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Effect of Fission Products on Fuel Reactivity Lifetime

R. S. Carlsmith, A. M. Perry

ABSTRACT

The reactivity-limited lifetimes of thorium-U²³⁵ fuels have been calculated for homogeneous, graphite-moderated systems. The fuel composition, core leakage, temperature, and specific power were varied over a wide range. The sensitivity of the results to assumed fission-product cross sections and yields was determined. Lifetimes of over 2 fissions per initial fissile atom were obtained with initial fission-product cross sections (excluding the high cross section nuclides) of $\gamma\sigma_0 = 43$ barns per fission and $\gamma I_a = 215$ barns per fission. Doubling the assumed fission-product cross sections gave a typical reduction in lifetime of only 15%.

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Subject: Effect of Fission Products on Fuel Reactivity Lifetime

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Page 15 (Fig. 7) change "Leakage = 0.5" to read "Leakage = 0.05".

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EFFECT OF FISSION PRODUCTS ON FUEL REACTIVITY LIFETIME

Introduction

In spite of a large amount of experimental work and careful analysis over the years, there remains a considerable degree of uncertainty in the aggregate cross section of the fission products.

The calculations described in this report represent an attempt to determine how the lifetime of fuels of various compositions and various assumed operating conditions is limited by reactivity considerations. A number of different assumptions were made regarding the aggregate cross section of the fission products, and the effect on reactivity lifetimes of each assumption was determined.

The reactor model used throughout was that of a large, homogeneous, graphite-moderated core fueled with a mixture of U^{235} and Th^{232} . An equilibrium cycle with graded exposure of the fuel is assumed, that is, the fuel is moved so that the flux is not a function of either time or position within the core.

The parameters which were varied were the initial carbon-to-uranium ratio, the initial thorium-to-uranium ratio, the specific power, the moderator temperature, the net neutron leakage from the core, and the cross sections of the fission products. All of the calculations were done with the LT-1 code,¹ a two-group lifetime program for the IBM-7090.

The reference point from which the fission-product cross sections were studied is provided by Nephew's work.² In brief, Nephew tabulated all of the known fission products with their measured yields and cross sections. He then attempted to find a smaller number of "fission product pseudo-elements" which would approximate the behavior of the actual fission products over a wide range of burnup conditions. His results give a recipe by which the behavior of the fission-product cross sections for each fissionable nucleus can be well represented by a total of six elements and pseudo-elements. Xe^{135} is represented explicitly. A second element represents the other nuclides



whose cross sections are above 10,000 barns: Sm^{149} , Sm^{151} , Gd^{155} , Eu^{155} , Cd^{113} , and Gd^{157} . The other four pseudo-elements together represent the remainder of the 200 or so fission product and daughter nuclei.

The yields of the xenon and of the samarium group are known reasonably well and were not varied in the present study. The variation of the yields, thermal cross sections and resonance integrals of the other four pseudo-elements make up the major part of this study. In particular, the values found by Nephew for these quantities are referred to hereafter as "normal" while other assumed values are compared to these "normal" values. The other assumed values for the fission-product cross sections are all greater than "normal", since it is believed that Nephew's method of adding up known nuclei could only result in an underestimate of the total (though perhaps only a slight underestimate).

It is often the custom when referring to fission-product aggregate cross section to give a thermal cross section and resonance integral, normalized to a yield of unity, in units of barns per fission. In these terms the cross sections of Nephew, excluding xenon and samarium, are at zero burnup $g\sigma_0 = 43.0$ barns per fission and $I = 215.3$ barns per fission for U^{235} . They are $g\sigma_0 = 37.0$ barns per fission and $I_a = 174.0$ barns per fission for U^{233} . As burnup occurs, the poisoning effect of the fission products depends not only on the thermal cross section and the resonance integral but also on the rate of saturation, which can be evaluated in terms of the assumed yield of a fission-product nuclide or pseudo-element. In the present study the thermal cross sections, resonance integrals, and yields of the pseudo-elements were varied independently to see what effect each assumption would have on fuel lifetime. Increasing the thermal cross section of a pseudo-element to a value greater than Nephew found is equivalent to assuming that some known nuclide or nuclides have actually higher thermal cross sections than the measured values. Similarly, increasing the resonance integral of a pseudo-element is equivalent to assuming that some nuclide has a higher resonance integral than has been measured. Increasing the yield of a pseudo-element, on the other hand, is equivalent either to assuming that some known nuclide

has actually a higher yield than has been measured, or that some nuclide which has escaped notice has a significant yield and cross section. This latter possibility is perhaps the most likely source of error in obtaining the aggregate fission-product cross section by totaling the contributions of individual nuclides. An increase in the yield of the fission products will always have more effect on the lifetime of the fuel than the same percentage increase in both thermal cross section and resonance integral, since the saturation effects will be smaller.

Still another possibility, for which no computations have been made in the present study, is that second order effects may be more significant than estimated by Nephew. He made the implicit assumption that new products formed by neutron capture in a fission-product nuclide have a negligible cross section except in the mass 151 chain. Sampson *et al.*³ have calculated that only 20% of the fission-product cross section at 2 fissions per initial fission atom is contributed by the higher order effects. Of this 20%, most is in the mass 151 chain.

In addition to the fission products, the nuclides taken explicitly into account in this study were: Th²³², Pa²³³, U²³³, U²³⁴, U²³⁵, U²³⁶, Np²³⁷, and the graphite moderator. The concentrations and absorptions of each of the foregoing were computed as a function of time in the calculation. The absorptions due to increase in samarium poisoning after a long shutdown were computed, and enough excess reactivity to offset these absorptions was allowed. (This is called samarium override.) In addition, an excess reactivity of 0.01 was allowed in each calculation for structural and control requirements. In accordance with the model of an equilibrium cycle with graded exposure the fast and thermal fluxes for a calculation must be those produced by the time-averaged concentrations of all the nuclides. This requirement necessitated an iterative procedure in the calculation whereby values were assumed for the fluxes, the average concentrations were computed and used to obtain improved values for the fluxes. This procedure was followed until convergence was obtained.

Results

In Fig. 1 are given some typical results of the calculations. Several features of the curves should be noted. The atomic ratios of carbon to uranium and thorium to uranium are used to specify completely the composition of the fresh fuel. A core leakage probability of 0.05 corresponds to a 15-ft-diam core with a void fraction of 0.39 and a graphite reflector, or a somewhat smaller core with a smaller void fraction. The legend that fission-product cross sections and yields were "normal" in the calculations for Fig. 1 indicates that the values from Nephew's report² were used. The specific power, expressed as Mw/kg of U^{233} plus U^{235} in the entire core, is a measure of the effects of samarium override, absorptions in Xe^{135} , and absorptions in Pa^{233} . The fuel reactivity lifetime is expressed here in units of fissions per initial fissionable atom (abbreviated fifa). Lifetimes are expressed in these units throughout this report except where otherwise noted. The temperature referred to is the moderator temperature.

The lifetime as a function of initial thorium-to-uranium ratio goes through a maximum for the following reasons. Fuels with little or no thorium have a small conversion ratio and are limited in lifetime to essentially the burnup of most of the original U^{235} or, in other words, to somewhat less than 1.0 fifa. As the proportion of thorium in the fresh fuel is increased, the conversion ratio increases, and the lifetime can include the burnup of most of the original fuel plus a substantial amount of U^{233} which has been bred into the system. At the other extreme of compositions, with a very high proportion of thorium in the fresh fuel, the conversion ratio is high but still less than unity. In these cases, the thorium absorbs so many neutrons that the initial k_{eff} of the system is only slightly greater than 1.0 and a very slight amount of burnup is sufficient to make it subcritical.

It can be seen that the specific power is important in determining the fuel lifetime. At low specific powers this is because of the effect of equilibrium xenon poisoning. Xenon poisoning approaches an asymptotic value as the specific power is increased, and does not increase much for specific powers above about 3 Mw/kg. At higher specific powers the control poison

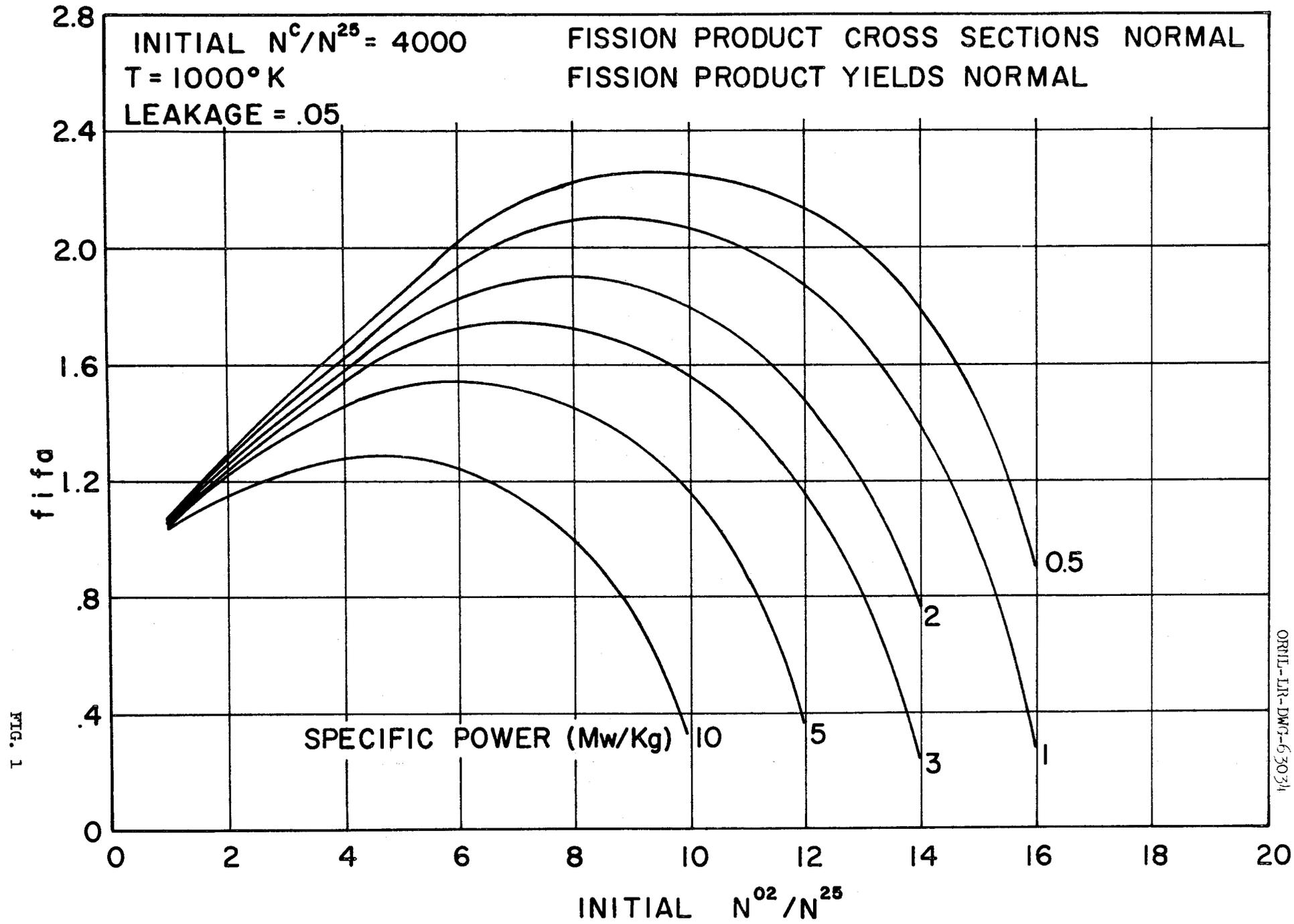


FIG. 1

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necessary for samarium override and the conversion ratio losses due to absorptions in Pa^{233} are important. The samarium override poisoning is always proportional to specific power while the protactinium losses particularly affect the systems with high conversion ratios (those with high thorium-to-uranium ratios). No allowance was made for xenon override (which would be extremely high at high specific powers) since it was argued that one could afford to wait for the peak xenon to decay.

In Fig. 2 the two opposing trends of conversion ratio and initial excess k_{eff} are shown explicitly. The conversion ratios that are shown are average values over the lifetime of the fuel. The initial excess k_{eff} is defined as the value which would obtain if the entire core were filled with fresh fuel.

Figure 3 shows another aspect of the effects of changing the initial thorium-to-uranium ratio. The power density in the fresh fuel is compared to the average for all of the fuel in the core. The cores with a higher proportion of thorium in the fresh fuel (those with higher conversion ratios) maintain a higher fuel value throughout their lifetime and thus exhibit a lower ratio of peak-to-average power density. It should be kept in mind that the peak-to-average power given here is that due to the variation of the fuel fission cross section alone and is in addition to any effects due to the spatial variation of the flux.

The lifetime calculations for the same conditions are replotted in Fig. 4, using a different definition and units for lifetime. In this case lifetime is given in megawatt-days per metric ton of uranium plus thorium in the fresh fuel. It can be seen that this type of lifetime does not go through a maximum with respect to initial thorium-to-uranium ratio, but instead increases with increasing uranium content in the fuel.

In Fig. 5 the lifetime and several other quantities of interest are plotted against specific power with initial thorium-to-uranium ratios which give the maximum fifa for each specific power. The subsequent curves all have the initial thorium-to-uranium ratio chosen to give maximum fifa .

Figure 6 compares the fuel lifetimes for different moderator-to-fuel ratios. It appears that an initial carbon-to-uranium ratio of about 4000 gives longer fuel lifetimes than either higher or lower values. This effect

FIG. 2

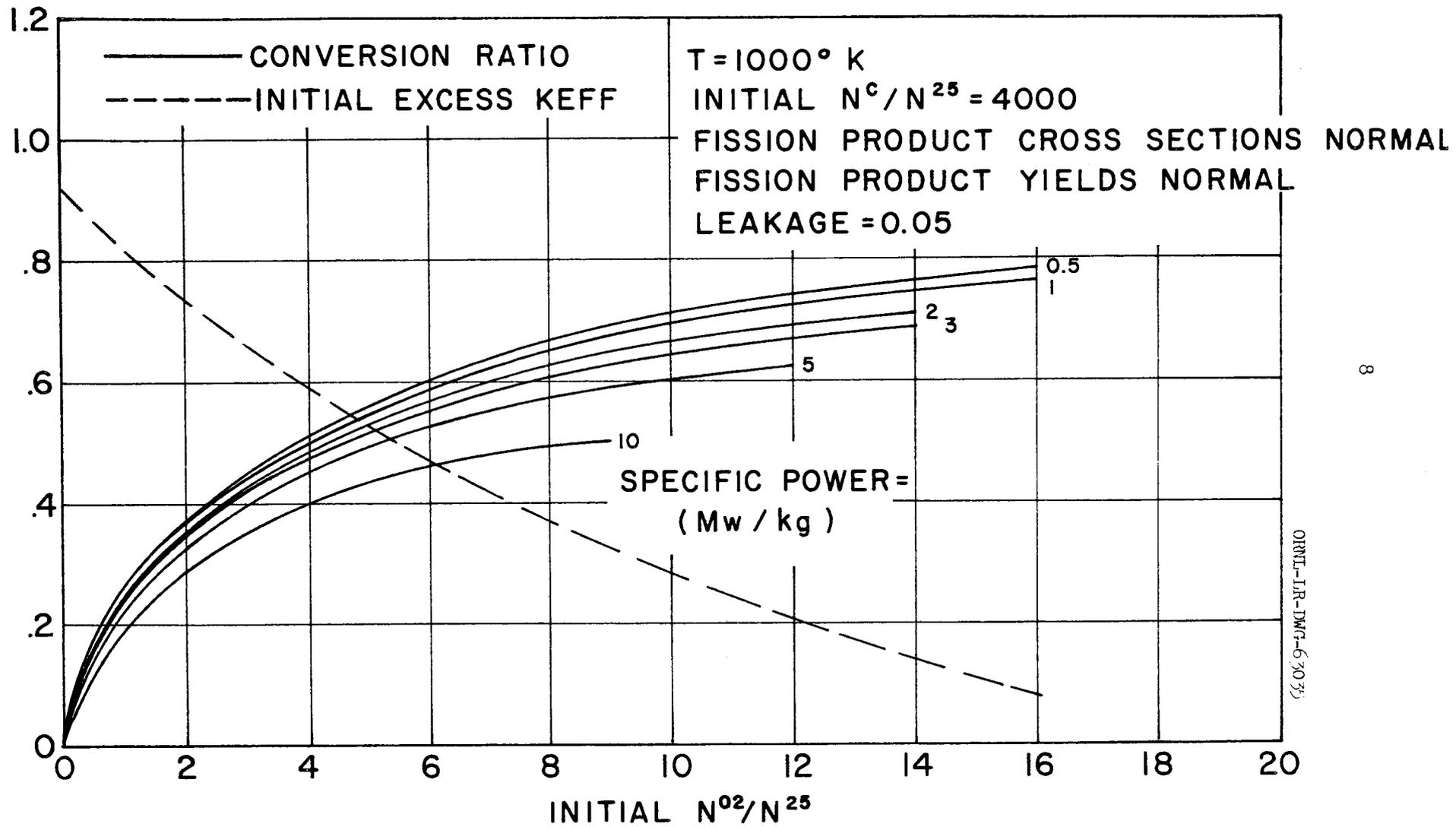
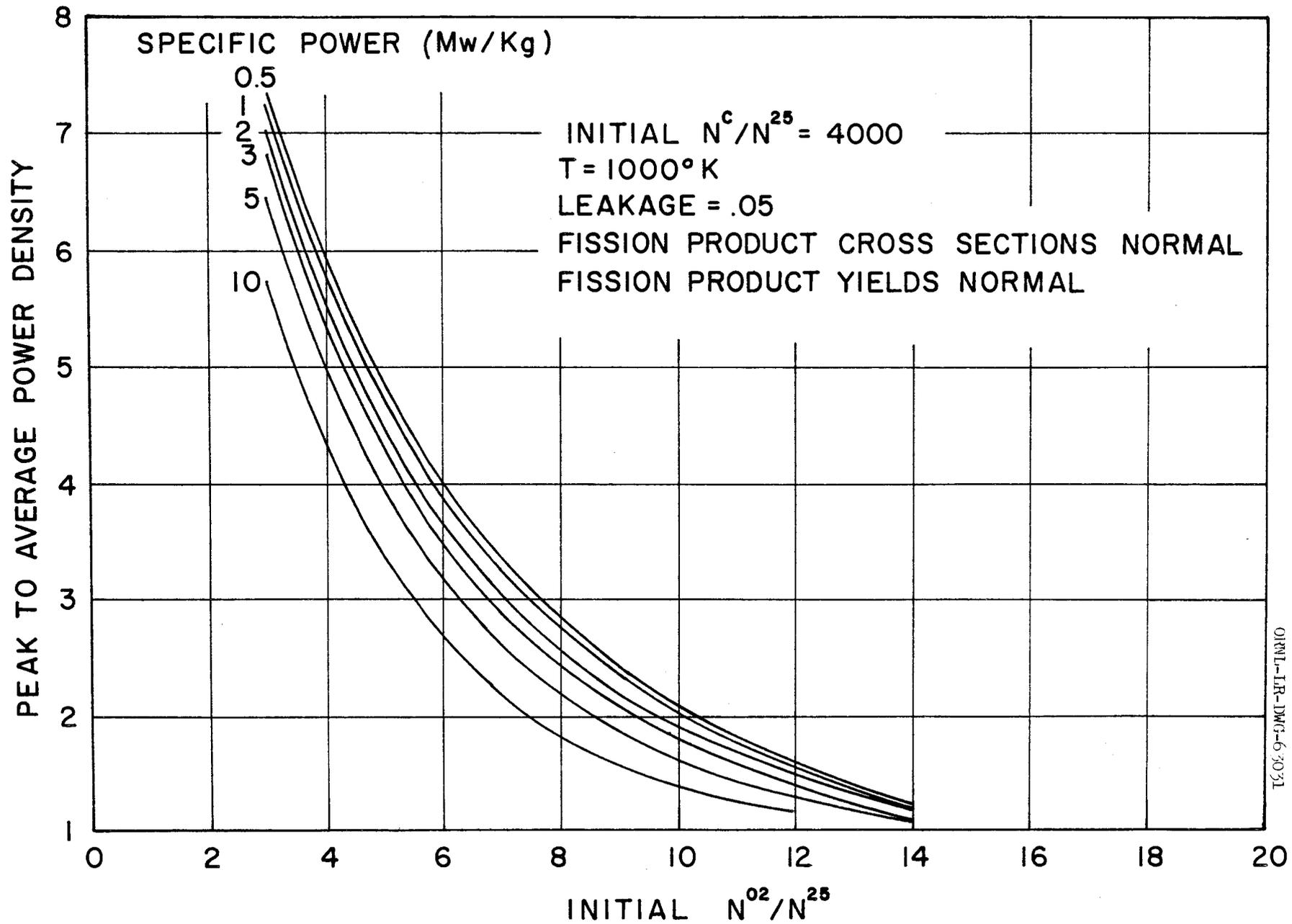


FIG. 3



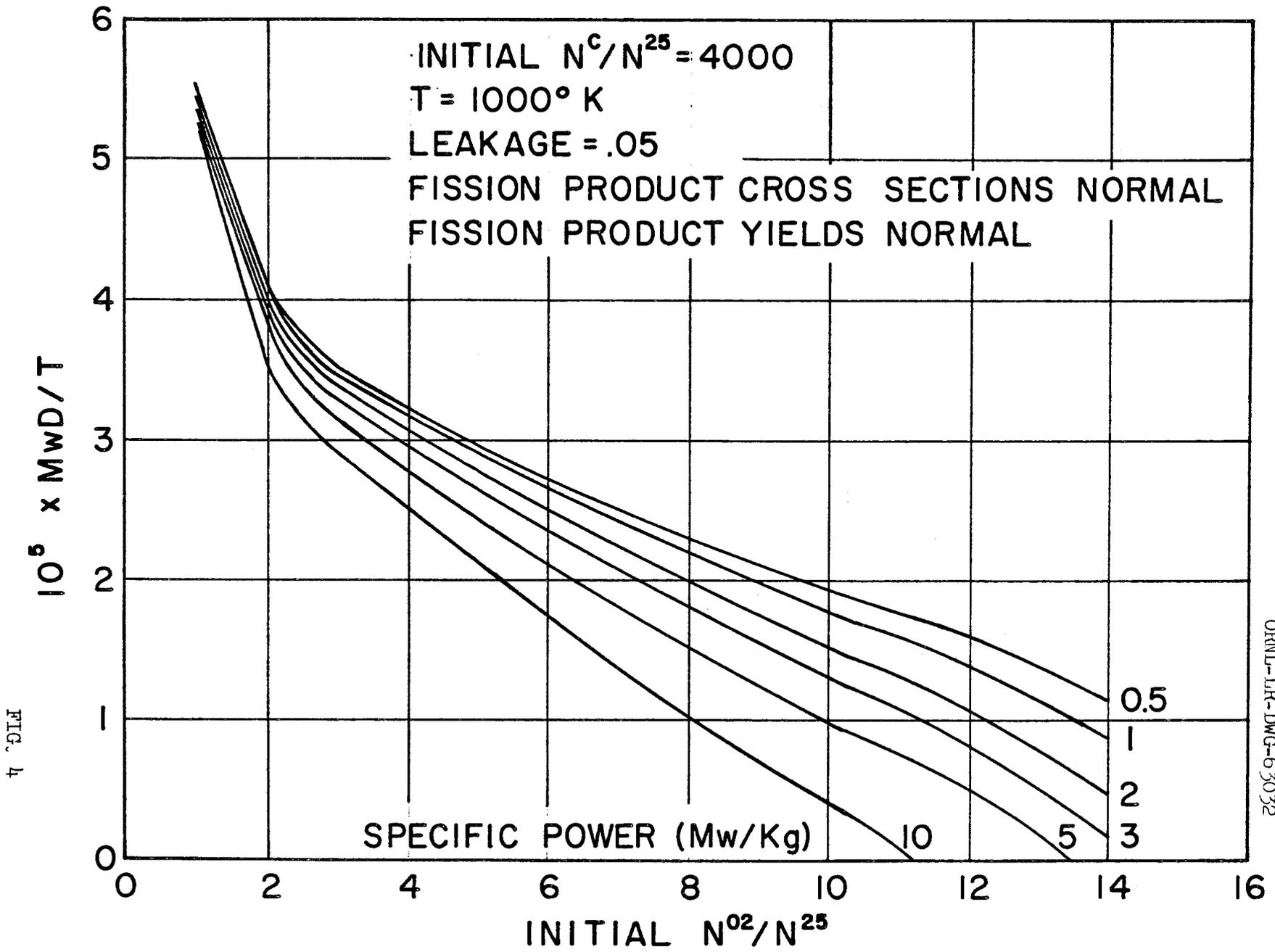


FIG. 4

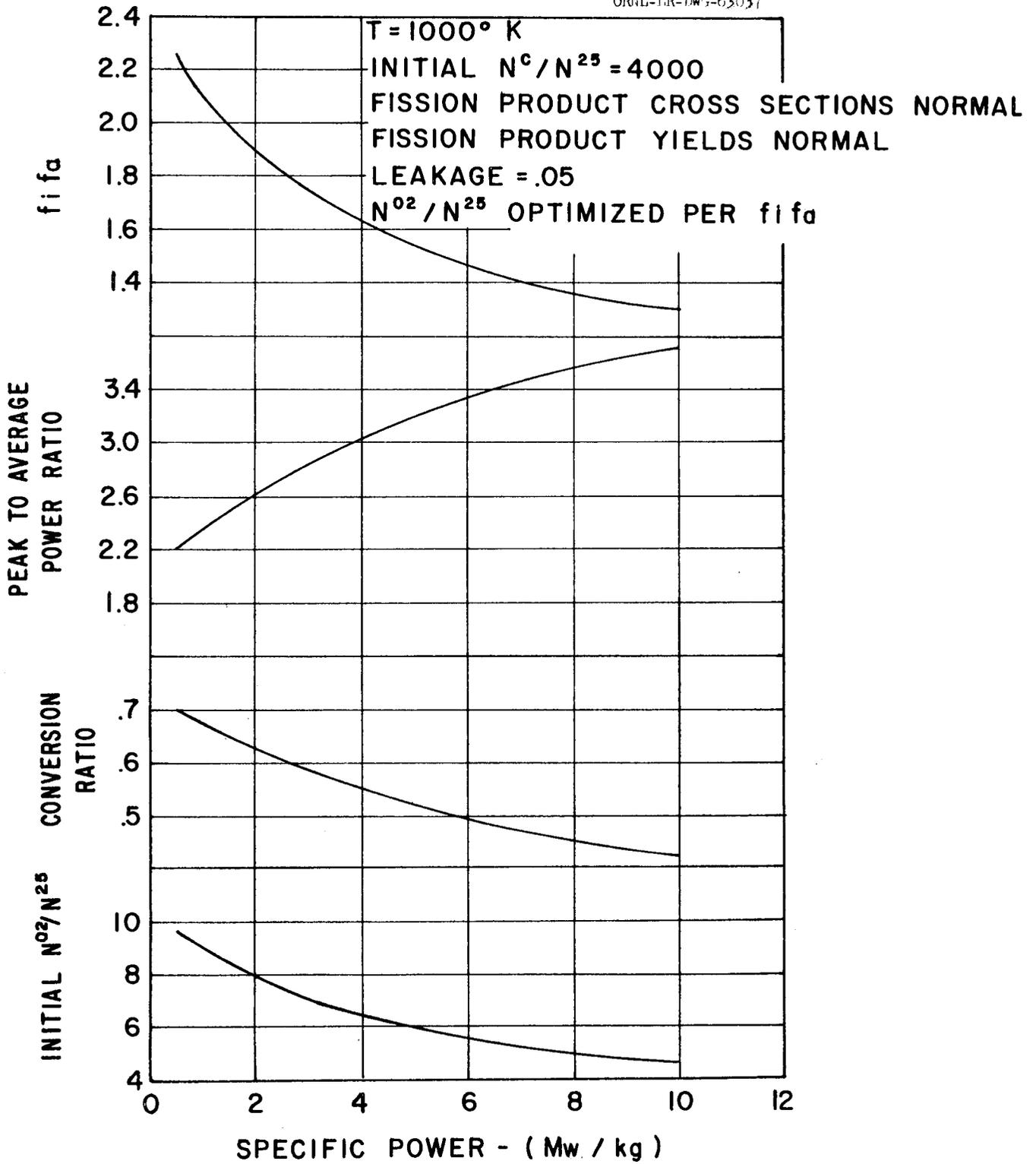
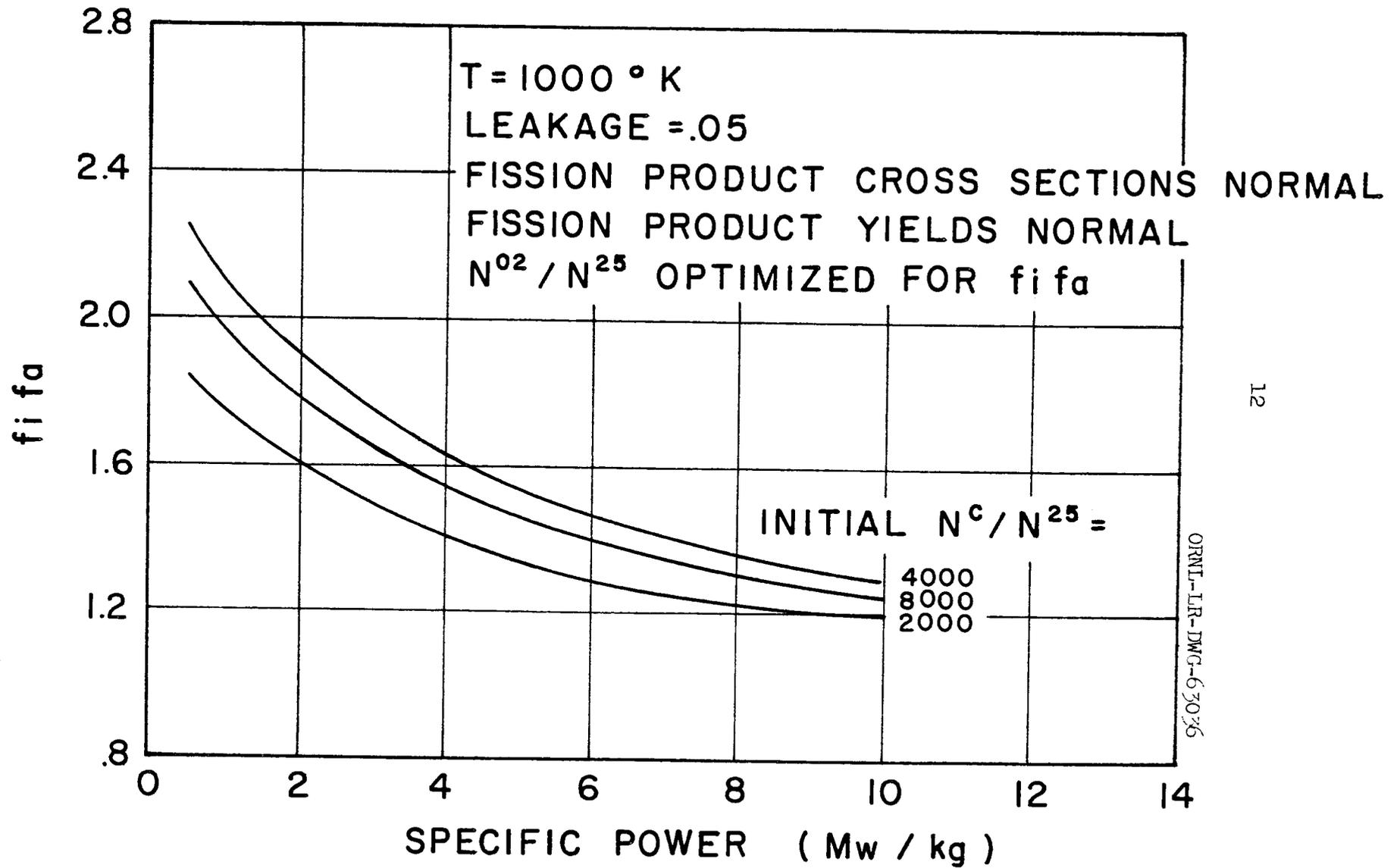


FIG. 5

FIG. 6



seems to be caused principally by the balancing of two compensating factors: the better value of $\bar{\eta}$ for the fuel in the more thermal spectrum produced by high N^C/N^{25} , and the fewer parasitic absorptions in the moderator with low N^C/N^{25} . The differences in fuel lifetime are small, however, over a wide range of moderator-to-fuel ratios so that other considerations are likely to be decisive in choosing a design value for this parameter.

The effect on lifetime of varying the moderator temperature can be seen by comparing Figs. 6 and 7 with Fig. 5. Over the range of 800°K to 1300°K it appears that longer fuel lifetimes are produced by lower temperatures, but the differences are almost certainly minor in comparison with other results of changing the core temperatures.

The decrease in lifetime associated with increased core leakage is shown in Fig. 9. This effect can be significant, and points to the necessity of large, low leakage cores where lifetime is important. It is seen that increasing the leakage from 0.05 to 0.10 reduces the lifetime by almost 30% at a specific power of 1.0 Mw/kg.

Finally, the results on lifetime of making different assumptions regarding the fission-product cross sections are given in Figs. 10 through 13. In each of these cases, the cross sections or yields of all of the fission product pseudo-elements except xenon and the samarium group have been raised by the same factor. The cross sections and yields of xenon and the samarium group were held constant throughout the calculations.

One may conclude that the fission-product cross sections have a noticeable, but not decisive, effect on reactivity lifetime. Thus, in Fig. 12 it is seen that doubling all of the known fission-product thermal cross sections and resonance integrals would decrease the reactivity lifetime by 15% or less. Changes in the fission-product yields produce somewhat greater effects on lifetime. In Fig. 13, a doubling of all of the yields of the known fission products with significant cross sections decreases the reactivity lifetimes by up to 30%.

Attention has been focused on the number of fissions obtainable from each fissionable atom because this parameter has an important, and often determining, effect on fuel cycle costs. The lifetime influences or

determines the fuel fabrication cost, the uranium burnup cost, and the fuel reprocessing cost. Another design parameter which can be seen as important in the types of reactor being considered here is the ratio of peak-to-average power density. In any design the power density is limited by either fuel temperature or thermal stress in the fuel. In many designs one does not have complete flexibility in shifting fuel elements around, so that any increase in the peak-to-average power ratio means a proportional decrease in the average core power density. This, in turn, results in an increase in inventory costs and also in core size with associated capital costs. In Fig. 3 one can see that some of the cases with low thorium-to-uranium ratios give peak-to-average power ratios which must make them unacceptable even if they were otherwise attractive.

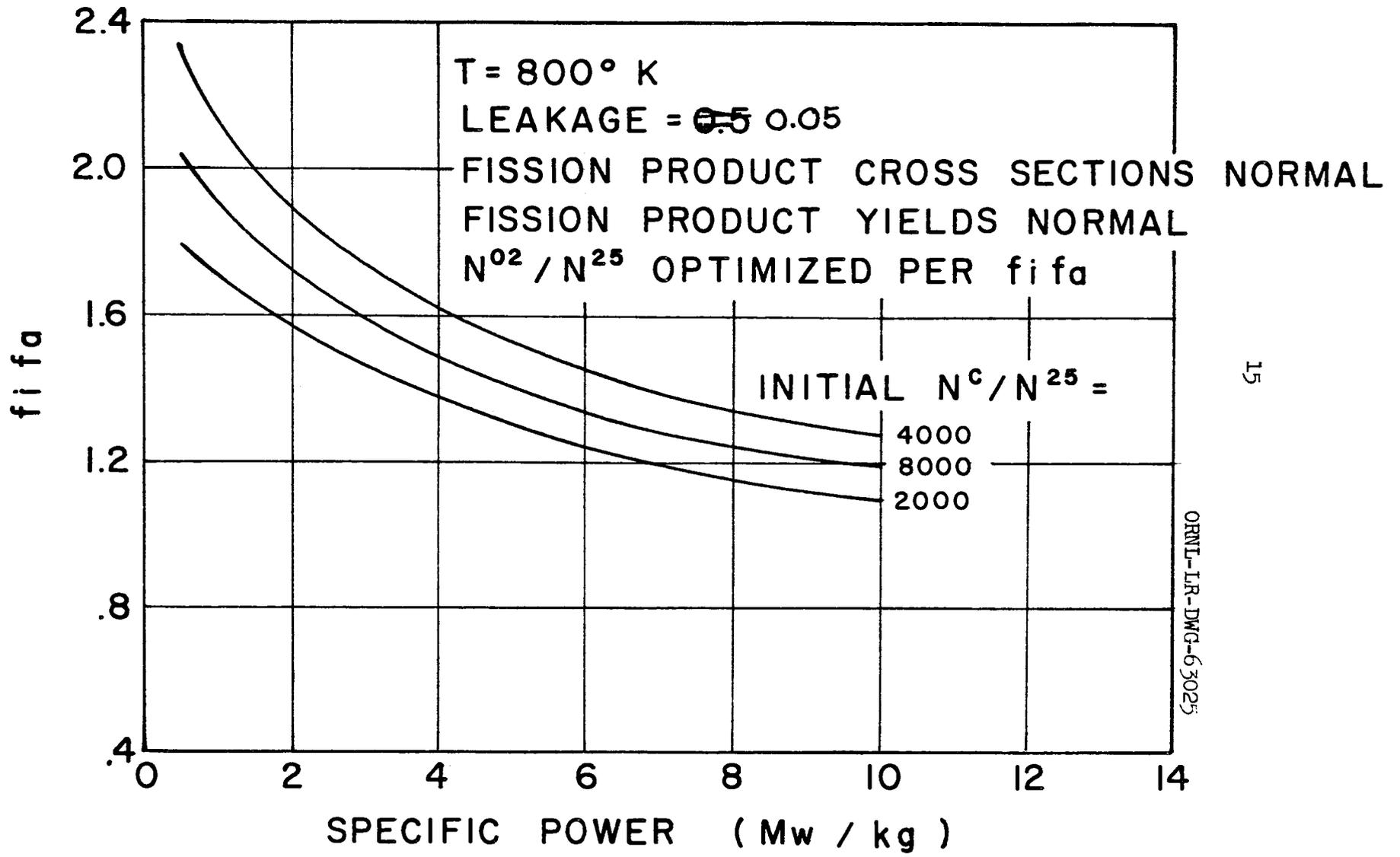
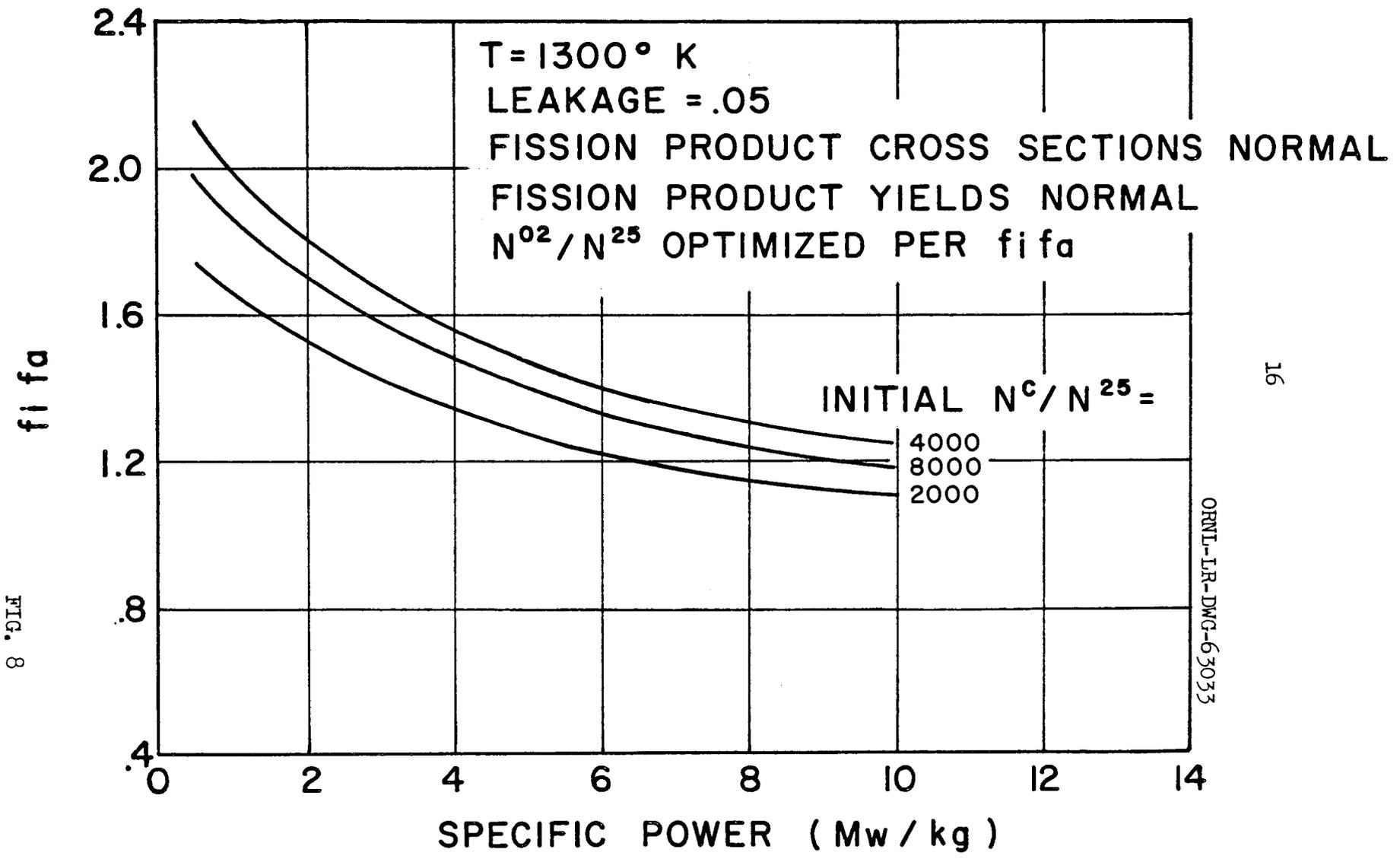


FIG. 7

FIG. 8



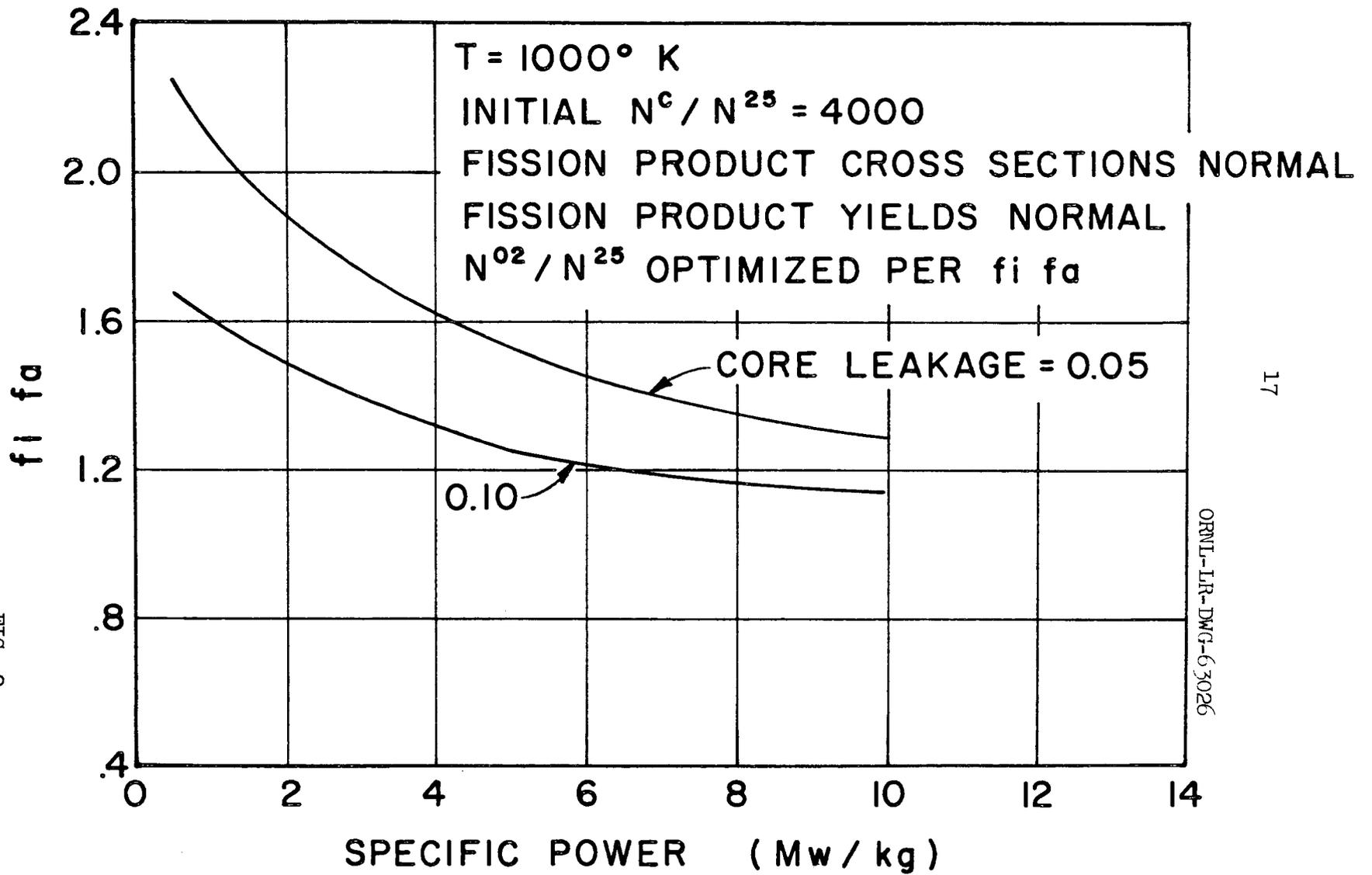
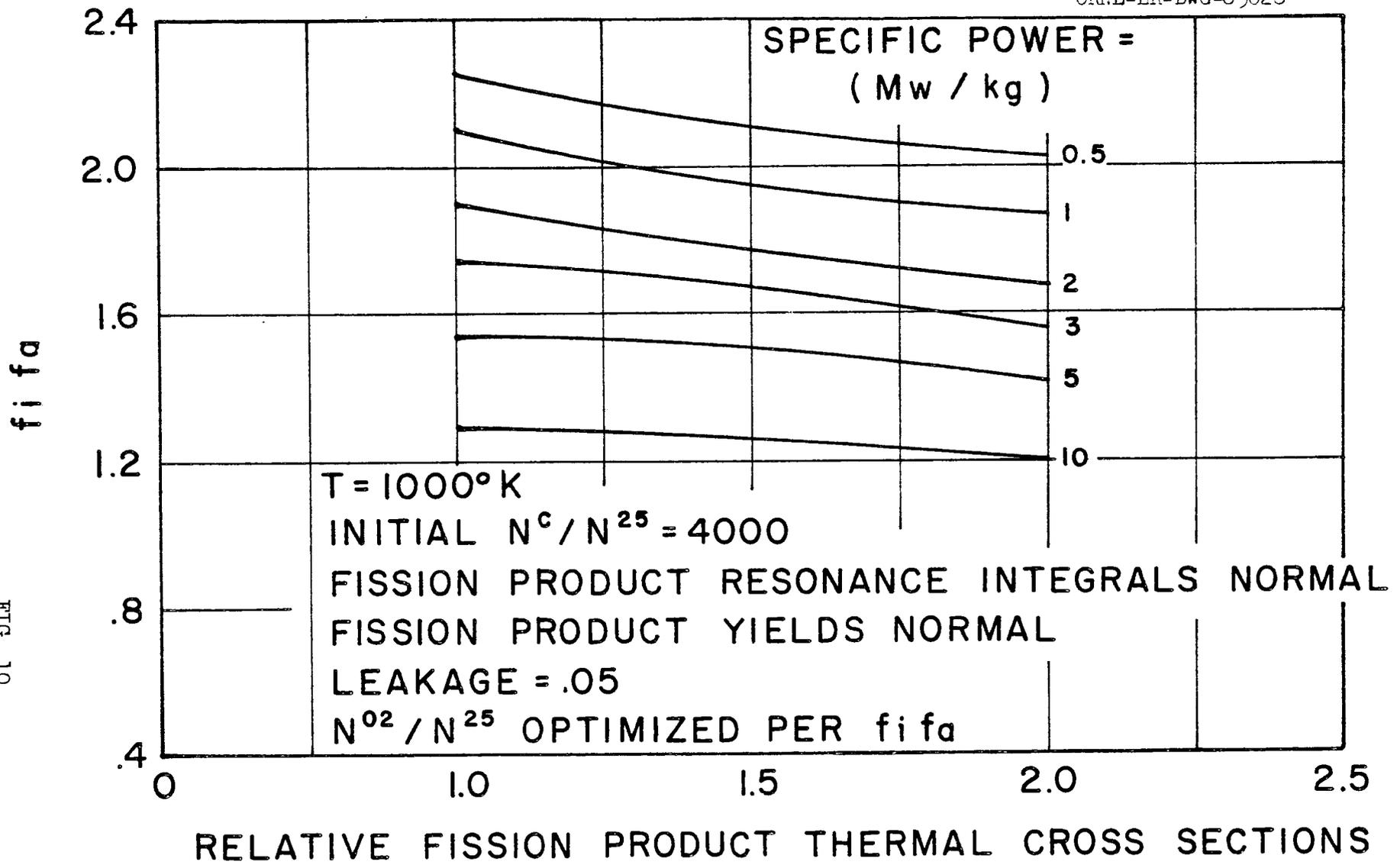


FIG. 9



T = 1000°K
INITIAL $N^C / N^{25} = 4000$
FISSION PRODUCT RESONANCE INTEGRALS NORMAL
FISSION PRODUCT YIELDS NORMAL
LEAKAGE = .05
 N^{02} / N^{25} OPTIMIZED PER $f_i f_a$

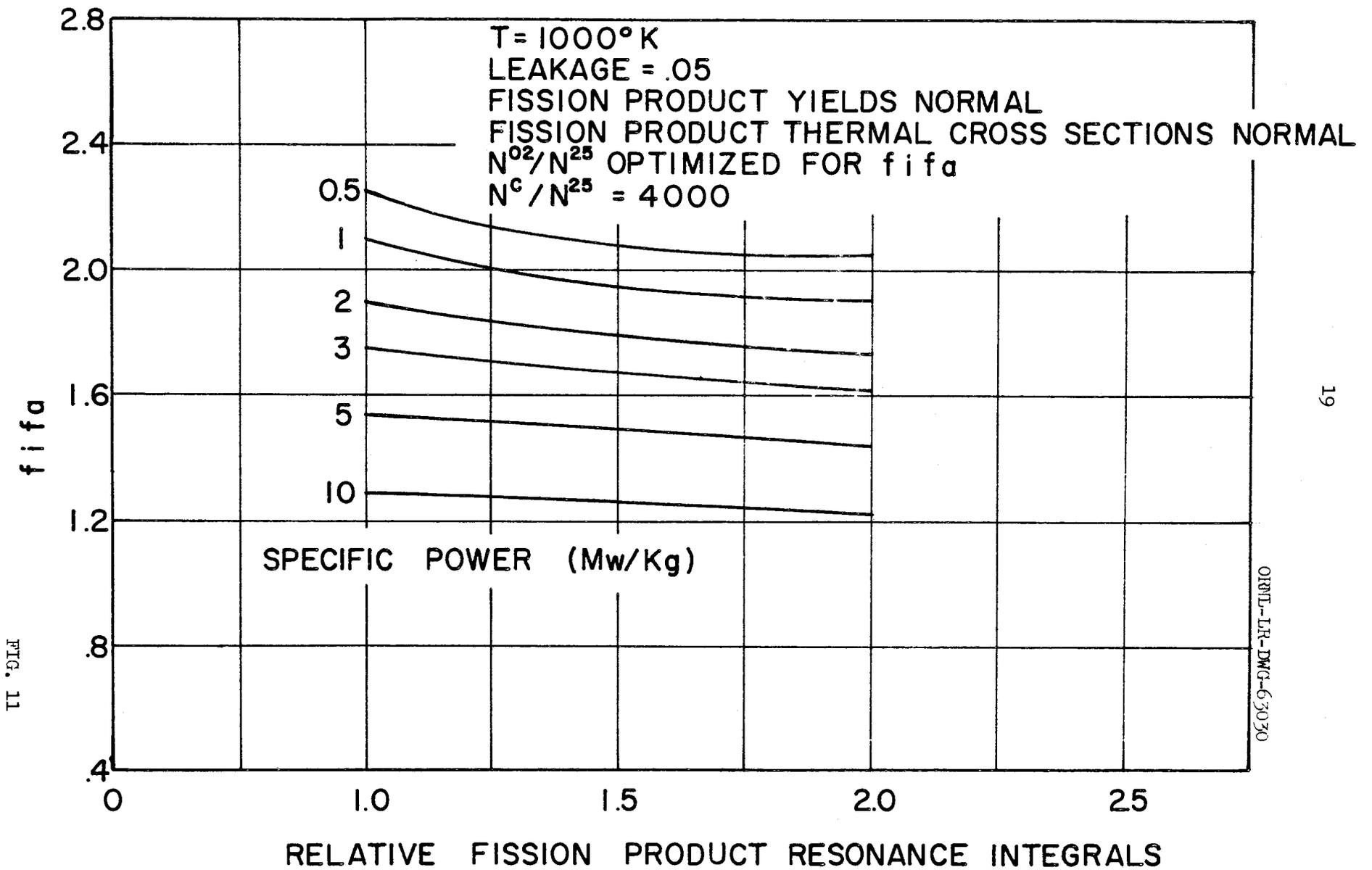


FIG. 11

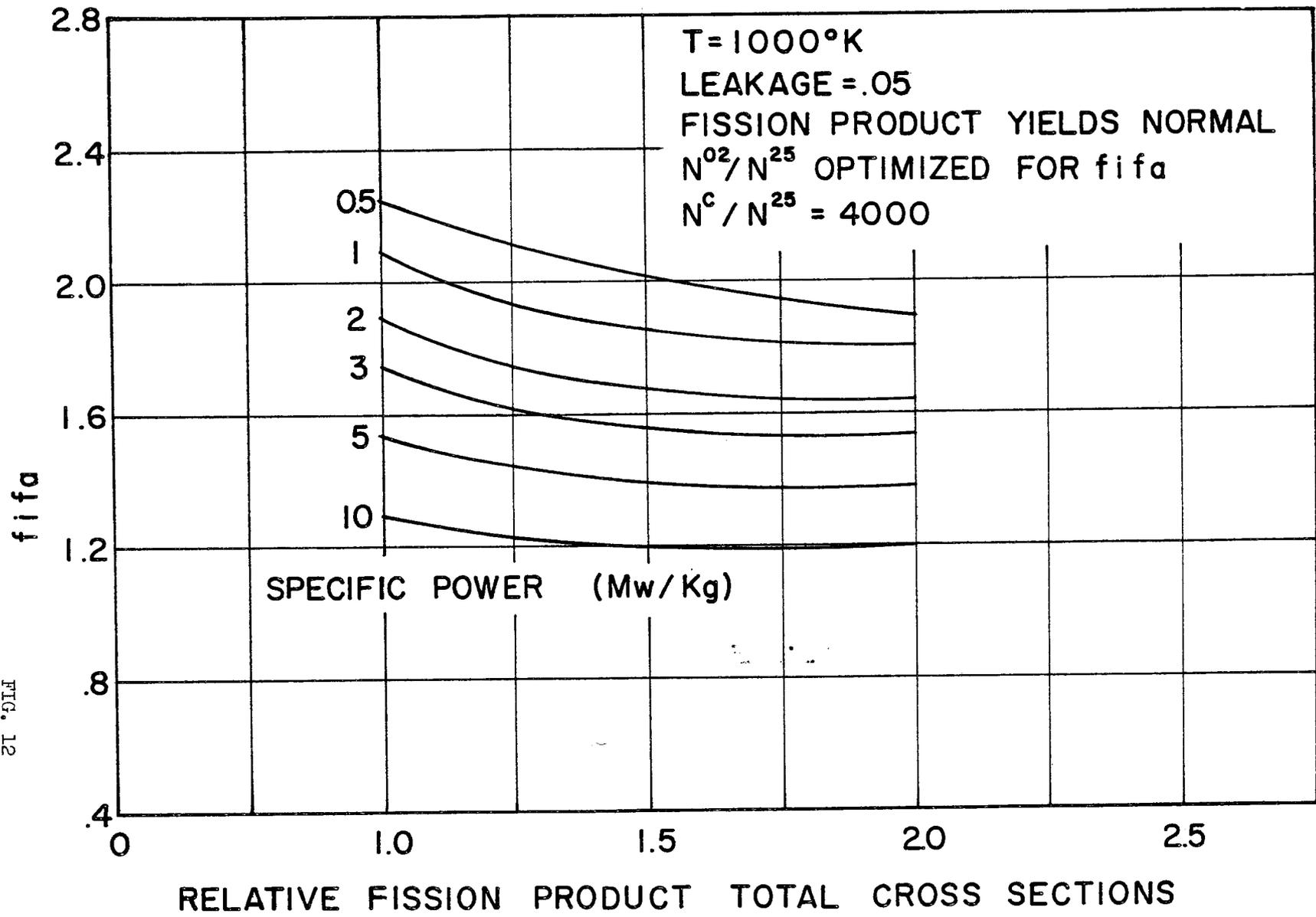


FIG. 12

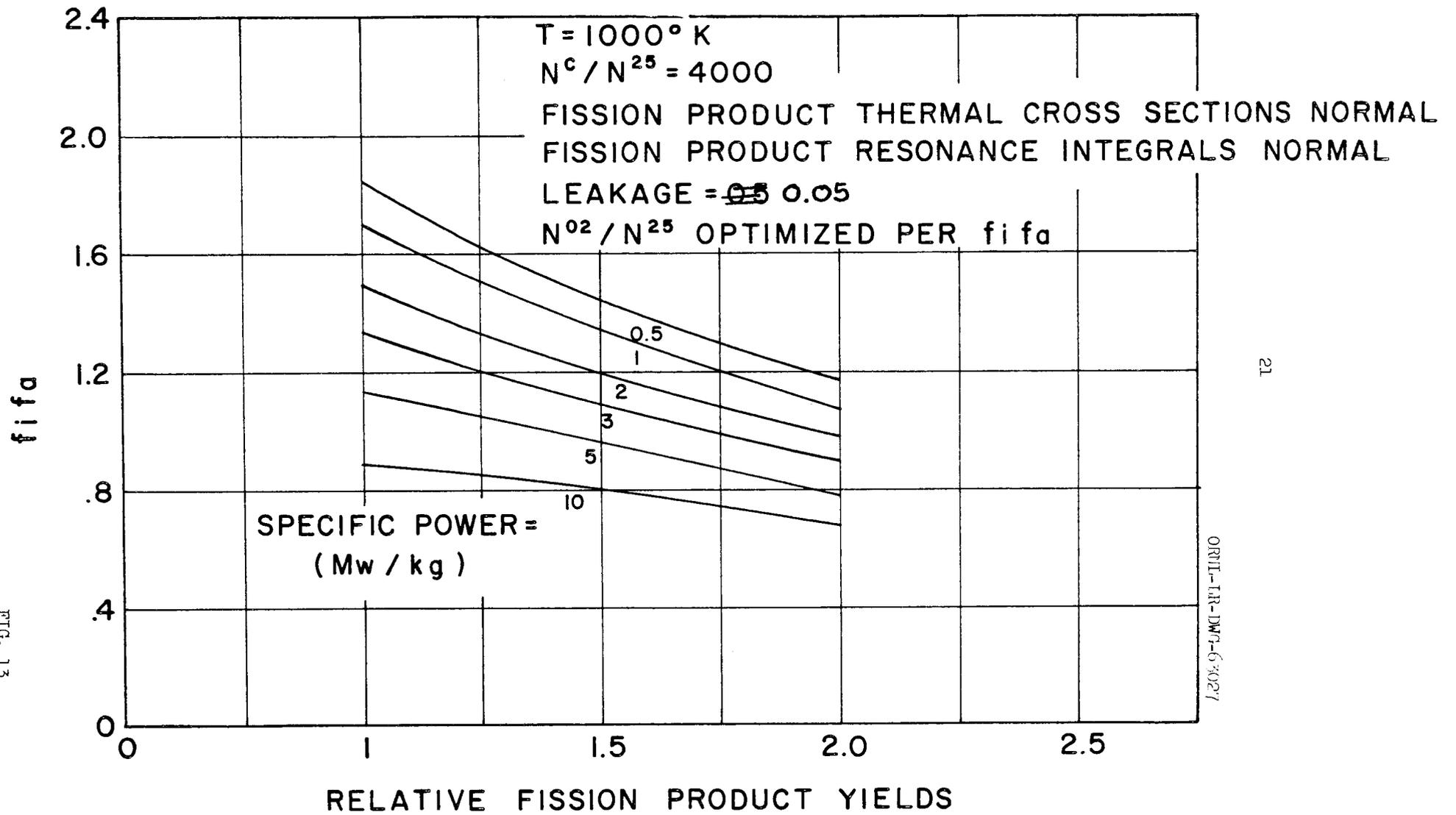


FIG. 13

APPENDIX A

Cross Section Calculations

1. Neutron Temperature

The relationship between moderator temperature and neutron temperature was that of Howland,⁴ who has deduced a modification to the Coveyou, Bate, and Osborn formula to fit graphite gas-model spectra to a Campbell-Freemantle model. The formula is

$$\frac{T_n}{T_m} = 1.0 + 1.4 \sigma_a \left(\frac{T_o}{T_m}\right)^{1/2},$$

where σ_a is the total absorption cross section at 2200 m/s per carbon atom, and T_o is 293.6°K. Different neutron temperatures were calculated for each carbon-to-uranium ratio, but the changes due to different thorium-to-uranium ratios were ignored.

2. U²³⁵

Contributions to thermal cross sections and resonance integrals were calculated from BNL-325 for $E \leq 0.4$ ev, using the ACS-1 code.⁵ Above 0.4 ev the experimentally determined values, $I_f = 271$ b⁶ and $I_a = 388$ b, were added.

3. U²³³

Contributions to thermal cross sections and resonance integrals were calculated from the data recommended by Nestor⁷ for the energy range $0.0001 \text{ ev} \leq E \leq 10^7 \text{ ev}$.

4. U²³⁴

Stoughton and Halperin's recommendation⁸ was used: $g\sigma_o = 95$ b and $I_a(E > 0.4 \text{ ev}) = 700$ b. This means that the total resonance integral is 747 b at a neutron temperature of 1140°K. The variation of cross sections with temperature was neglected.

5. U²³⁶

Stoughton and Halperin's recommendation⁸ was used: $g\sigma_0 = 66$ and $I_a(E > 0.4 \text{ ev}) = 400 \text{ b}$. This means that $I_a = 403 \text{ b}$ at a neutron temperature of 1140°K . The variation of cross sections with temperature was neglected.

6. Np²³⁷

The cross sections were obtained from the parameters given by Westcott:⁹ $g\sigma_0 = 245.5 \text{ b}$ and $I_a = 655.5 \text{ b}$ at a neutron temperature of 1140°K . The variation with temperature was neglected.

7. Th²³²

The thermal values were taken from BNL-325 ($\sigma_0 = 7.5 \text{ b}$) and Westcott⁹ ($g = 1.000 \text{ b}$), and were assumed to be independent of temperature. Doppler-broadened effective resonance integrals were taken from Blake.¹⁰ Values were specified for each of the fuel temperatures at each thorium-to-carbon ratio.

8. Xe¹³⁵

The thermal cross sections of S. Bernstein as reported by Westcott⁹ were used. It was assumed that the resonance integrals were negligible.

9. Sm¹⁴⁹

The cross sections were obtained from the parameters given by Westcott.⁹ The yield of the "Sm group" was defined to include the yields of the other high-cross-section fission products: Sm^{151} , Gd^{155} , Eu^{155} , Cd^{113} , and Gd^{157} . This gives a total of 1.66% yield for U^{235} fissions and 0.95% yield for U^{233} fissions.

10. The cross sections for the pseudo-elements representing the remainder of the fission products were taken from Nephew,² and adjusted for a Campbell-Freemantle type slowing-down spectrum in which

$$\begin{aligned} \phi(E) dE &= \frac{dE}{E} & E &\geq 7kT \\ &= \frac{1}{7kT} & 5kT &\leq E < 7kT \end{aligned}$$

$$\begin{aligned}\phi(E) dE &= \frac{E - 2kT}{21(kT)^2} & 2kT \leq E < 5kT \\ &= 0 & E < 2kT\end{aligned}$$

The values used were:

		<u>FP-1</u>	<u>FP-2</u>	<u>FP-3</u>	<u>FP-4</u>	
U^{233}	$g\sigma_0$	228.31	30.88	147.56	134.29	$\sum_1^4 yg\sigma_0 = 37.0$
	I_a	235.33	300.91	914.74	2883.43	$\sum_1^4 yI_a = 174.0$
	y	9.43%	18.1%	5.13%	1.75%	
U^{235}	$g\sigma_0$	216.81	31.52	140.94	129.15	$\sum_1^4 yg\sigma_0 = 43.0$
	I_a	186.33	299.87	980.50	2518.53	$\sum_1^4 yI_a = 215.3$
	y	11.24%	21.0%	5.82%	2.95%	

Nephew found that the cross sections of these pseudo-elements were only very slightly dependent on temperature or specific power. These effects were neglected.

11. Graphite

It was assumed that $g\sigma_0 = 0.004$ b, $g = 1.0$, $S = 0$. Hence, at 1140°K $I_a = 0.00183$. Temperature variation was ignored.

12. Pa^{233}

Stoughton and Halperin's recommendation⁸ was used: $g\sigma_0 = 70$ b and $I_a(E > 0.4 \text{ ev}) = 1200$ b. Thus, $I_a = 1235$ b.

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