blanket radius was 75 centimeters, which differed from the assumption of an infinite blanket thickness by Hall (7).

The core and blanket dimensions were fed into the computer as fixed point constants. Then the following quantities were calculated.

1) The atoms/cm^3 of each element or isotope was calculated from the volume fraction of the various nuclides.

2) For each energy group, $\Sigma_{n,n'}\ j \longrightarrow j + k$ was calculated. This

is the cross section for neutrons transferred from group j to group j + k by inelastic scattering. The inelastic scattering cross section, $\Sigma_{n,n'}$, for case $j \longrightarrow j + 1$ was adjusted to include Σ_{er} which was the cross section for neutrons removed to the next lowest energy group by elastic scattering. Due to the high atomic weights of the elements considered to be present in the reactor, a neutron could not be scattered from group j to a group greater than j + 1 by elastic scattering. These cross sections were called CSELA and consisted of a 16x6 array CSELA (16,6) which included the probability for each group of being scattered to the next lowest six groups.

3) The quantity FCSBN was calculated. This was the product $(\Sigma_f)(\nu)$ for each group where Σ_f was the macroscopic fission cross section and ν was the average number of neutrons per fission for a reaction with a neutron in that specific energy group. There were 16 values of FCSBN.

4) The diffusion coefficient for each energy group was calculated from DELAN equals $1/3\ \Sigma_{tr}$ where Σ_{tr} was the transport cross section for that particular energy group.

5) The absorption cross sections, which were calculated for each energy group, included all removals from that group $\Sigma_a = \Sigma_f + \Sigma_{n,\gamma} + \Sigma_{er} + \Sigma_{n,n'}$, where $n \neq n$. Then Σ_a was used to calculate the constant BBLAN for each group, where BBLAN was defined as $2 + (\Sigma_a/D)$.

In addition to the above calculation, a similar set of values was computed for the reactor core. It was also necessary to estimate the values of FISOR, which was the relative number of fissions at each radius point. FISOR was normalized so that $1.0 = \sum_{IR=1}^{75}$ FISOR (IR). Also, the value of FISPE was required, which was the distribution of fission neutrons

between the various energy groups, stated by Yiftah et al. (19) to be identical for all fissionable nuclei.

Once the above data had been prepared on cards, the reactivity of the system could be calculated by use of the digital computer. The calculation was made in the following steps.

1) SORCE, a 16 x 75 or 1,200 word array, was calculated first. SORCE was a function of both radius point and energy. The asterisk indicates multiplication.

$$\text{SORCE(IE,IR)} = \text{FISOR(IR)} * \text{FISPE(IE)}.$$

Since both FISOR and FISPE were normalized, SORCE was also normalized as shown in Equation 62.

$$1.0 = \sum_{IE=1}^{16} \sum_{IR=1}^{75} \text{SORCE(IE,IR)}. \tag{62}$$

2) The neutron flux was now calculated at each point and for each energy group (1,200 values). The following equations were used.

$$\overset{I}{\Phi}_n = \frac{\overset{I}{\Phi}_{n+1} + \beta_n}{\alpha_{n+1}} . \tag{63}$$

$$\beta_n = \frac{\beta_{n-1}}{\alpha_n} + \frac{nS}{D} . \tag{64}$$

$$\alpha_{n+1} = b - \frac{1}{\alpha_n} . \tag{65}$$

For each energy group, the sequence of calculations was α, β, and $\overset{I}{\Phi}$. The flux was first calculated for the energy group I whose only source of

neutrons was that of fission, since there were no higher groups for scattering down.

3) For energy group I, the program began with the calculation of α. To make the flux finite at the center of the core, α_1 equals ∞. Thus α_2 equals b, α_3 equals b - $(1/\alpha_2)$, etc., where b is equal to $2 + (\Sigma_a/D)$ which was read in as input data. Once the core blanket boundary was reached, α_{m+1} was evaluated by Equation 66.

$$\alpha_{m+1} = \frac{DCORE}{DBLAN}\left[\frac{r_{m+1}}{r} - \frac{r_{m+1}}{(r_{m-1})(\alpha_m)}\right] + \frac{r_{m+1}}{r_m}. \quad (66)$$

The remaining values of α were calculated using BBLAN instead of BCORE.

4) Next, values of β were calculated. Since α_1 equals ∞, the value of β_1 equals $(1)S_1/D$, β_2 equals β_1/α_2 plus $(2)S_2/D$, etc. The calculation was continued to the core blanket boundary, at which point β_m was evaluated by Equation 67.

$$\beta_m = \frac{DCORE\ (\beta_{m-1})(r_m + 1)}{DBLAN\ (\alpha_m)(r_{m-1})}. \quad (67)$$

The remaining values of β were calculated using DBLAN instead of DCORE where m was the core blanket boundary radius point.

5) Next FLUXR was calculated. FLUXR is the product of the flux and R, e. g., FLUXR = $\Phi = \varphi r$. This quantity was calculated working from the outside of the reactor in, and the neutron flux at the boundary was set equal to zero as illustrated in Equation 68 and Equation 69.

$$FLUXR\ (IE,75) = 0\ . \quad (68)$$

$$\text{FLUXR (IE, 74)} = \frac{\text{FLUXR (IE, 75)} + \beta_{74}}{\alpha_{75}} . \qquad (69)$$

6) The flux was calculated at each point for the first energy group and was stored to be printed out at the end of the computation. It was also used to determine the number of neutrons that were scattered at a radius point to each of the next six lower energy groups at that radius point. This was done by multiplication of flux by the read-in arrays CSCOR and CSBLA. When the existing lower energy groups, which were initially due only to fission, had been adjusted to include scattering from energy group I, energy group II was calculated.

7) When all 16 energy groups had been calculated, a 16x75 array had been stored which gave the value of the flux in each energy group at each radius point. By use of these values of flux and the read-in values of FCSCN and FCSBN, which are products of fission cross sections and ν in each energy group, the number of fissions was calculated. This was the variable RECFS(IR) or the recomputed fission source. This was printed out together with the flux values at this point in the program.

The initially assumed fission source, FISOR, and the fission source calculated from the fluxes, RECFS, were now both available. The source which had been assumed, integrated over all space in the reactor, should equal the source calculated from fluxes, over all space in the reactor if the reactor was just critical. The total source, as shown in Equation 70, can be put into difference equation form as in Equation 71.

$$\text{Total Source} = \int_0^R 4\pi r^2 S_{(r)} dr . \qquad (70)$$

$$\text{Total Source} = \sum_{n=1}^{75} 4\pi r_n^2 S_n \Delta r . \qquad (71)$$

Thus, the assumed source is proportional to $\sum_{n=1}^{75} r_n^2$ FISOR(n) and the calculated source is proportional to $\sum_{n=1}^{75} r_n^2$ RECFS(n).

According to Murray (13, p. 290), the reactivity may be calculated by Equation 72.

$$\text{Reactivity} = \frac{S_{\text{total}}(\text{calculated})}{S_{\text{total}}\ (\text{assumed})} = \frac{\sum_{n=1}^{75} r_n^2 \ \text{RECFS(n)}}{\sum_{n=1}^{75} r_n^2 \ \text{FISOR(n)}} . \qquad (72)$$

VII. RESULTS AND CONCLUSIONS

The changes in the isotopic composition of the plutonium used as the initial fast reactor fuel loading resulted in significant changes in the Pu-239 critical mass as shown in Table 1. The concentration of Pu-239 in the fuel of a fast reactor which yielded a critical mass was reduced from 14.1% for the Pu-239 case to 7.43% for the isotopic mixture produced in a thermal reactor for flux times of 10^{22} cm^{-2}. By comparison, in a thermal reactor the critical masses of Pu-239 required for mixed isotopic composition were from 11% to 18% higher than the pure Pu-239 masses for irradiation flux times between $\theta = 1$ and $\theta = 10$.

Table 1. Critical Pu-239 concentration in fuel

θ, 10^{21} cm.$^{-2}$	% Pu-239 in fuel
0	14.1
1	11.1
5	8.57
10	7.43

The ratio of the critical mass of Pu-239 in an isotopic mixture to the critical mass of pure Pu-239 was plotted in Figure 2 for the isotopic mixtures produced by various flux times. The three curves plotted are the 16 neutron energy group digital computer solution, a one neutron energy group hand calculation for the fast reactor to serve for comparison purposes, and a hand calculation showing the effect in a thermal reactor.

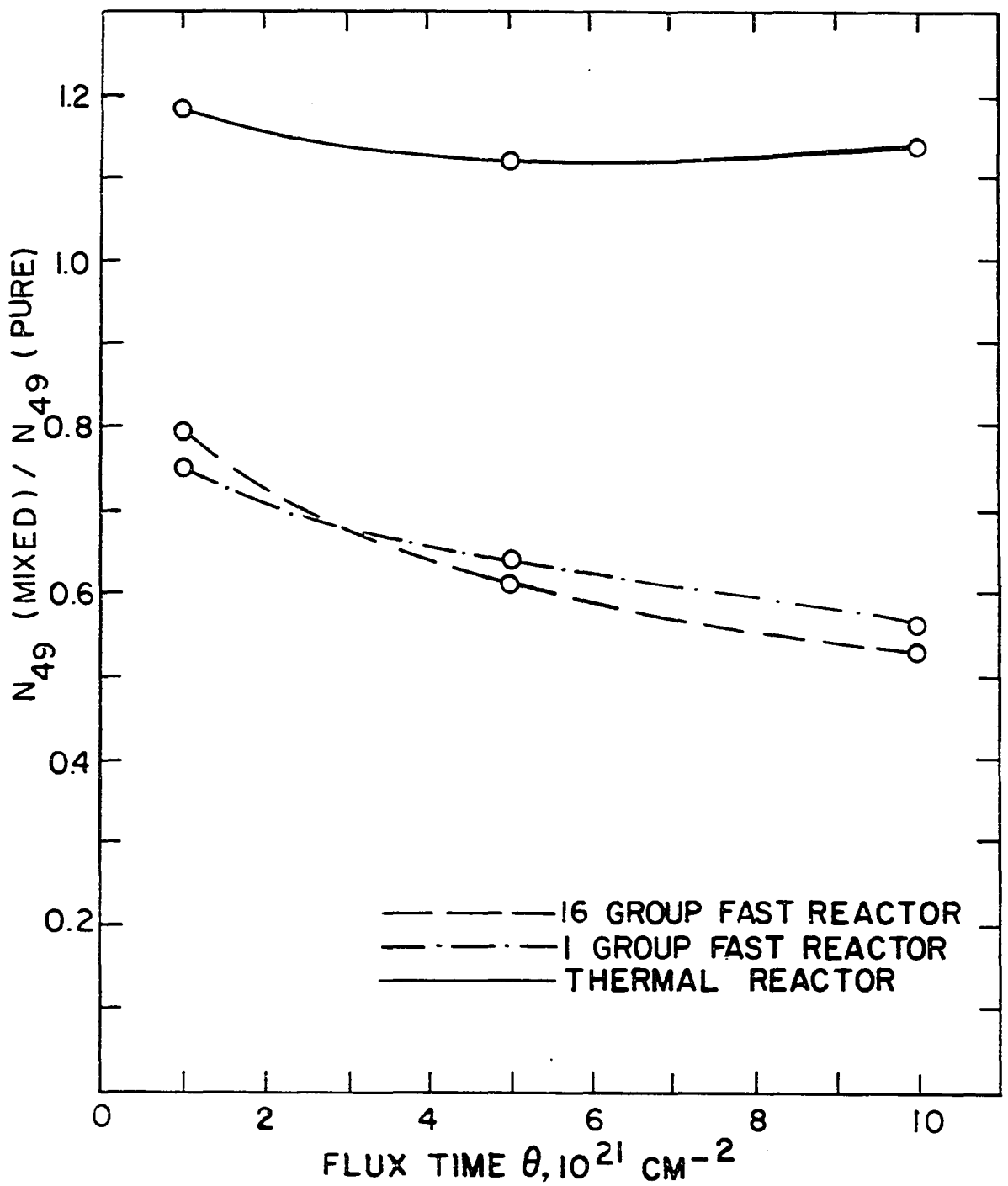

Figure 2. Ratio of the plutonium critical masses for various flux times

The mixed Pu-239 fast reactor critical mass was in all cases smaller than the pure Pu-239 fast reactor critical mass. This was due to the fact that Pu-240 and Pu-242 will fission in a fast neutron flux while these isotopes will not fission when the neutron energy is smaller than 0.11 MEV. The mixed Pu-239 critical mass was reduced to 79% of the pure Pu-239 critical mass for flux times as low as $\theta = 1$. For flux times as high as $\theta = 10$, the critical mass was reduced to 53% of the pure Pu-239 value. When these reductions were considered in terms of power reactors having critical masses in the neighborhood of 500 kilograms, the reduction in investment was considered to be significant. If the power reactor were to contain 500 kilograms of pure Pu-239 valued at $12 per gram, the investment would reach $6 million. The substitution of a mixture of plutonium isotopes irradiated to flux times of $\theta = 10$ containing 265 kilograms of Pu-239 could result in a considerable reduction in fuel investment. The actual price of Pu-239 when mixed with higher isotopes has not, of course, been determined. It would seem that the price of Pu-239 would be somewhat less than $12 per gram for this so-called "dirty" plutonium since the higher isotopes result in a reduction in reactivity in a thermal reactor. The increased fuel loading for the mixed plutonium isotopes in a thermal reactor is shown in Figure 2 for purposes of comparison with the fast reactor. The initial depression in the curve was due to the buildup of fissionable Pu-241, and the later rise was due to the continued buildup of non-fissionable Pu-242. The agreement in Figure 2 between the highly simplified one neutron energy group hand calculation and the 16 neutron energy group digital computer calculation may be somewhat fortuitous and may be due to the cancelling effects of the various assumptions made for

the one group calculation in this particular case. In other cases the one group calculation might not show such good agreement.

Table 2 shows the variation of reactivity with the percentage of Pu-239 in the fuel for $\theta = 0$, $\theta = 1$, $\theta = 5$, and $\theta = 10$ as calculated with the digital computer.

Table 2. Reactivity of a fast reactor for various plutonium compositions and concentrations

% Pu-239 in fuel	θ (10^{21}cm^{-2})	Reactivity
12.0	0	0.862
14.0	0	0.996
17.0	0	1.267
10.8	1	0.964
11.2	1	1.019
8.94	5	1.031
9.42	5	1.073
0.0760	10	1.012
0.0834	10	1.064

The percentage of Pu-239 which gave a "just critical" reactor (reactivity equals 1.000) was determined by the assumption of a linear relationship between reactivity and percentage of Pu-239 in the fuel. For the case of $\theta = 0$, the reactivity was essentially a linear function of Pu-239 concentrations within the range of Pu-239 concentrations from 12% to 17% of the fuel for the three plutonium concentrations calculated.

reactivity varied from 0.862 to 1.267. This linearity simplified interpolations to find the critical mass. The interpolated concentrations were in all cases within 4% of a value of the plutonium concentration for which a digital computer calculation was made. The results of $\theta = 0$ were compared to the calculations of Hall (7). Hall obtained criticality with a Pu-239 concentration of 17% in the fuel. The calculated Pu-239 concentration of 14.1% of the fuel to achieve criticality was in reasonable agreement with Hall when the difference in cross sections used was noted. Yiftah et al. (19) stated that the set of cross section data he computed tended to give somewhat higher reactivities than the earlier sets of assumed cross sections, thus leading to a somewhat lower critical mass.

The flux plots shown in Figures 3 and 4 indicate the variation in neutron flux with position for the various neutron energy groups. The value of the flux increased for energy groups I to VI and decreased for groups VII to XVI. This agreed with the statement of Murray (13, p. 69), who predicted flux peaking at about 0.3 MEV for EBR-type reactors. The changes in the flux spectra due to the introduction of the higher isotopes of plutonium were negligible. The changes in the amount of flux in any particular energy group due to changes in plutonium isotopic concentration in the core were less than 6% within the range $\theta = 1$ to $\theta = 10$. No particular pattern was noted in these small changes in the distribution of flux within the various energy groups. The rapid decrease with distance of the neutron flux within the blanket is shown in Figure 3 and 4. This decrease in neutron flux supported the assumption that a 75 centimeter blanket is essentially infinite. The 75 centimeter blanket was chosen in accordance with the criteria of Murray (13, p. 72) that a blanket

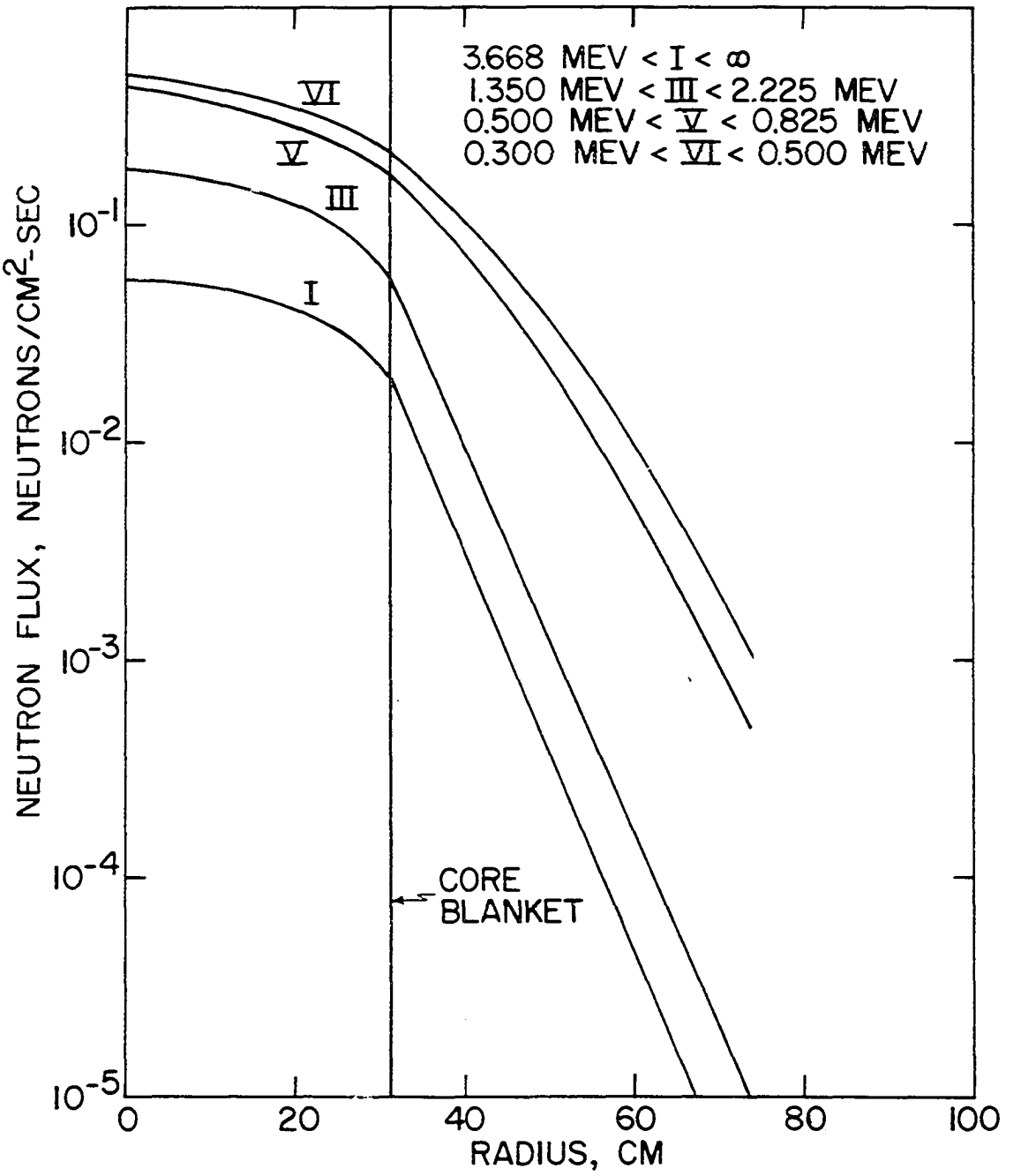

Figure 3. Neutron flux in the reactor for energy groups I through VI

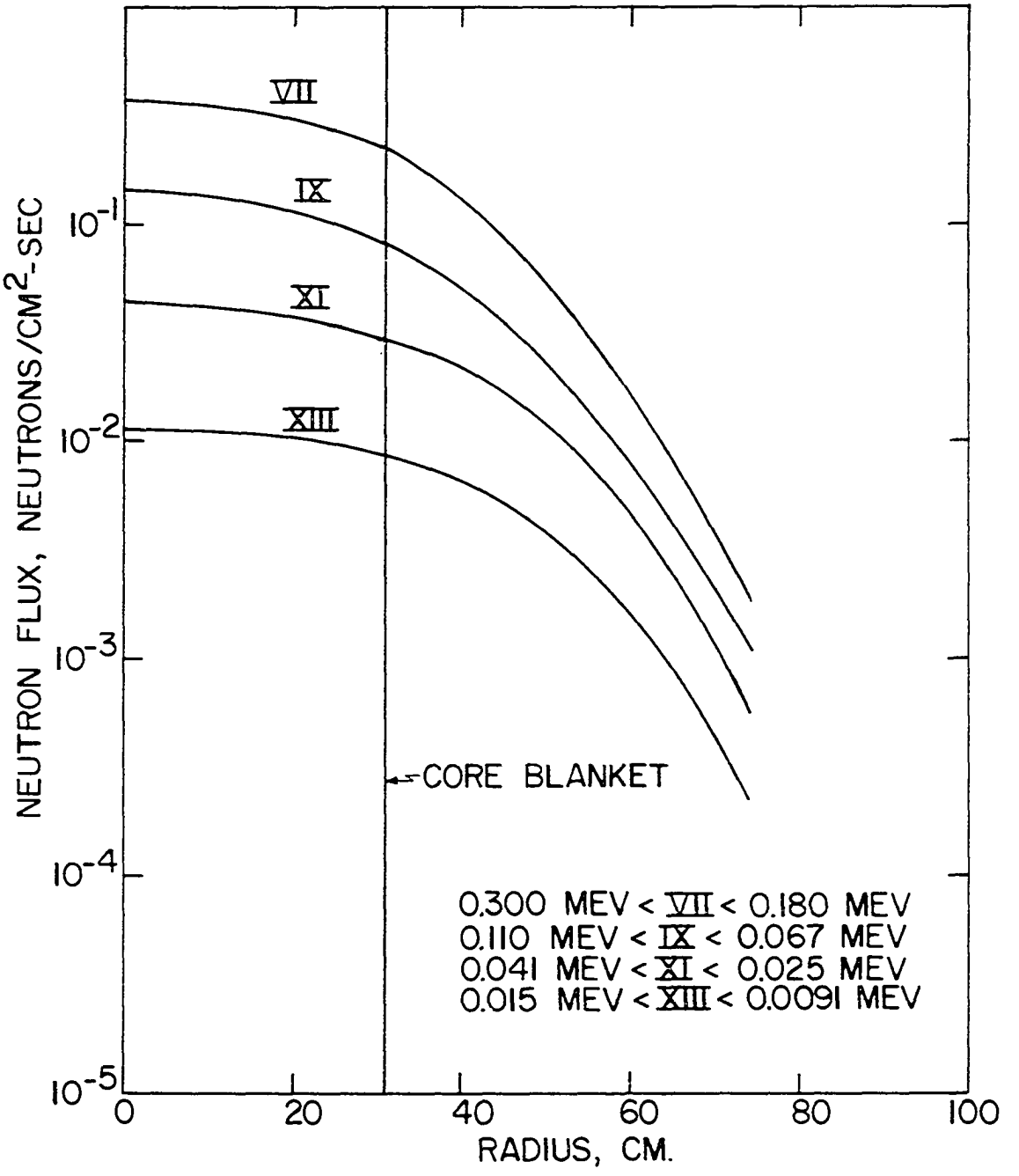

Figure 4. Neutron flux in the reactor for energy groups VII through XIII

thickness of $T' = 2.65\ L_r$ was comparable to an infinite blanket.

The changes in the plutonium isotopic composition as a function of flux times of irradiation were determined by use of the Heathkit analog computer to solve the set of differential equations describing the buildup of the higher plutonium isotopes in a thermal reactor. The nuclear parameters of the thermal reactor chosen were those used by Benedict and Pigford (2, p. 76). Figure 5 shows plots of the change in concentrations in units of atoms/atom U-238 as a function of the flux time θ, $10^{21} cm^{-2}$. The plots are for various values of R, which was defined as the average ratio of N_{25}/N_{28} during irradiation. The results of the analog computation shown in Figure 5 were used to obtain the isotopic plutonium composition of the plutonium used as the fast reactor fuel. A ratio R, average N_{25}/N_{28} during irradiation, of 1.0 was chosen. Plutonium composition at flux times of $\theta = 1$, $\theta = 5$, and $\theta = 10$ is shown in Table 3.

Table 3. Isotopic plutonium at various irradiation flux times

Nuclide	$\theta = 0$	$\theta = 1$	$\theta = 5$	$\theta = 10$
Pu-239	1.000	0.700	0.496	0.362
Pu-240	-----	0.140	0.147	0.107
Pu-241	-----	0.160	0.225	0.168
Pu-242	-----	-----	0.132	0.362
	-----	-----	-----	-----
	1.000	1.000	1.000	1.000

The analog computer results shown in Figure 5 agreed with the computa-

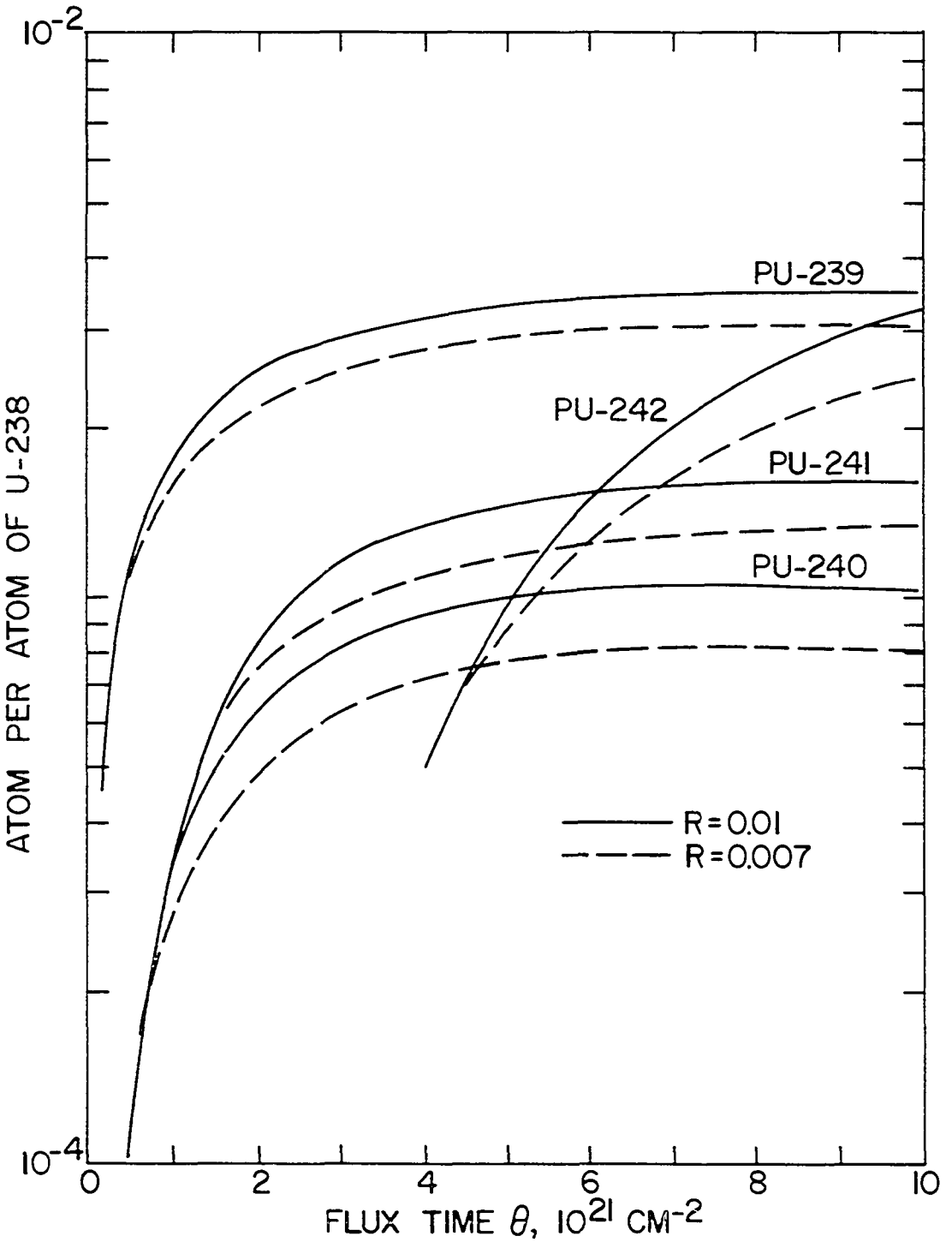

Figure 5. Plutonium buildup in the thermal reactor

tions of Benedict and Pigford (2, p. 111) for the conditions of low flux times of irradiation. For this case the assumptions made for the analytical solution by Benedict and Pigford were valid. For higher flux times of irradiation, the concentrations of Pu-239, Pu-240 and Pu-241 reached the equilibrium values calculated by Benedict and Pigford. The flux times studied were not large enough for Pu-242 to come to equilibrium. Thus the electronic analog computer solution was in agreement with the results of Benedict and Pigford for the two limiting conditions that they calculated analytically.

The digital computer program which was developed could be used to calculate the critical masses of a wide variety of one dimensional spherical fast reactors. No restriction, other than available computer storage, was placed on the physical dimensions of the core and blanket. The program is completely flexible in choice of materials, allowing for the use of any desired mixture of reactor fuel, coolant and structural materials. While the program was written for a two region reactor, it can easily be adapted for the one region case. Although the program was written in the IBM 650 FORTRANSIT language, it is in general compatible with any FORTRAN system, including IBM 650 FORTRAN, IBM 1620 FORTRAN, IBM BASIC 7070/7074 FORTRAN, IBM 7070/7074 FORTRAN, IBM 705 FORTRAN, IBM 704 FORTRAN, and IBM 709/7090 FORTRAN. Minor adaptations must be made, of course, depending upon the specific card or tape input-output used.

VIII. LITERATURE CITED

1. Andriot, J. and Gaussens, J. Fuel cycles, the production of uranium power plant program, and relevant investments. Second International Conf. on the Peaceful Uses of Atomic Energy Proc. 13: 165-183. 1958.

2. Benedict, Manson and Pigford, Thomas H. Nuclear chemical engineering. New York, N. Y., McGraw-Hill Book Co., Inc. 1957.

3. Carlson, B. J. and Bell, G. I. Solution of the transport equation by S_n method. Second Internatl. Conf. on the Peaceful Uses of Atomic Energy Proc. 16: 535-547. 1958.

4. Dayal, M., Parajpe, S. R., Prasad, N. B., and Singh, B. Study of fuel cycle with reference to a power programme. Second Internatl. Conf. on the Peaceful Uses of Atomic Energy Proc. 13: 184-188. 1958.

5. Ehrlich, R. and Hurwitz, H. Multigroup methods for neutron diffusion problems. Nucleonics 12, No. 2: 23-30. Feb. 1954.

6. Greebler, P., Harker, W. H., Harriman, J. M., and Zebroski, E. L. Recycle of plutonium in low enrichment light water reactors. Second Internatl. Conf. on the Peaceful Uses of Atomic Energy Proc. 13: 251-264. 1958.

7. Hall, D. B. Plutonium fuel for fast reactors. Second Internatl. Conf. on the Peaceful Uses of Atomic Energy Proc. 13: 300-306. 1958.

8. Hansen, G. E. Properties of elementary fast-neutron critical assemblies. Second Internatl. Conf. on the Peaceful Uses of Atomic Energy Proc. 12: 84-88. 1958.

9. Kazachkovsky, O. D. The economics of nuclear fuel for fast power reactors. Second Internatl. Conf. on the Peaceful Uses of Atomic Energy Proc. 13: 307-313. 1958.

10. Kiehn, R. M. Breeding: internal or external. U. S. Atomic Energy Commission Report ANL-6122: 101-113. [Argonne National Lab., Lemont, Ill.] 1959.

11. Loewenstein, W. B. and Okrent, D. The physics of fast power reactors; a status report. Second Internatl. Conf. on the Peaceful Uses of Atomic Energy Proc. 12: 16-37. 1958.

12. Long, J. K., Loewenstein, W. B., Branyan, C. E., Brunson, G. S., Kirn, F. S., Okrent, D., Rice, R. E., and Thalgott, F. W. Fast neutron power reactor studies with ZPR-III. Second Internatl. Conf. on the Peaceful Uses of Atomic Energy Proc. 12: 119-141. 1958.

13. Murray, Raymond L. Nuclear reactor physics. Englewood Cliffs, N. J., Prentice-Hall, Inc. 1957.

14. Okrent, D. The sensitivity of breeding ratio in fast reactors to uncertainties in cross sections. U. S. Atomic Energy Commission Report ANL-6122: 93-100. [Argonne National Lab., Lemont, Ill.] 1959.

15. Roach, W. H. A computational survey of idealized fast breeder reactors. U. S. Atomic Energy Commission Report ANL-6122: 77-87. [Argonne National Lab., Lemont, Ill.] 1959.

16. Scarborough, James B. Numerical mathematical analysis. Baltimore, Md., The John Hopkins Press. 1950.

17. Shanstrom, R. T., Benedict, M., and McDaniel, C. T. Fuel cycles in nuclear reactors. U. S. Atomic Energy Commission Report AECD-3595. [Technical Information Service Extension, AEC.] 1954.

18. Starr, C. Fuel enrichment and reactor performance. Second Internatl. Conf. on the Peaceful Uses of Atomic Energy Proc. 13: 282-290. 1958.

19. Yiftah, S., Okrent, D., and Moldauer, P. A. Fast reactor cross sections. New York, N. Y., Pergamon Press. 1960.

IX. ACKNOWLEDGEMENTS

The author wishes to thank Dr. Morton Smutz for his able guidance and helpful encouragement in this project. Thanks are also due to Dr. Glenn Murphy for his encouragement of the initial formulation of the problem and his continued interest throughout the past four years.

This thesis has been completed under a fellowship granted by the Atomic Energy Commission. Their financial support has made full-time academic work possible and is sincerely appreciated.

The use of the IBM 650 Data Processing System was made possible through a grant for unsponsored research by the Iowa State University Computing Center.

X. APPENDIX

A. Fortransit Program

```
00100  DIMENSION SORCE (16, 75), FISOR (75), FISPE (16), ALPHA (75),
       BCORE (16), BBLAN (16), DCORE (16), BETA (75)
00101  DIMENSION FLUXR (16,75), RECFS (75), FCSCN (16) CSCOR (16, 6),
       CSBLA (16, 6), DBLAN (16)
00102  EQUIVALENCE (FLUXR, SORCE)
00103  READ, FISOR
00104  READ, FISPE
00105  READ, IBLAN, ICORE
00106  READ, BBLAN
00107  READ, DCORE
00108  READ, CSCOR
00109  READ, CSBLA
00110  READ, FCSCN
00111  READ, FCSBN
00112  READ, DBLAN
00113  READ, BCORE
00023  CONTINUE
00114  DO 1 IE = 1, 16
00115  DO 2 IR = 1, IBLAN
00116  SORCE (IE, IR) = FISOR (IR)*FISPE (IE)
00002  CONTINUE
00001  CONTINUE
00000  PUNCH, SORCE
```

```
00117  DO 3 IE = 1,16
00119  IR = 2
00120 ALPHA (2) = BCORE(IE)
00004  ALPHA(IR + 1) = BCORE(IE) - (1/ALPHA(IR))
00121  IR = IR + 1
00122  IF(IR - ICORE)   4,123,12.
00123  CORE = ICORE
00124  A1 = (CORE + 1.0)/CORE
00205  A2 = (CORE + 1.0)/(CORE-1.0)
00206  ALPHA(ICORE + 1) = (DCORE(IE)/DELAN(IE)) * (A1 - (A2/ALPHA(ICORE)))
00207  ALPHA(ICORE + 1) = ALPHA(ICORE + 1) + A1
00201  IF(ALPHA(ICORE + 1) - 1.0)  202, 203, 203
00202  ALPHA(ICORE + 1) = 1.0
00203  IR = IR + 1
00006  ALPHA(IR + 1) = BELAN(IE) - (1/ALPHA(IR))
00125  IR = IR + 1
00126  IF(IR-IBLAN)  6, 7, 7
00007  CONTINUE
00127  BETA(1) = + SORCE(IE, 1)/DCORE(IE)
00128  IR = 2
00008  BETA(IR) = (BETA(IR-1)/ALPHA(IR)) + ((IR * SORCE (IE, IR))/
       DCORE(IE))
00129  IR = IR + 1
00130  IF(IR-ICORE)  8, 9, 9
00009  BETA(ICORE) = (DCORE(IE) * BETA(IR - 1) * (ICORE + 1))/
       ((ICORE - 1) * ALPHA(ICORE) * DELAN(IE))
```

```
00135  IR = ICORE + 1
00010  BETA(IR) = (BETA(IR - 1)/ALPHA(IR)) + ((IR * SORCE(IE, IR))
       /DELAN(IE))
00136  IR = IR + 1
00137  IF(IR - IELAN)  10, 11, 11
00011  CONTINUE
00138  FLUXR   (IE, IELAN) = 0
00139  IR = IELAN - 1
00012  FLUXR(IE,IR) = ((1/ALPHA(IR + 1)) * (FLUXR(IE, IR + 1) +
       BETA(IR)))
00140  IR = IR - 1
00141  IF(IR)   13, 13, 12
00013  CONTINUE
00000  PUNCH, ALPHA, BETA, FLUXR
00142  DO 14   IR = 1, ICORE
00143  INCR = 1
00015  IGRUP = IE + INCR
00144  SORCE(IGRUP, IR) = SORCE(IGRUP, IR) + ((FLUXR(IE, IR) *
       CSCOR(IE, INCR))/IR)
00145  INCR = INCR + 1
00146  IF(INCR - 6)   15, 15, 14
00014  CONTINUE
00147  ICORP = ICORE + 1
00148  DO 16 IR = ICORP, IELAN
00149  INCR = 1
00017  IGRUP = IE + INCR
```

```
00150  SORCE(IGRUP, IR) = SORCE(IGRUP, IR) + ((FLUXR(IE, IR)
       * CSELA(IE, INCR))/IR)
00151  INCR = INCR + 1
00152  IF(INCR-6)   17, 17, 16
00016  CONTINUE
00000  PUNCH, SORCE
00200  PAUSE
00003  CONTINUE
00153  DO 18 IR = 1, ICORE
00154  RECFS (IR) = 0
00155  DO 19 IE = 1,16
00156  RECFS(IR) = RECFS(IR) + ((FCSCN(IE) * FLUXR(IE, IR))/IR)
00019  CONTINUE
00018  CONTINUE
00157  ICORP = ICORE + 1
00158  DO 20 IR = ICORP, IELAN
00159  RECFS(IR) = 0
00160  DO 21 IE = 1,16
00161  RECFS(IR) = RECFS(IR) + ((FCSEN(IE) * FLUXR(IE, IR))/IR)
00021  CONTINUE
00020  CONTINUE
00024  PUNCH FLUXR
00025  PUNCH RECFS
00169  END
```