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SELF-SUSTAINING PLUTONIUM RECYCLE IN SODIUM GRAPHITE REACTORS

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I. INTRODUCTION

Plutonium is formed in any reactor fueled with uranium by the capture of neutrons in uranium-238. If the fuel is predominantly uranium-238, as in natural or slightly enriched uranium, the quantity of plutonium formed plays a significant role in the fuel cycle of the reactor. Although some of this plutonium is fissioned in situ, much of it will be in the fuel at the time of discharge from the core. The recovery of this plutonium and its reuse as fuel in the same reactor is referred to as plutonium recycle.

The incentives for recycling plutonium vary somewhat with the reactor in question. In the case of natural uranium reactors the plutonium can extend the reactivity lifetime of the fuel. A study of this subject was made recently for the case of a gas-cooled graphite reactor.[1] In reactors requiring slightly enriched uranium, the recycled plutonium lowers the consumption and inventory of uranium-235. Pigford and others[2] recently made an investigation of this subject for the case of a light-water-moderated reactor.

A special case of plutonium recycle may arise with slightly enriched uranium reactors in which the reactivity effect of the recycled plutonium is sufficient to reduce the required uranium-235 concentration in the feed material to that of natural uranium. This case may be referred to as self-sustaining plutonium recycle; a reactor which initially requires a source of reactivity above that supplied by natural uranium can, once steady-state plutonium recycle is established, operate with natural uranium as its only raw fuel material. In situations where a dependence on uranium enrichment may be undesirable, plutonium recycle offers the possibility of extending the range of acceptable reactor designs.

The present study was undertaken to determine whether a sodium graphite reactor (SGR) could be designed to achieve self-sustaining plutonium recycle. Studies of this subject have been presented by Roderick[3] for the case of an SGR of the Sodium Reactor Experiment (SRE) design, and by Benedict and Pigford.[4] The current design[5] of the SGR under development by Atomics International, a Division of North American Aviation, Inc., makes use of stainless steel for fuel and moderator cladding. It requires a fuel enrichment of about 3% uranium-235.

The reactivity requirements of such a reactor are too high for self-sustaining plutonium recycle to be achieved, because eta for the equilibrium plutonium is less than is required for the fuel. However, certain design modifications and the development of cladding materials of lower neutron absorption offer good promise of reducing SGR fuel reactivity requirements. The purpose of the present study was to determine whether, in the light of these probable advances, self-sustaining plutonium recycle could be achieved in a SGR. This effort was limited in scope to the technical feasibility, although certain economic factors such as fuel burnup are also considered.

II. DISCUSSION

The establishment of self-sustaining plutonium recycle in a given reactor depends on: (1) the reactivity of plutonium in the neutron spectrum of that reactor, (2) the concentration of plutonium which can be maintained in the fuel, and (3) the criticality requirements of the reactor.

A. REACTIVITY OF PLUTONIUM

The simplest index of the reactivity of a fuel material is η*, the number of neutrons produced per absorption in the fuel. Because plutonium is a mixture of isotopes, its nuclear characteristics are a function of its isotopic composition. For example,

$$\eta_{Pu} = \frac{N_9\sigma_9\eta_9 + N_1\sigma_1\eta_1}{N_9\sigma_9 + N_0\sigma_0 + N_1\sigma_1 + N_2\sigma_2} .$$

The concentration of each isotope, neglecting processing losses, is obtained by the solution of the typical isotope buildup differential equations,[4]

$$\frac{dN_9}{d(\phi t)} = \dot{N}_9 = R_c - N_9\sigma_9$$

$$\dot{N}_0 = N_9\sigma_9^c - N_0\sigma_0$$

$$\dot{N}_1 = N_0\sigma_0 - N_1\sigma_1$$

$$\dot{N}_2 = N_1\sigma_1^c - N_2\sigma_2 .$$

The term R_c expresses rate of conversion. While this rate is not a constant with irradiation, such an assumption does not lead to serious errors. The cross sections used in these equations are effective values which express the relative rates of the various neutron reactions in the total spectrum of the reactor. The estimation and use of these cross-sections has been discussed.[6]

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*See Appendix F for a table of nomenclature.

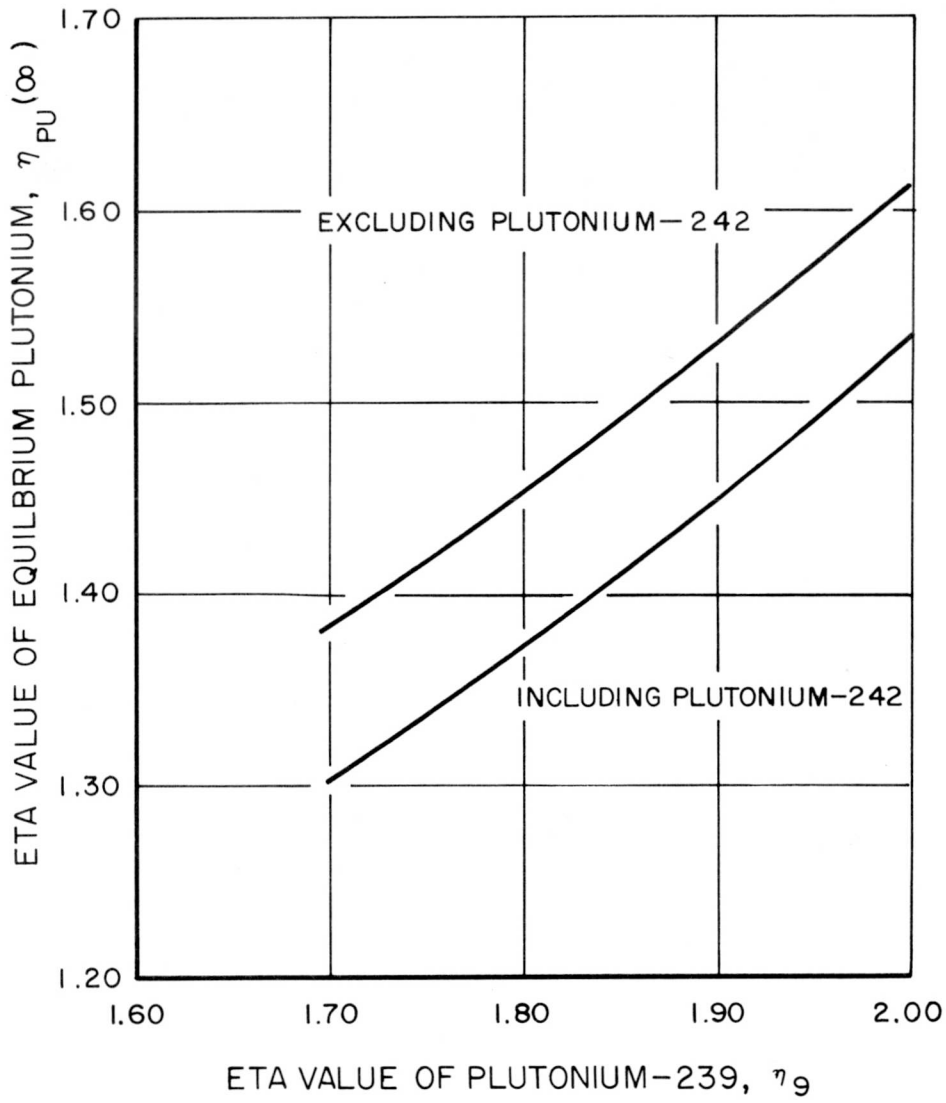

Figure 1. Eta Value of Equilibrium Mixture of Plutonium Isotopes

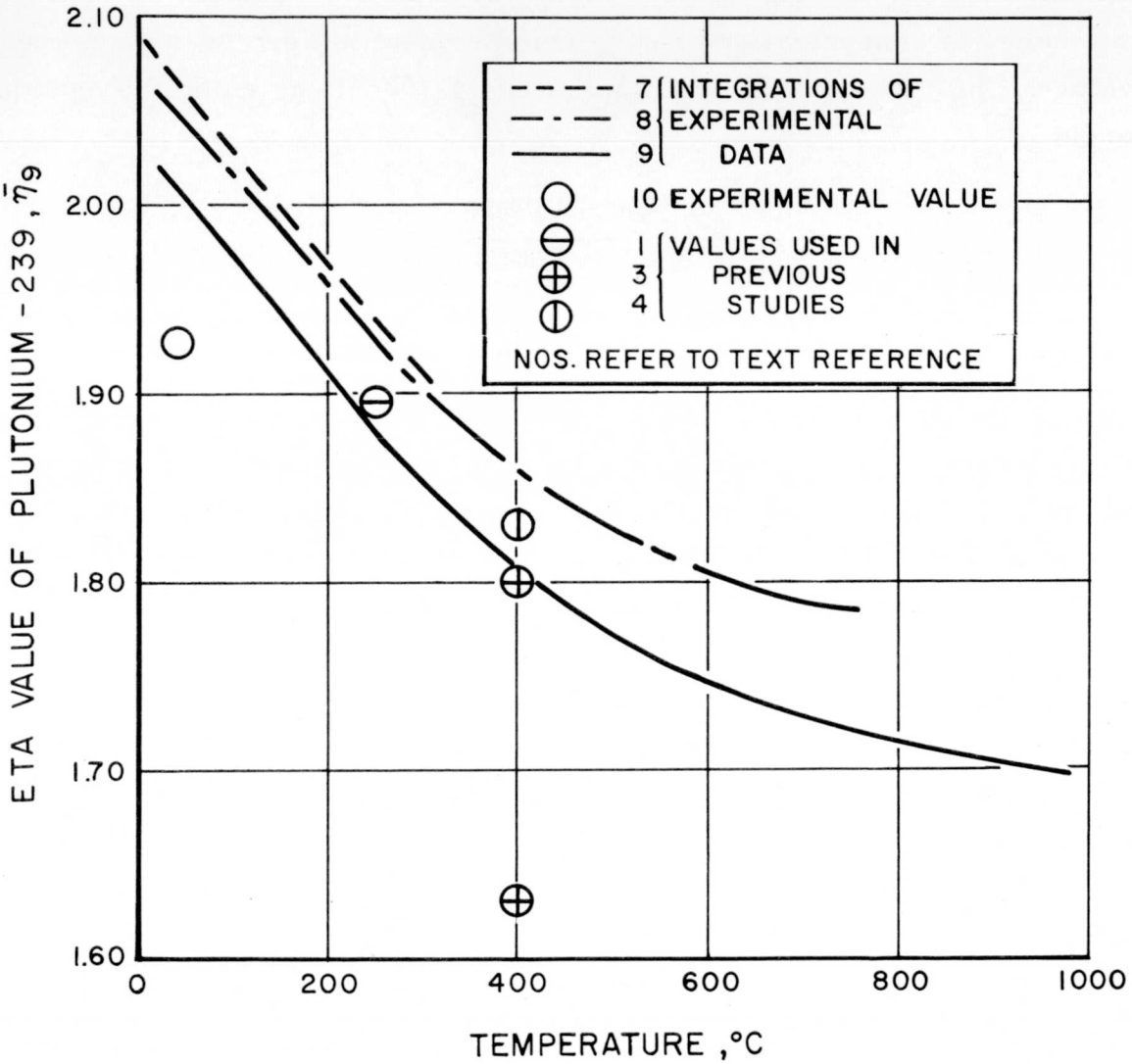

Figure 2. Maxwell-Averaged Eta Value for Plutonium-239

It is apparent that the value of η_{Pu} will be a function of the irradiation of the plutonium and also of the neutron spectrum. One idealized situation which can be easily calculated and which is useful in establishing limits of self-sustaining plutonium recycle is the case of equilibrium plutonium. Equilibrium plutonium is the isotope combination that would eventually be reached if the conversion rate is maintained constant. The isotopic concentration is calculated by setting the rate of change of concentration of each isotope equal to zero in the above equations. The value of η_{Pu} for equilibrium plutonium, $\eta_{Pu}^{(\infty)}$ is given by the equation (Appendix A)

$$\eta_{Pu}^{(\infty)} = \frac{2.00 + 0.313\eta_9}{3.27 - 0.780\eta_9} .$$

This value is plotted as a function of η_9 in Figure 1.

One of the principal differences between the composition of the idealized "equilibrium plutonium" and plutonium in an actual fuel cycle is that in the latter case plutonium-242 will not reach the concentration as indicated by $N_2\sigma_2 = N_1\sigma_1^c$ because of processing losses. The cross section of plutonium-242 is relatively low and, as a result, the equilibrium value of N_2 is high and the burnout per cycle is small. In such a case processing losses constitute the important loss of plutonium-242. Consequently, this isotope would reach an effective equilibrium concentration significantly below the ideal case. In order to show the limiting effect of this phenomenon on the reactivity of plutonium, the value of $\eta_{Pu}^{(\infty)}$ with a zero concentration of plutonium-242 has been calculated and is plotted in Figure 1.

It is apparent from Figure 1 that the reactivity of plutonium is a sensitive function of the value of η_9. The value of η_9 is a function of neutron temperature (and, therefore, moderator temperature) as shown in Figure 2. The lines drawn represent integrated average values for a Maxwellian distribution of neutrons of the given temperature. The line of Skarsgard and Kenward[7] represents averages using their own experimental values of $\eta_9(E)$. The other two lines[8, 9] were obtained by averaging over an assortment of experimental data weighted according to the judgment of the individual authors. The single experimental point of

Gaerttner, et al.[10] was obtained by measuring the reactivity effect on a thermal test reactor. The discrepancy between this value and other experimental measurements on plutonium-239 is not easily explained. It does illustrate the tentative nature of the data. As a matter of interest, the values of η_9 used in other studies of plutonium recycle[1, 3, 4] are shown.

The actual neutron spectrum in a SGR is expected to deviate from a Maxwellian.[1] However, orienting calculations indicated that for the lattices of interest in this study, deviations from a Maxwellian spectrum had a negligible effect on the value of η_9. Westcott's compilation[8] also indicates that η_9 is insensitive to the epithermal fraction of the neutron population in the temperature range of interest. For these reasons it appears that the effective value of η_9 in a reactor may be assumed to be equal to the Maxwell-averaged value.

B. EQUILIBRIUM CONCENTRATION OF PLUTONIUM IN FUEL

Relations for the equilibrium concentration of the plutonium isotopes under conditions of plutonium recycle, have been derived.[4] For plutonium-239 the equation is

$$N_9(\phi t) = \frac{N_8\bar{\sigma}_8 + N_1\sigma_1\eta_1 P}{\gamma} + \left(\frac{N_5^0\sigma_5\eta_5 P}{\gamma - \sigma_5}\right)\left(\frac{e^{-\sigma_5\phi t} - e^{-\gamma\phi t}}{1 - e^{-\gamma\phi t}}\right)$$

$$\gamma = \sigma_9(1 - \eta_9 P),$$

where ϕt is the irradiation per cycle. The term P is a lattice parameter which expresses the number of resonance captures in uranium-238 per fission neutron. In a lattice with only uranium fuel, P may be expressed in terms of the more customary enrichment, ϵ, and initial conversion ratio, ICR,

$$P = \frac{ICR - \dfrac{100 - \epsilon}{\epsilon}\dfrac{\bar{\sigma}_8}{\bar{\sigma}_5}}{\eta_5}.$$

Since these two parameters, ϵ and ICR, are used extensively in the following pages, it may be well to stress that they refer to a given lattice when fueled with

uranium only. The variation in equilibrium plutonium-239 concentration as a function of ϵ and ICR is shown in Figure 3. These values are for the limiting case of zero irradiation ($\phi t = 0$) per cycle. Figure 4 indicates the variation of plutonium concentration with irradiation per cycle, expressed as burnup of uranium-235 in natural uranium, for a specified lattice. The values calculated are very nearly independent of the η_9 value. They are, of course, dependent on assumed values for certain nuclear constants; in the present case σ_8/σ_5 and σ_9/σ_5, for which values of 0.0040 and 4.0, respectively, were used. Numerous calculations have indicated, however, that the significant results of this type calculation are not sensitive to the particular values used for these constants. As a further simplification, it was assumed that the higher plutonium isotopes have the concentration in equilibrium plutonium calculated according to their buildup differential equations (Section A). Under these conditions the concentration of equilibrium plutonium is given by the equation (Appendix A)

$$N_{Pu}\sigma_{Pu} = N_9\sigma_9(3.27 - 0.780\eta_9).$$

C. SELF-SUSTAINING PLUTONIUM RECYCLE CONDITION

The principal reactivity effect of the addition of plutonium to uranium fuel is in the ηf product for the lattice. The requisite condition for self-sustaining plutonium recycle which is applied here is that the ηf product with natural uranium plus equilibrium plutonium equals or exceeds the ηf product with the original enriched uranium fuel. The equation

$$\eta f = \frac{N_5^0\sigma_5\eta_5 + N_{Pu}\sigma_{Pu}\eta_{Pu}}{\frac{1}{f_0}(N_5^0\sigma_5 + N_8\sigma_8) + N_{Pu}\sigma_{Pu}}$$

is a reasonable approximation in the present case (Appendix C). N_5^0 represents the concentration of uranium-235 in natural uranium and f_0 is the thermal utilization value in the same lattice with natural uranium fuel. The values of N_{Pu} and of $N_{Pu}\sigma_{Pu}$ were discussed in the previous sections. Figure 5 shows the results of the application of this criterion. The lines drawn there represent loci of conditions under which lattice will have an equal ηf product whether fueled

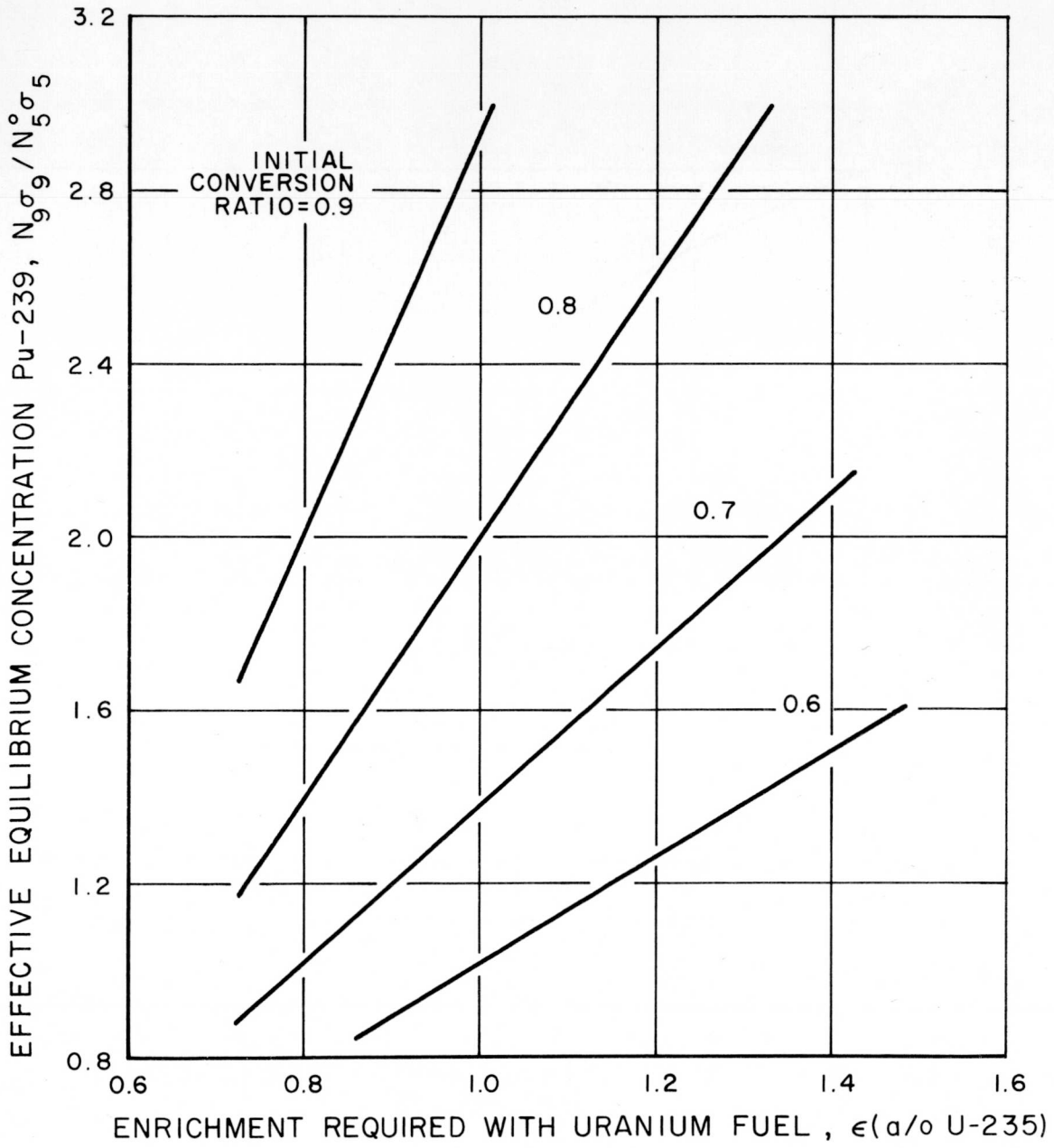

Figure 3. Equilibrium Concentration of Recycled Plutonium-239 in Fuel (Zero Irradiation per Cycle)

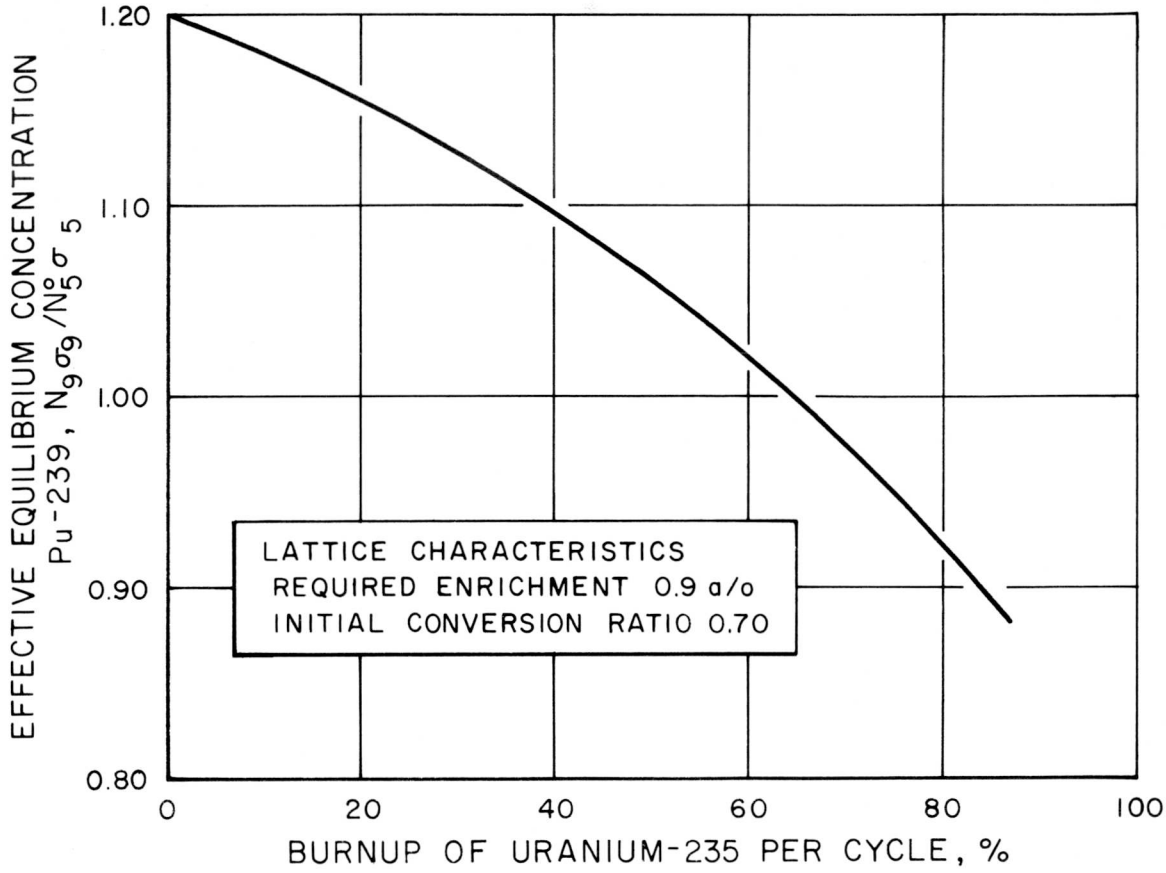

Figure 4. Effect of Fuel Irradiation per Cycle on Equilibrium Concentration of Plutonium-230

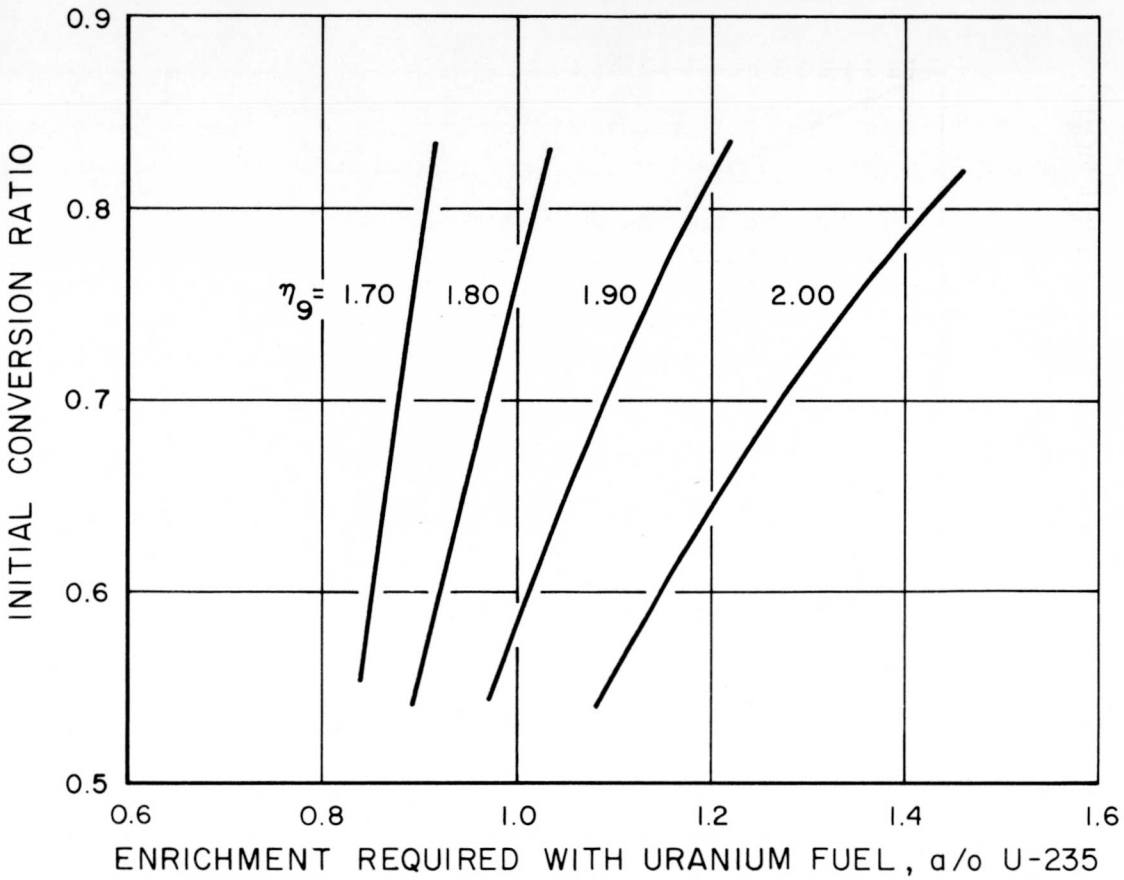

Figure 5. Conditions for Self-Sustaining Plutonium Recycle (Equilibrium Plutonium Concentration Corresponds to Zero Irradiation per Cycle)

with enriched uranium or with natural uranium plus equilibrium plutonium. For any given curve, the region to the left represents conditions of possible self-sustaining plutonium recycle.

There are several reasons why conditions in an actual fuel cycle will not correspond to those of Figure 5. The failure of plutonium-242 to reach its theoretical equilibrium concentration has been mentioned. The equilibrium concentration of plutonium will be reduced in a fuel cycle of practical irradiation or uranium-235 burnup per cycle. The significance of these considerations will be discussed with reference to a specific lattice in a later section. The next step is to determine whether an SGR can be designed within the limits of fuel enrichment and conversion ratio defined by Figure 5.

D. SGR LATTICE DESIGN FOR LOW ENRICHMENT

Figure 5 makes it clear that an SGR designed to operate on self-sustaining plutonium recycle must be one that can be made critical with uranium fuel of low enrichment, probably below 1% uranium-235. That a graphite-moderated, water-cooled reactor can be made critical with natural uranium fuel is obvious from the history of reactor development. The use of sodium as a coolant introduces negative reactivity effects of about the same magnitude as those from water cooling, but the higher moderator temperature associated with sodium cooling makes enrichment of the fuel practically a necessity. This is a result of the poorer eta values for both U-235 and Pu-239 at higher temperatures. In the first sodium graphite reactor, the SRE[12], the fuel was 2.8 a/o uranium-235. A small core (6 feet by 6 feet cylinder) as well as the use of stainless steel cladding on the fuel contributed to this enrichment requirement. In the current design of an SGR power reactor[5] the enrichment remains about 3% due to rather extensive use of stainless steel for cladding both fuel and moderator elements and for other core structural components. Both these designs utilize the canned-moderator concept of SGR design in which hexagonal blocks of graphite are individually canned in zirconium or stainless steel in order to prevent the possibility of sodium contamination of the graphite. However, advanced concepts of SGR designs have been developed which would reduce the cladding material required. The basic design change is from a core with individually canned hexagonal blocks of graphite

to the so-called calandria design in which there is, in effect, a single block of graphite pierced by holes for the fuel elements. Each fuel element is located within a process tube through which the sodium flows. This design is shown schematically in Figure 6. Development of this design is currently in progress. A second change which would effect a reduction in SGR enrichment requirements would be the substitution of a zirconium alloy for stainless steel in the core. At the present time, due to the difficulty of making a joint or bond between stainless steel and zirconium, the use of zirconium in the core would mean that the entire calandria tank would be of zirconium alloy. Such extensive use of zirconium presents technical and economic problems which are beyond the scope of this study.

On the basis of these considerations, a calandria core design with cladding and structural material limited to zirconium alloy was selected as the model for calculations. The reactor is described in Table I. Many of the values were chosen on the basis of studies conducted under the Advanced SGR Program. Others, notably fuel enrichment, initial conversion ratio, and core diameter, were selected on the basis of calculations to be described here. The fuel element selected was a 7-rod cluster of uranium metal rods as shown in Figure 7. The volume of sodium in the process tube is the estimated minimum required to remove the heat subject to the restrictions of a 15 psi pressure drop (due to friction) across the core and a temperature rise of 375°F. The zirconium liner shown in Figure 7 is to protect the graphite from sodium impregnation in the event of a leak in the process tube. The 40 mil gap between the process tube and the liner would permit any leaking sodium to fall to the bottom of the calandria tank without contaminating the graphite. This design feature affects the moderator temperature which has been shown previously (Figure 2) to be an important factor in plutonium reactivity. Practically all heat generation in the graphite must be transferred radially to the sodium. This fact establishes the sodium temperature as the minimum graphite temperature, assuming there is no separate cooling system for graphite. Moreover, the helium gap between the process tube and the graphite represents a large thermal resistance with the result that the graphite temperature is considerably above the sodium temperature. The estimated volume-average graphite temperature is 600°C (1112°F).

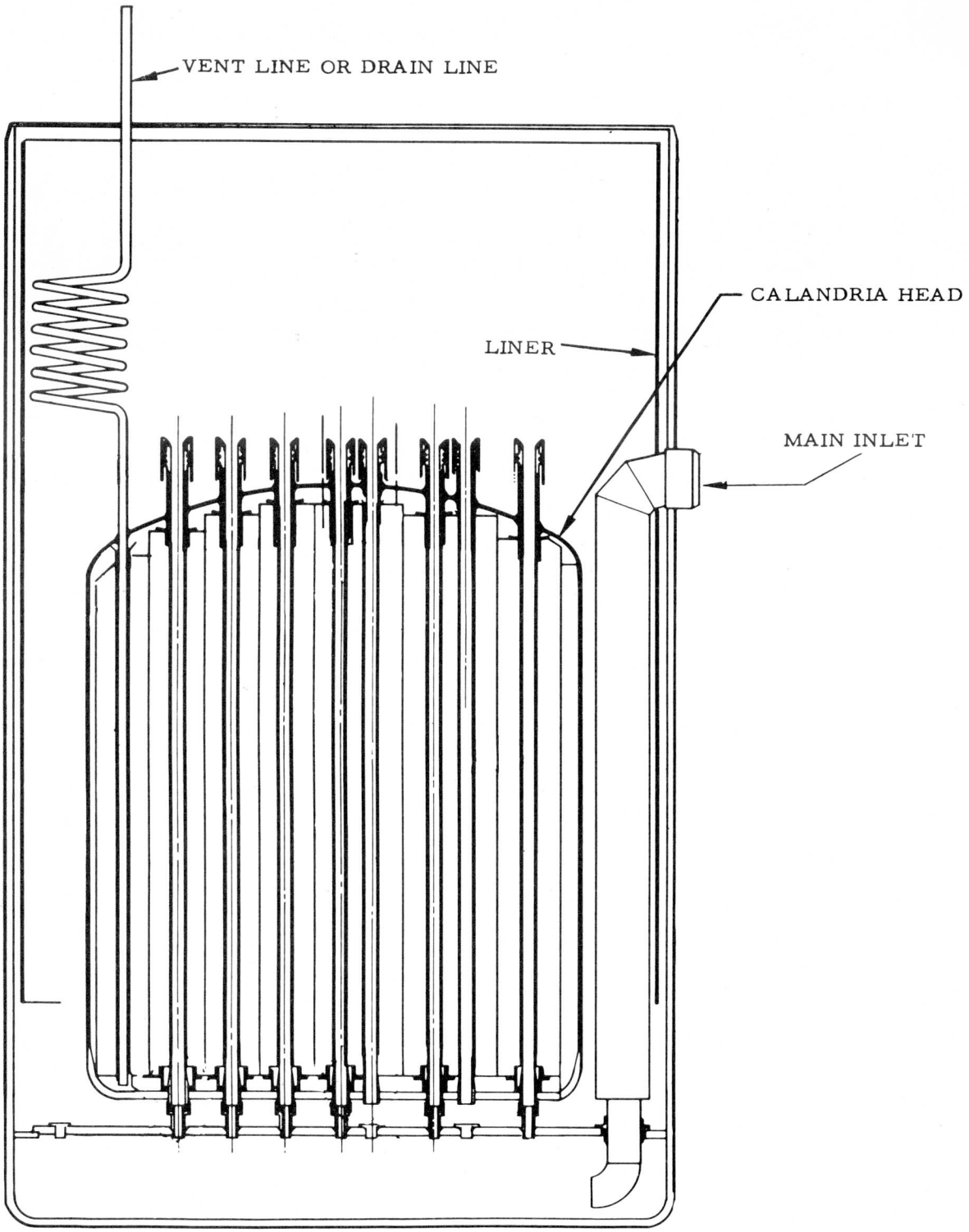

Figure 6. Calandria Core

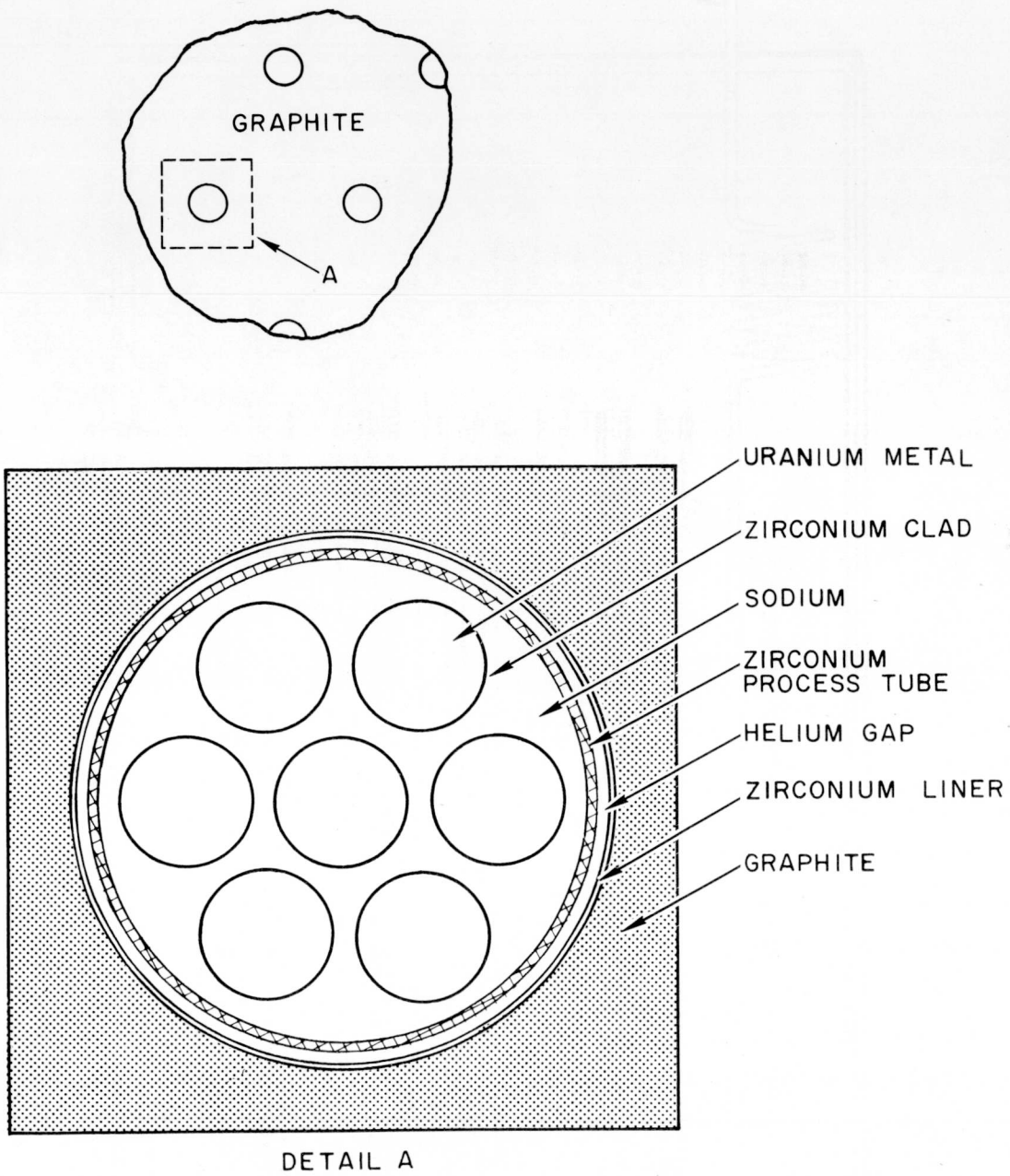

Figure 7. Low-Enrichment SGR Lattice and Fuel Element

TABLE I

GENERAL SPECIFICATIONS OF AN SGR DESIGNED FOR SELF-SUSTAINING PLUTONIUM RECYCLE

Power	
Thermal power	630 Mw
Electrical power	250 Mw
Core	
Diameter	24 feet
Height	18 feet
Lattice spacing	14 inches
Average reflector thickness	2 feet
Average moderator temperature	600°C
Fuel	
Material	Uranium metal
Enrichment	0.9 a/o uranium-235
Fuel element configuration	7-rod cluster
Rod diameter	0.75 inches
Number of fuel elements	378
Fuel specific power	8.3 Mw/t
Cladding material	Zirconium alloy
Cladding thickness	0.010 inches
Process tube inside diameter	2.79 inches
Process tube thickness	0.050 inches
Primary Coolant	
Sodium inlet temperature	625°F
Sodium outlet temperature	1000°F
Sodium pressure drop in core	15 psi
Heat flux, average	236,000 Btu/hr, sq ft

The initial lattice calculations were carried out in accordance with procedures for sodium graphite reactors previously outlined.[12] The results of these calculations are shown in Figure 8 as a plot of core diameter versus lattice spacing and fuel enrichment to give a hot clean k_{eff} of 1.05. This value is to provide the estimated excess multiplication required for fission product poisoning and modest burnup. The locus of points which satisfy the heat transfer requirements of 630 Mw thermal is also plotted. The intersections of this line with the lines of constant enrichment represent an average heat power generation of 95 kw per foot of fuel element or 13.6 kw per foot of rod. This value corresponds to a heat power density of 8.3 Mw per ton of uranium and a heat flux of 236,000 Btu per hour per square foot.

The conditions selected from these survey calculations, as indicated in Table I, were a core diameter of 24 feet, lattice spacing of 14 inches, and a fuel enrichment of 0.9 a/o uranium-235. Although the selection was somewhat arbitrary, it was largely determined by the curves in Figure 5. In general, the lower enrichment core, in spite of having a lower initial conversion ratio (in the lattice which meets both heat transfer and reactivity requirements; see Figure 8), better satisfies the conditions for self-sustaining plutonium recycle. On the other hand, since the enrichment for an infinite diameter core in these calculations was 0.85%, there is little to be gained below 0.9%. The selected core model was then analyzed in greater detail. Intracell thermal flux was estimated by the P_3 transport approximation,[13] neutron absorption in the fast neutron region was evaluated by a multigroup Fourier transform code[14] and criticality and burnup were calculated by the four-group diffusion theory code, CANDLE[15], using the results of the cell calculations for input. The results of these calculations are summarized in Table II in the form of a neutron balance for the uranium-fueled core. These calculations confirm the survey calculations except that the initial conversion is 0.70 instead of 0.74 as indicated. The burnup calculation results are shown in Table III. They indicate that the hot, clean k_{eff} of 1.05 is sufficient to give an average energy yield of 2400 Mwd/ton or a peak energy yield of about 4000 Mwd/ton. Since the criticality calculations were one dimensional radial calculations, the energy yield figure is the effective value rather than the average axial value. However, the results indicate that the core chosen would have sufficient reactivity to achieve modest burnups when charging 0.9 a/o enriched uranium fuel.

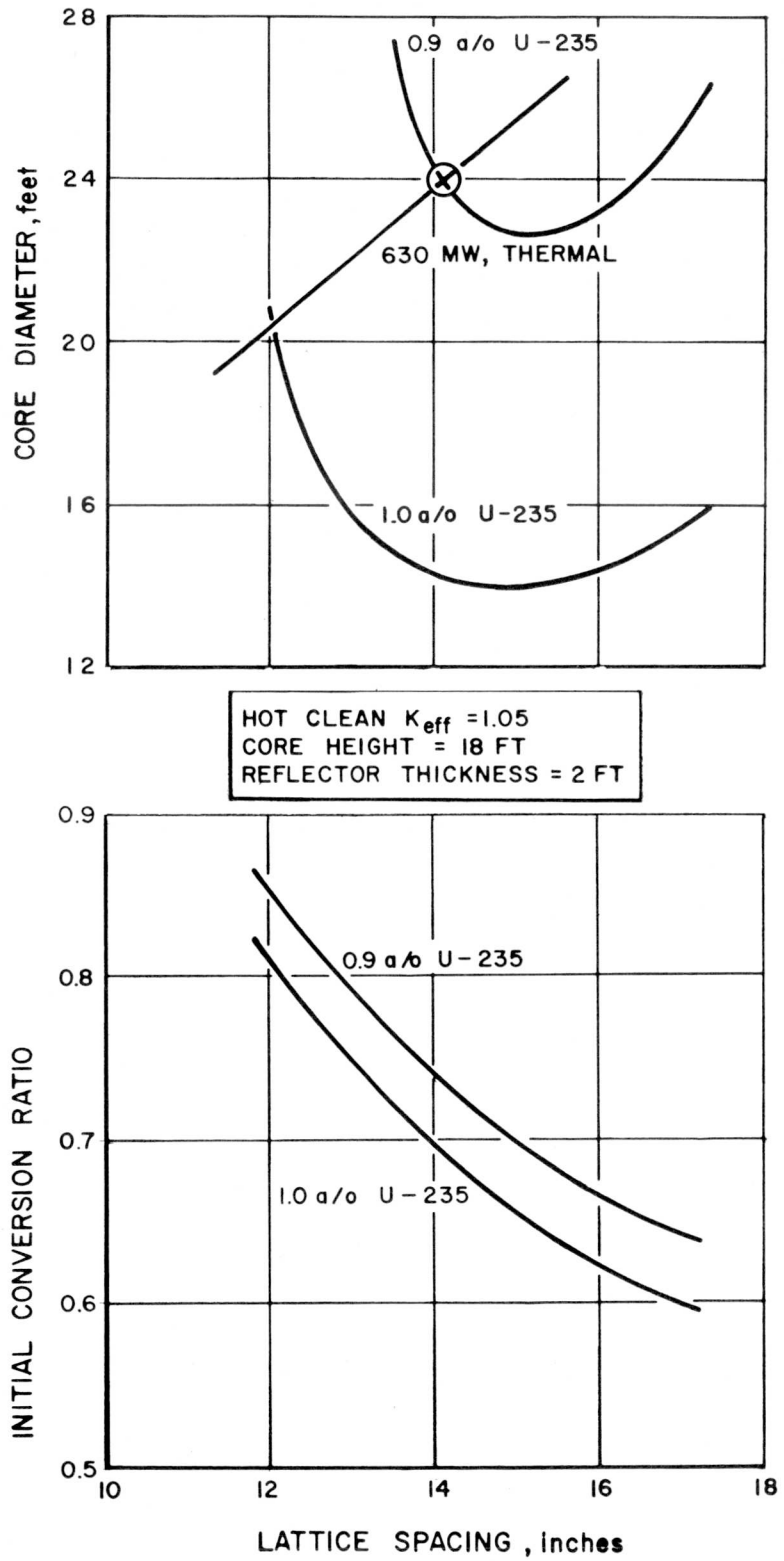

Figure 8. Lattice Parameter Inter-Relation for Low-Enrichment SGR

TABLE II

NEUTRON BALANCE

REFERENCE SGR - HOT CLEAN CONDITIONS

Neutron Energy Group	Number of Neutrons	Event	Fission Neutrons Produced	Atoms U-235 Destroyed	Atoms Pu-239 Produced
Fast	1.80	Leakage			
	2.74	Fission in U-235	6.74	2.74	
	1.12	Capture in U-235		1.12	
	1.26	Fission in U-238	3.36		
	12.82	Capture in U-238			12.82
	0.36	Capture in other material			
Thermal	2.60	Leakage			
	36.52	Fission in U-235	89.90	36.52	
	7.36	Capture in U-235		7.36	
	20.70	Capture in U-238			20.70
	7.82	Capture in other material			
	4.90	Capture in control rods			
	100.00		100.00	47.74	33.52

$$\text{Initial Conversion Ratio} = \frac{33.52}{47.74} = 0.70$$

TABLE III

REACTIVITY CHANGE IN REFERENCE SGR

Initial Enrichment	0.9 a/o Uranium-235
Initial Conversion Ratio	0.70

Specific Energy Yield (Mwd/t)		Uranium-235 Burnup (%)		
Radial Average	Radial Maximum	Radial Average	Radial Maximum	k_{eff}
0	0	0	0	1.054
480	840	6.4	10.9	1.034*
960	1670	12.0	19.8	1.031
1450	2470	17.7	28.3	1.023
1930	3260	22.5	34.9	1.014
2410	4000	27.1	40.9	1.003

Note: The above values were obtained from one-dimensional (radial) flux and criticality calculations.[15] If the k_{eff} is to apply to a reactor, then the values of energy yield and burnup must be regarded as effective axial values rather than maximum or average.

*Includes xenon, samarium effect

E. FUEL CYCLE ANALYSIS

The SGR described in Table I has a uranium fuel enrichment requirement of 0.9 a/o uranium-235 and an initial conversion ratio of 0.70. Figure 5 indicates that such a reactor should achieve self-sustaining plutonium recycle. The practicability of such a reactor fuel cycle would depend, however, on the specific energy yield (Mwd/t, megawatt-days of thermal energy per metric ton of uranium charged to the reactor) or the uranium-235 burnup attainable. The problem is: In a cyclic program of charging and discharging fuel, what is the maximum burnup of uranium-235 in the discharged natural uranium elements which can be tolerated from the standpoint of criticality? The estimation of an exact answer to this question is extremely complex if not impossible at the present time. Not only

are the data uncertain and calculation procedures untested but there are countless possible variations in the fuel cycle. For these reasons, the possible energy yield from the fuel will be examined by approximate methods.

When equilibrium plutonium recycle is established, the contribution of plutonium to the reactivity of the reactor should be nearly constant. The decline in reactivity during burning periods between fuel changes would be due to uranium-235 burnup and fission product accumulation. A calculation of fuel reactivity decline based on this simplifying assumption is outlined in Appendix D. The results of this calculation applied to the low-enrichment SGR described in Table I are given in Figure 9. The decline in reactivity is indicated by the decline in the ηf product. The curve does not contain any xenon or samarium effect. An η_9 value of 1.80 was used as reasonable for this SGR. The uncertainty in this value arising both from the data and from prediction of the moderator temperature should be recognized.

The next problem is the estimation of the reactivity of the fuel composed of fresh natural uranium and equilibrium plutonium. This calculation can be made for the case of plutonium of the theoretical equilibrium composition (Figure 1) by the procedure of Appendix C. As has been mentioned, however, plutonium-242 will not reach its theoretical equilibrium value due to unavoidable plutonium processing losses. It is necessary to estimate plutonium equilibrium quantity and composition in a more realistic case involving such losses. To facilitate this estimation an idealized model of the plutonium recycle process is given in Figure 10.

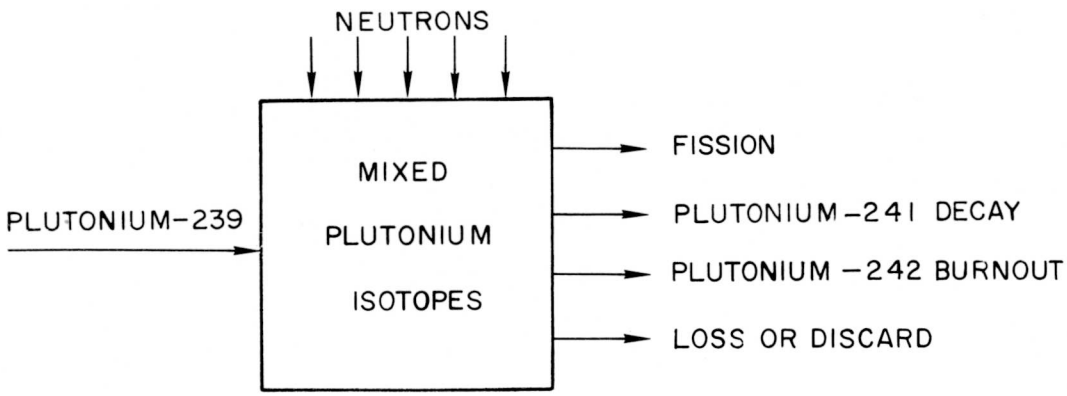

Figure 10. Schematic of Mixed Plutonium Irradiation and Flow

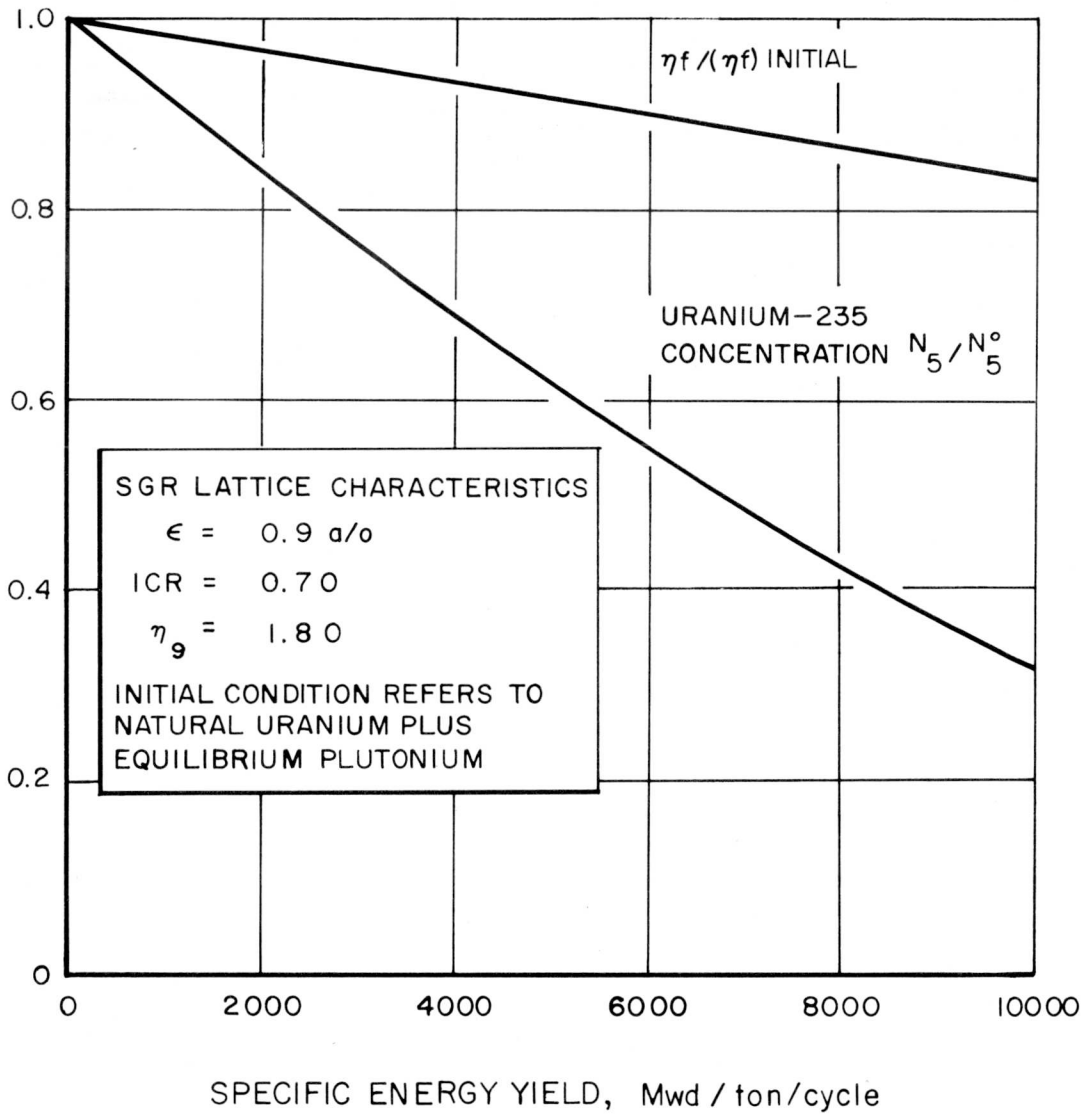

Figure 9. Decline in Reactivity of Uranium-Plutonium Fuel

In this representation, plutonium-239 is produced at a constant rate within the mixture of plutonium isotopes while the various dissipation processes for all plutonium isotopes take place continuously and at a constant rate. This model represents the situation in which recycled plutonium is mixed uniformly with fresh natural uranium. The most obvious discrepancy in the model is that the loss or discard of plutonium would take place at discrete intervals rather than continuously. The calculation of the equilibrium plutonium quantity and composition for this idealized case is outlined in Appendix E. The results, given in Figure 11, show a slight gain in reactivity of the plutonium-natural uranium mixture when plutonium is lost or discarded. The principal effect is that plutonium-242 equilibrium concentration is reduced, thus increasing the reactivity of the plutonium.

The information given in Figure 9 and 11 provides a basis for an estimate of the specific energy yield achievable. The uranium-plus-plutonium fuel has an ηf product 3% above that of 0.9 a/o enriched uranium, which gives a k_{eff} of 1.05 in the lattice under study. Allowing 4% reactivity for xenon and samarium there would be about 4% for burnup. Figure 9 indicates that the fuel should show a 4% decline in the ηf product (or k_{eff}) at a specific energy yield of about 2500 Mwd/t or a uranium-235 burnup of 20%. However, this does not represent the maximum achievable specific energy yield because the reactivity of the reactor is some weighted average of the reactivity of the fuel. The amount by which the discharged fuel can exceed the average in irradiation or specific energy yield depends on the fueling program or the arrangement of fuel in the reactor. Spinrad[16] has shown that in an arrangement of uniformly graded irradiation, the achievable energy yield is extended. The subject of fuel programming in an SGR to achieve uniformly graded irradiation has been discussed.[6] In such an arrangement the same average irradiation of the fuel is maintained radially in the core, and the fuel element of maximum irradiation exceeds the average in energy yield by a factor of about 1.8. In a reactor in which a fuel element extends the length of the core, this averaging can only be achieved in the radial direction. Using the above value of specific energy yield as a radial average, therefore, one would expect a maximum value of nearly 4500 Mwd/t. Then, if a value of 1.2 is used as the ratio of effective-to-average energy yield in the axial direction, the

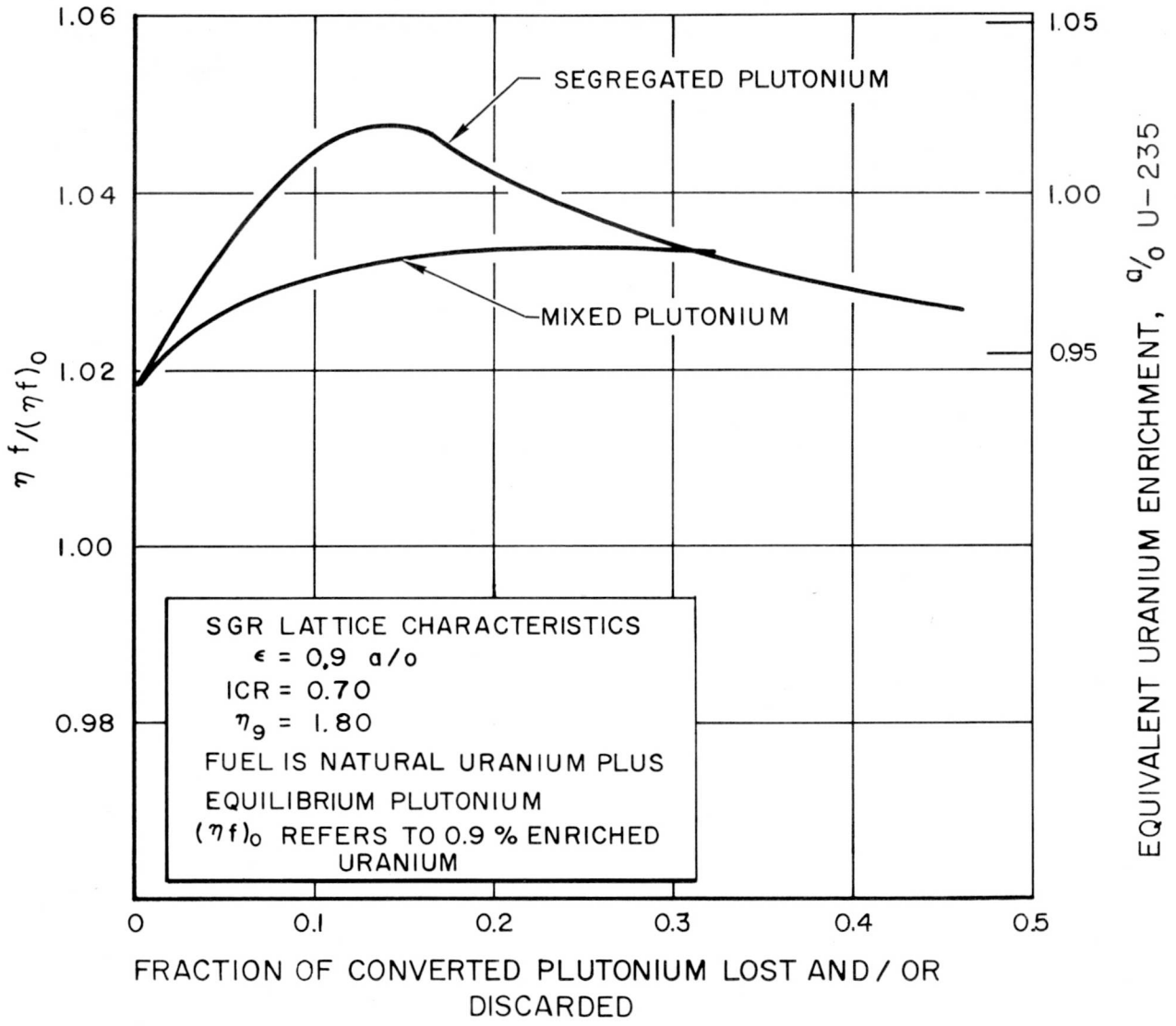

Figure 11. Fuel Reactivity at Beginning of Burning Period

average energy yield of the discharged fuel would be about 3750 Mwd/t, corresponding (Figure 9) to a uranium-235 burnup of 30%.

One objection to the "mixed plutonium" scheme of plutonium recycle as depicted in Figure 10 is that freshly converted plutonium-239 is continuously mixed with highly irradiated plutonium and, therefore, they are lost or discarded together. The difference in the nuclear fuel properties of plutonium-239 and irradiated plutonium-239 can be seen in Figure 12 which gives the variation in these properties as initially pure plutonium-239 is irradiated. The reactivity falls very rapidly and then remains roughly constant for a considerable period as plutonium-241 rises through a maximum in concentration. However, while there still remains about 15% of plutonium based on original plutonium-239, the reactivity becomes low. It would be more effective in increasing fuel reactivity if this plutonium, rather than a mixture of all plutonium, was discarded. In a practical fuel cycle, segregation of each differential quantity of plutonium converted is not possible. However, once plutonium is separated from discharged uranium fuel, it would be possible to keep it segregated in separate plutonium fuel elements. An estimate of the effect of such a scheme of plutonium segregation is made by idealizing the process as depicted in Figure 13.

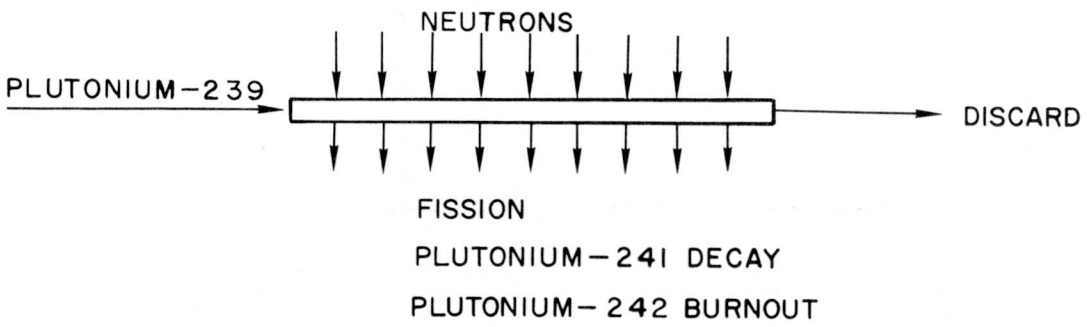

Figure 13. Schematic of Segregated Plutonium Irradiation and Flow

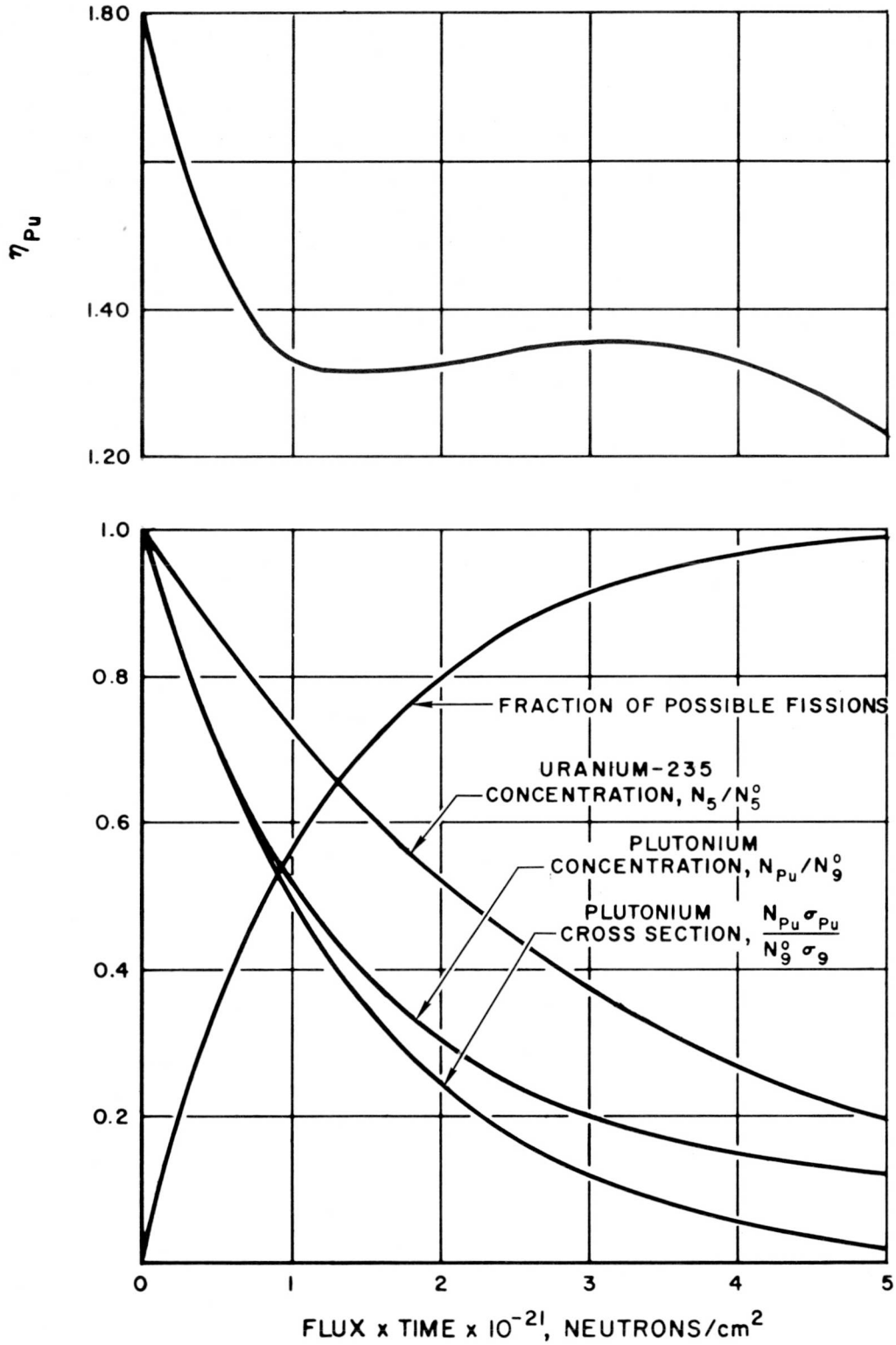

Figure 12. Characteristics of Segregated Plutonium-239 under Irradiation

In this idealized scheme, plutonium-239 is continuously segregated from uranium as it is converted. Furthermore, each differential quantity of plutonium remains segregated until a certain irradiation is reached and is then discarded. Such a scheme represents the limiting case of plutonium segregation. The properties of plutonium in the reactor at equilibrium can be found simply by integrating over the irradiation range, θ_0 ,

$$N_{Pu}\sigma_{Pu} = \int_0^{\theta_0} \frac{N_{Pu}\sigma_{Pu}}{N_9^0\sigma_9} (\theta_0 - \theta) R \, d\theta,$$

where R is the rate of conversion of plutonium-239 and $\theta = \phi t$.
θ_0 represents the irradiation at which plutonium is discarded. Also,

$$N_{Pu}\sigma_{Pu}\eta_{Pu} = \int_0^{\theta_0} \frac{N_{Pu}\sigma_{Pu}\eta_{Pu}}{N_9^0\sigma_9} (\theta_0 - \theta) R \, d\theta.$$

These terms may be found by integrating the appropriate values plotted in Figure 12. The ηf product of natural uranium plus equilibrium plutonium can then be calculated as in Appendix C. The results are plotted in Figure 11. The selective discarding of plutonium makes possible a significant increase in reactivity of the fuel, a maximum being reached at about 14% discard.

Again an estimate of the specific energy yield and uranium-235 burnup may be made. Figure 11 indicates that with the segregated plutonium scheme the ηf product of natural uranium plus plutonium is 5% higher than that of 0.9 a/o enriched uranium. This means that if 4% reactivity is allowed for xenon and samarium, about 6% is available for burnup. Figure 9 shows that the fuel would have declined 6% at about 3500 Mwd/t. Applying the same reasoning as previously to the question of reactor reactivity versus fuel reactivity, the average energy yield of discharged fuel would be 5250 Mwd/t and the uranium-235 burnup, 40%. It is assumed that although the plutonium is segregated from the uranium, it is in the same neutron flux. It should be noted that in the case of segregated

plutonium, the specific energy yield quoted would not all be released in the uranium fuel, a fact which might be important if irradiation damage was a limitation. Finally, although no estimate of the effect is made here, it appears likely that if the plutonium is segregated, a further reactivity gain could be effected by advantageous location of the plutonium, possibly in a seed and blanket arrangement.

F. CONCLUSIONS

The analysis of the lattice requirements for achievement of self-sustaining plutonium recycle in an SGR indicates that a high degree of neutron conservation is necessary. Specifically, a reactor which requires a uranium enrichment of 1 a/o uranium-235 or less, when plutonium is not recycled, appears necessary. Although current SGR designs specify enrichments of about 3 a/o uranium-235, it does appear that by careful attention to neutron economy, a 1 a/o enriched SGR is within the state of the art. Neutron economy would require, for example, that cladding and core structure material be restricted to low-cross-section materials, that sodium volume be minimized, that lattice spacing be relatively large, and that the core be relatively large.

Admitting of the technical feasibility of such a low-enrichment SGR, there are important engineering problems involved in designing and constructing such a reactor. The usual expedient available to the designers of slightly enriched reactors, that of increasing enrichment whenever the necessity for decreasing neutron economy arises, would not be admissible. The desirability of decreasing neutron economy could arise for many reasons such as alloying of uranium fuel or change from uranium metal to the carbide or oxide, reduction of core size, or decrease in fuel rod diameter to obtain higher power density. For example, a uranium carbide or oxide lattice will have a 2 to 3% lower maximum k_{eff} than a uranium metal lattice. In short, a low-enrichment design SGR could not achieve the high performance characteristics of current designs which do not have such an enrichment restriction. The result promises to be a reactor of high capital cost.

A factor of equal importance with the achievement of self-sustaining plutonium recycle is the value of specific energy yield or uranium-235 burnup which may be

attained. The estimation of specific energy yield is difficult because of uncertainties in the data and in the calculation procedures, and because of the many possible modifications of the fuel cycle. The estimates made here indicate that a specific energy yield of 3000 Mwd/t and a uranium-235 burnup of 25% should be attainable. Segregation of recycled plutonium in separate fuel elements, as opposed to mixing with fresh natural uranium, gives promise of increasing the energy yield substantially. Opportunities for further gain in energy yield lie in the advantageous location of the segregated plutonium in the core and in the possibility of reducing moderator temperature. The specific energy yields will, however, be less than those attainable in enriched reactors or in natural uranium reactors with plutonium recycle.[1]

The conclusion of this effort may be stated very simply. It appears technically feasible to design a SGR to operate on self-sustaining plutonium recycle. Such a reactor, however, would of necessity have a large core and low (for an SGR) volumetric power density, and would require exclusive use of a low cross-section material such as zirconium alloy for cladding and core structure. The fuel specific energy yield and uranium-235 burnup attainable is uncertain, but appears to be on the low side, perhaps 3000 to 5000 Mwd/t, and 25 to 40% uranium-235 burnup. Such a system would almost certainly compare unfavorably with a slightly enriched SGR if the fuel inventory annual rate and the unit costs for uranium-235 enrichment and for fuel fabrication and reprocessing, currently prevailing in the United States are applied. Expressed in a different way, it appears that an optimized plutonium recycle case will require some enrichment of the uranium feed. These conclusions in large measure reflect the low reactivity of plutonium as a reactor fuel and, therefore, they apply in general to other thermal reactor systems.

APPENDIX A
THE ETA VALUE OF EQUILIBRIUM PLUTONIUM

$$\eta_{Pu} = \frac{N_1\sigma_1\eta_1 + N_9\sigma_9\eta_9}{N_0\sigma_0 + N_1\sigma_1 + N_2\sigma_2 + N_9\sigma_9}.$$

At ideal equilibrium, with no processing losses,

$$N_0\sigma_0 = N_9\sigma_9^c,$$

$$N_1\sigma_1 = N_0\sigma_0 = N_9\sigma_9^c,$$

$$N_2\sigma_2 = N_1\sigma_1^c = \frac{N_9\sigma_9^c\sigma_1^c}{\sigma_1}.$$

Therefore,

$$\eta_{Pu}(\infty) = \frac{N_9\sigma_9^c\eta_1 + N_9\sigma_9\eta_9}{N_9\sigma_9^c + N_9\sigma_9^c + \frac{N_9\sigma_9^c\sigma_1^c}{\sigma_1} + N_9\sigma_9}.$$

Dividing by $N_9\sigma_9$,

$$\eta_{Pu}(\infty) = \frac{\frac{\sigma_9^c}{\sigma_9}\eta_1 + \eta_9}{1 + \frac{2\sigma_9^c}{\sigma_9} + \frac{\sigma_1^c\,\sigma_9^c}{\sigma_1\sigma_9}}.$$

Since

$$\eta_9 = \left(1 - \frac{\sigma_9^c}{\sigma_9}\right)\nu_9$$

or

$$\frac{\sigma_9^c}{\sigma_9} = 1 - \frac{\eta_9}{\nu_9},$$

then

$$\eta_{Pu}(\infty) = \frac{\eta_9 + \eta_1\left(1 - \frac{\eta_9}{\nu_9}\right)}{1 + \left(2 + \frac{\sigma_1^c}{\sigma_1}\right)\left(1 - \frac{\eta_9}{\nu_9}\right)} .$$

A value of $\nu_9 = 2.91$ was used. It appears that for plutonium-241, unlike plutonium-239, the values of η, and σ_1^c/σ_1 will not vary much with temperature.[8] Therefore, constant values of $\eta_1 = 2.213$ and $\sigma_1^c/\sigma_1 = 0.27$ were used. The former value was reduced to 2.00 to allow for a radioactive decay of plutonium-241 (Appendix B). These substitutions give

$$\eta_{Pu}(\infty) = \frac{\eta_9 + 2.00\left(1 - \frac{\eta_9}{2.91}\right)}{1 + 2.27\left(1 - \frac{\eta_9}{2.91}\right)}$$

and

$$\eta_{Pu}(\infty) = \frac{2.00 + 0.313\eta_9}{3.27 - 0.780\eta_9} .$$

If the concentration of plutonium-242 is assumed to be zero,

$$\eta_{Pu}(\infty) = \frac{2.00 + 0.313\eta_9}{3.00 - 0.687\eta_9} .$$

APPENDIX B
EFFECT OF PLUTONIUM-241 DECAY

The concentration of plutonium-241 in the fuel is affected by the fact that it undergoes radioactive decay with a half-life of 13.2 years. The rate of disappearance of plutonium-241 in the core is proportional to $\phi\sigma_1 + \lambda_1^B$. The flux value used in this term must be discounted for the time the plutonium is in processing as well as for the load factor of the reactor. In the present situation a value of 1.5×10^{13} n/cm^2/sec appears reasonable. Using a value of 1250 barns for the cross section of plutonium-241 leads to the following comparison of these values

$$\phi\sigma_1 = 1.625 \times 10^{-8} sec^{-1}$$

$$\lambda_1^B = 0.166 \times 10^{-8}.$$

The relative values of these terms indicate that at equilibrium the concentration of plutonium-241 will be about 10% less than would be calculated if radioactive decay was neglected. An allowance for this effect has been made by reducing the η_1 value from 2.213 to 2.0 in the calculation of $\eta_{Pu}(\infty)$ (Appendix A), continuing to set the $N_1\sigma_1$ term equal to $N_9\sigma_9^c$. The reasoning here is that the neutron capture rate is maintained by americium-241 formed by the decay of plutonium-241.

APPENDIX C
ESTIMATION OF EQUIVALENT PLUTONIUM AND URANIUM-235 CONCENTRATIONS

Postulate that the primary effect on reactivity resulting from addition of plutonium or uranium-235 to natural uranium is in the change of the ηf term. For natural uranium fuel,

$$\eta_0 f_0 = \frac{N_5^0 \sigma_5 \eta_5 f_0}{N_5^0 \sigma_5 + N_8 \sigma_8}$$

$$\eta_0 f_0 = \frac{\eta_5 f_0}{1 + 138 \dfrac{\sigma_8}{\sigma_5}}$$

If plutonium is added to the natural uranium in the same lattice,

$$\eta f = \frac{\eta_5 + \dfrac{N_{Pu} \sigma_{Pu} \eta_{Pu}}{N_5^0 \sigma_5}}{\left(\dfrac{1}{f_0}\right)\left(1 + 138 \dfrac{\sigma_8}{\sigma_5}\right) + \dfrac{N_{Pu} \sigma_{Pu}}{N_5^0 \sigma_5}}$$

It is assumed here that the relative flux in the fuel and non-fuel components of the lattice is unaffected by the addition of plutonium. While this assumption may produce a significant error in the estimation of absolute reactivity, it is sufficiently accurate for the present purpose of comparing effects of plutonium and uranium-235. The equivalent expression for the case of "addition" of uranium-235 to the fuel is,

$$\eta f = \frac{\eta_5 \dfrac{138}{100 - \epsilon}}{\left(\dfrac{1}{f_0}\right)\left(1 + 138 \dfrac{\sigma_8}{\sigma_5}\right) + \dfrac{139\epsilon - 100}{100 - \epsilon}} .$$

For any values of $N_{Pu}\sigma_{Pu}$ and η_{Pu}, the equivalent enrichment, ϵ, is regarded as that which makes the resulting ηf values equal.

Values used for the various constant values in the above expression were

$$f_0 = 0.875$$

$$\eta_5 = 2.00$$

$$\sigma_8/\sigma_5 = 0.0040$$

$$\sigma_9/\sigma_5 = 4.0$$

APPENDIX D
REACTIVITY CHANGE WITH BURNUP IN A PLUTONIUM-FUELED CORE

For the sake of computational simplicity the criterion for self-sustaining plutonium recycle (Figure 5) was taken as the equality of reactivity of an enriched-uranium fuel and a natural-uranium-plus-recycled-plutonium fuel at the beginning of a burning period. Actually, because of a difference in the rate of decline of reactivity with burnup between these two fuels, this criterion is strictly applicable only to the limiting case of zero burnup. An approximation to the rate of decline of reactivity of natural-uranium-plus-plutonium fuel can be made if it is assumed that the plutonium contribution remains unchanged, and that, after equilibrium is established, the decline is due only to the disappearance of uranium-235 and the buildup of fission products. As in Appendix C, the ηf product at the beginning of a burning period is

$$\eta f = \frac{\eta_5 + \dfrac{N_{Pu}\sigma_{Pu}\eta_{Pu}}{N_5^0\sigma_5}}{\left(\dfrac{1}{f_0}\right)\left(1 + 138\dfrac{\sigma_8}{\sigma_5}\right) + \dfrac{N_{Pu}\sigma_{Pu}}{N_5^0\sigma_5}}.$$

After some burnup, reducing N_5^0 to N_5, the expression may be written

$$\eta f = \frac{\dfrac{N_5\eta_5}{N_5^0} + \dfrac{N_{Pu}\sigma_{Pu}\eta_{Pu}}{N_5^0\sigma_5}}{\left(\dfrac{1}{f_0} - 1\right)\left(1 + 138\dfrac{\sigma_8}{\sigma_5}\right) + \dfrac{N_5}{N_5^0} + 138\dfrac{\sigma_8}{\sigma_5} + \dfrac{(N\sigma)_{fp}}{N_5^0\sigma_5} + \dfrac{N_{Pu}\sigma_{Pu}}{N_5^0\sigma_5}}.$$

The values of $N_{Pu}\sigma_{Pu}$ of η_{Pu} are assumed to remain constant. These values are a function of lattice characteristics and burning period length as indicated in Figures 1, 3, 4. The cross section value of a fission product pair was assumed to be $0.1\sigma_5$. The specific energy yield for any given uranium-235 burnup is calculated from the total fissions in uranium and plutonium. The resulting decline in the ηf product as a function of specific energy yield for one set of lattice parameters is given in Figure 9.

APPENDIX E
COMPOSITION OF EQUILIBRIUM PLUTONIUM WITH LOSS OR DISCARD

For the case of mixed plutonium as idealized in Figure 10, the isotope change differential equations may be written

$$\frac{dN_9}{d(\phi t)} = \dot{N}_9 = R_c - N_9\sigma_9 - R_d F_9,$$

$$\dot{N}_0 = N_9\sigma_9^c - N_0\sigma_0 - R_d F_9 \frac{N_0}{N_9},$$

$$\dot{N}_1 = N_0\sigma_0 - N_1\sigma_1 - R_d F_9 \frac{N_1}{N_9},$$

$$\dot{N}_2 = N_1\sigma_1^c - N_2\sigma_2 - R_d F_9 \frac{N_2}{N_9},$$

where

R_c = Rate of conversion of plutonium-239

R_d = Rate of discard of plutonium

$$F_9 = \frac{N_9}{N_{Pu}}.$$

For a given rate of conversion and discard, the composition of equilibrium plutonium can be calculated by setting the derivatives equal to zero and solving the equations. The rate of conversion depends on the lattice characteristics. The reactivity of the resulting plutonium and its mixture with natural uranium can then be calculated by the procedure of Appendix C. One example of the resulting reactivity as a function of discard rate is given in Figure 11.

APPENDIX F
NOMENCLATURE

E = Fuel exposure or specific energy yield (Mwd/metric ton of uranium)

ϵ = Uranium enrichment (a/o uranium-235 in uranium)

f = Thermal utilization

f_0 = Thermal utilization when lattice is fueled with natural uranium

ICR = Initial conversion ratio

k_{eff} = Effective multiplication

N = Atomic concentration

P = Resonance region capture in uranium-238 per fission neutron from thermal fission

R_c = Rate of conversion of uranium-238 to plutonium-239

R_d = Rate of loss or discard of plutonium

η = Fission neutrons produced per absorption in fuel material

$\overline{\eta}$ = Eta value in a Maxwellian population of neutrons

$\eta(\infty)$ = Eta value of equilibrium plutonium

ν = Neutrons produced per fission

σ = Effective absorption cross section in neutron spectrum of the reactor

$\overline{\sigma}$ = Cross section in a Maxwellian population of neutrons

θ = Irradiation (flux x time)

Subscripts

0 = Plutonium-240

1 = Plutonium-241

2 = Plutonium-242

5 = Uranium-235

6 = Uranium-236

8 = Uranium-238

9 = Plutonium-239

Pu = Plutonium isotopic mixture

Superscripts

c = Radiative capture

f = Fission

o = Initial value

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