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NEUTRON SOURCE STRENGTH IN FFTF FUEL AS A FUNCTION OF IRRADIATION

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I. INTRODUCTION

The neutron source strength associated with fuel irradiated in the FFTF reactor is important in determining the reactivity of the core when it is shut down or being refueled. The monitoring of subcritical reactivity for FFTF will be done with the Modified Source Multiplication (MSM) technique which requires an accurate knowledge of the neutron source strength in high exposure fuel elements replaced with fresh fuel elements in a shut down reactor.

As a means of obtaining the most accurate source evaluations possible with existing nuclear data for FFTF irradiated fuel, a technique for generating one-group actinide cross sections for use in isotopic buildup and depletion codes was developed. A further objective of the work was to use the cross sections and recently evaluated spontaneous fission and (α,n) reaction neutron yield data to calculate the neutron source strength in typical compositions of FFTF-type fuel as a function of flux level and irradiation time and then to compare the results with those of a previous study to assess the differences in the nuclear data. This paper will highlight the important aspects of this study and present some results.

II. NEUTRON SOURCES IN FFTF FUEL

In FFTF fuel the source neutrons are produced by certain isotopes which are both present in fresh fuel and built up by neutron irradiation. Source neutrons result predominantly from isotopes that either fission spontaneously or alpha decay and then induce (α,n) reactions with the oxygen isotopes in oxide fuel.

Initially, the fuel is composed of natural uranium plus plutonium isotopes from ^{238}Pu to ^{242}Pu. In addition, ^{241}Am is present as a result of the decay of the relatively short-lived ^{241}Pu. Irradiation of fuel in FFTF can produce a wide variety of additional

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SOURCE STRENGTH IN FFTF FUEL

Source neutrons result from:

1. Spontaneous fission of actinide isotopes.

2. Alpha particles emitted by actinide isotopes that react with isotopes ^{17}O and ^{18}O.

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A. ACTINIDES PRESENT IN UNIRRADIATED FUEL

U235, U238, Pu238, Pu239, Pu240, Pu241, Pu242,

$^{241}Pu \xrightarrow[14.8\ yrs]{\beta^-} {}^{241}Am$

B. ACTINIDES PRESENT IN IRRADIATED FUEL

URANIUM ISOTOPES:	U235 TO U239
NEPTUNIUM ISOTOPES:	Np236 TO Np239
PLUTONIUM ISOTOPES:	Pu236 TO Pu244
AMERICIUM ISOTOPES:	Am240 TO Am244
CURIUM ISOTOPES:	Cm240 TO Cm246

isotopes. Some of these have substantially larger neutron yields than the initial isotopes. Hence it is found that the build up of even trace quantities of these isotopes results in a substantial increase in the neutron source strength.

III. SOURCE DENSITY

The neutron source density at some location in the fuel as a function of time is related to the isotopic composition of the fuel by Equation 1. Here, $S_{TOT} = \Sigma S_i$, is the total neutron source density at a point, where S_i is the contribution to the source density produced by isotope i. The quantity $S_i = \rho_i Y_i$, where ρ_i is the density of isotope i at some time, and Y_i is the total neutron yield for isotope i. Equation 2 shows that Y_i is equal to the sum of the spontaneous fission and (α,n) yields for isotope i. In this study the change in the isotopic density of each isotope as a function of irradiation was calculated with the Oak Ridge isotope depletion and buildup code ORIGEN. In addition, the neutron yield terms were treated as input parameters in the code SOURCE, used to evaluate the source strength.

IV. NEUTRON YIELDS

Measured yields are not available in the literature for all isotopes that can contribute to the neutron source. Therefore, it was necessary to refer to available nuclear data in order to evaluate the yield terms for each isotope whose yields have not been measured.

A. Spontaneous Fission Yields

Neutron yields for spontaneous fission were evaluated by D. L. Johnson of HEDL for thirty-three isotopes from ^{235}U to ^{246}Cm. The general trend is for spontaneous fission yield terms to increase substantially when one goes to heavier elements. Table 1 shows some calculated spontaneous fission yields and a comparison of these yields for a few isotopes for which measured yields also exist. In general, the calculated data exhibit deviations of up to 15% relative to the measured data.

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NEUTRON SOURCE DENSITY

$$S_{TOT} = \sum_i S_i = \sum_i \rho_i Y_i \qquad (1)$$

$$Y_i = Y_i(S.F.) + Y_i(\alpha,n) \qquad (2)$$

S_{TOT} = TOTAL NEUTRON SOURCE DENSITY (n/cm^2-sec)

$S_i = \rho_i Y_i$ = SOURCE DENSITY PRODUCED BY ISOTOPE i OF DENSITY ρ_i (gm/cm^3)

Y_i = TOTAL NEUTRON YIELD (n/gm-sec) OF ISOTOPE i

$Y_i(S.F.)$ = SPONTANEOUS FISSION YIELD OF ISOTOPE i

$Y_i(\alpha,n)$ = ALPHA-NEUTRON REACTION YIELD OF ISOTOPE i

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TABLE 1

SPONTANEOUS FISSION YIELDS (n/g-sec)

Isotope	Calculated Yields	Measured Yields	% Maximum Deviation
Pu238	2.197 (+3)	2.54±0.32+(3)	13.0
Pu239	2.082 (-2)	(2.259) (-2)	8.0
Pu240	8.95 (+2)	(8.95±0.81)(+2)	9.0
Pu242	1.716 (+3)	(1.577±0.039)(+3)	10.0
Am241	5.686 (-1)	None	-
Cm242	1.969 (+7)	(1.967) (+7)	0.1
Cm244	(1.159) (+7)	(1.151±0.069)(+7)	7.0

B. (α,n) Reaction Yields

The neutron yields from spontaneous (α,n) reactions were evaluated for the same thirty-three isotopes ranging from ^{235}U to ^{246}Cm by Ombrellaro and Johnson of HEDL.

Table 2 shows a comparison of the (α,n) yield for a few isotopes for which measured yields also exist. In general, the calculated data exhibit maximum derivations of 10% relative to the measured data.

V. ANALYTIC TECHNIQUE USED FOR DEPLETION AND BUILDUP CALCULATIONS

The computer code ORIGEN, used for calculating the depletion and buildup of isotopes in FFTF fuel as a function of irradiation, solves a set of coupled differential equations represented by Equation 1 of Figure 1.

The first term on the right represents the loss of isotope i by radioactive decay, where λ_i is the decay constant of isotope i. The second term represents the loss of isotope i by neutron absorption, where σ_i is the effective absorption cross section (including fission).

Increases in isotope i due to radioactive decay of other isotopes j are represented by the third term. Here, contributions from beta decay, alpha decay, electron capture and isomeric transitions were included wherever they were significant.

Increases in the isotope i due to neutron-induced reactions on other isotopes are represented by the sum of terms

$$\sum_k \sigma_{ki} \phi N_k.$$

For these, contributions from (n,γ) and (n,2n) reactions are included where they might be significant and can be evaluated.

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TABLE 2

(α,n) REACTION YIELD (n/g-sec) IN FFTF FUEL AND OTHER MATERIALS

Isotope	Calculated Yields	Measured Yields	% Maximum Deviation
Pu238	1.396 (+4)	1.4 (+4)	0.4
Pu239	4.060 (+1)	3.8 (+1)	7.0
Pu240	1.520 (+2)	1.45 (+2)	5.0
Pu242	2.116 (+0)	2.13 (+0)	0.8
Am241	2.790 (+3)	2.68 (+3)	4.0
Cm242	3.83 (+6)	-	
Cm244	8.026 (+4)	-	

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$$\frac{dN_i}{dt} = -N_i\lambda_i - N_i\sigma_i\phi + \sum_j \lambda_j iN_j + \sum_k \sigma_{ki}\phi N_k \qquad (1)$$

N_i = ATOM DENSITY ISOTOPE i

$N_i\lambda_i$ = LOSS OF ISOTOPE i DUE TO RADIOACTIVE DECAY

$N_i\sigma_i\phi$ = LOSS OF ISOTOPE i BY NEUTRON ABSORPTION

$\sum_i \lambda_{ji} N_j$ = INCREASE IN ISOTOPE i DUE TO RADIOACTIVE DECAY OF ISOTOPE j

$\sum_k \sigma_{ki}\phi N_k$ = INCREASE IN ISOTOPE i BY NEUTRON-INDUCED REACTIONS IN ISOTOPES k

Figure 1. Isotope Buildup and Depletion Equation.

It should be noted that effective cross sections used here were obtained for particular neutron spectra at specific locations in the reactor. This was done so that the spatial variations in the depletion and buildup calculations would take into account variations in both the total flux and the spectrum.

VI. INPUT NUCLEAR DATA

In calculations on FTR fuel performed in 1975 at HEDL by Boroughs, Bunch and Johnson, effective cross sections were derived mainly from ENDF/B-IV and used in the code ALCHEMY. The effective cross sections were generated by averaging multigroup cross sections with a neutron spectrum believed to be representative of the equilibrium spectrum in the center of the core.

In the present study, energy-dependent cross sections were obtained primarily from ENDF/B-V. These were processed by R. E. Schenter and F. M. Mann into 42-group constants using the ETOX code. The 42-group cross sections were then processed through the 1DX code to correct for self-shielding effects and to evaluate the elastic removal cross sections. The 1DX model was developed in terms of homogenized zones, each representing one of the six rows of subassemblies in the FFTF core, and includes temperatures, dimensions and atom densities corresponding to full power conditions.

In each zone of the 1DX mode, the 42-group cross sections were collapsed to 12 groups using the equilibrium spectrum of the zone and thereby producing a set of 12-energy group cross sections for each zone. Effective one-group cross sections for capture, fission, and (n,2n) reactions appropriate for ORIGEN were obtained by averaging the 12-group cross section sets with 12-group fluxes obtained from three-dimensional FFTF core calculations. The three-dimensional flux calculations were performed for three core configurations; namely, beginning of life (BOL), beginning of cycle 4 (BOC-4) and end of cycle 4 (EOC-4).

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TABLE 3

CROSS SECTION EVALUATION PROCESS

ETOX - PRODUCED 42-GROUP ACTINIDE CROSS SECTIONS BASED ON ENDF/B-V DATA

1DX - 42-GROUP CROSS SECTIONS WERE SELF-SHIELDED AND TEMPERATURE-CORRECTED TO FULL POWER CONDITIONS AND THEN COLLAPSED TO 12 GROUPS

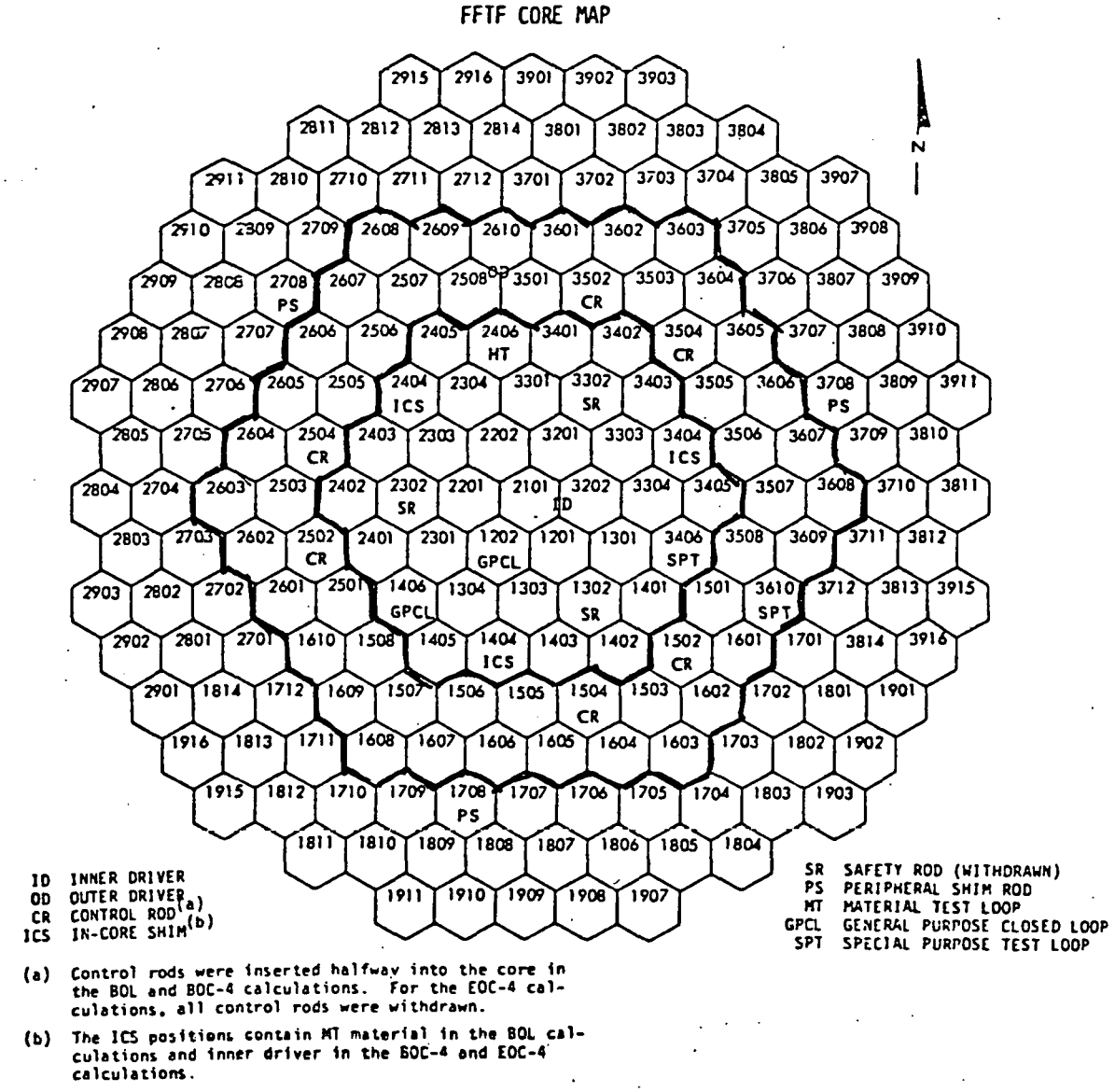
BURN - 12-GROUP CROSS SECTIONS WERE AVERAGED WITH THREE-DIMENSIONAL, 12-GROUP FLUXES REPRESENTATIVE OF BOL, BOC-4 AND EOC-4 CONDITIONS TO OBTAIN ONE-GROUP EFFECTIVE CROSS SECTIONS FOR ORIGEN

To clarify the details of the procedure we refer to the core map of Figure 2 which represents the midplane configuration of Core 1 in hexagonal geometry. The set of 91 subassemblies in the core contains 27 inner driver and 46 outer driver fuel subassemblies. Each fuel subassembly was zoned separately, allowing the radial and axial dependence of the fuel burnup rate to be accounted for by subassembly. In this manner, the fuel subassemblies were represented by 73 different zones. BOL, BOC-4 and EOC-4 effective one-group cross sections for each zone were obtained by averaging the 12-group cross sections with the three-dimensional, 12-group fluxes. The results of such calculations showed that the midplane values of the cross sections are suitable for use over much of the axial region. Therefore, by this calculation and selection process, three sets of one-group, midplane, cross sections for each of the 73 zones were obtained.

To minimize the number of cross sections for general depletion and buildup calculations, the cross sections in each set were spatially averaged to provide BOL, BOC-4 and EOC-4 effective cross section sets for the inner driver, and outer driver row 5 and row 6 regions.

Figure 3 shows the results of using BOL inner driver cross sections in ORIGEN. As the mixed oxide fuel of FFTF is exposed to high neutron flux levels for long periods of time, the build up of trace quantities of heavy actinide isotopes will occur. Moreover, as shown in the figure, except for ^{242}Am concentrations, the ENDF/B-V based cross sections generally predict larger concentrations than those calculated with the earlier set of effective cross sections based on ENDF/B-IV data.

Typical results of source buildup evaluations based on BOL cross sections are shown in Figure 4 for irradiation of FFTF inner driver fuel for up to 300 days in a flux of 5×10^{15} n/cm^2-sec.



(a) Control rods were inserted halfway into the core in the BOL and BOC-4 calculations. For the EOC-4 calculations, all control rods were withdrawn.

(b) The ICS positions contain MT material in the BOL calculations and inner driver in the BOC-4 and EOC-4 calculations.

Figure 2. Core configuration of FFTF Core 1.

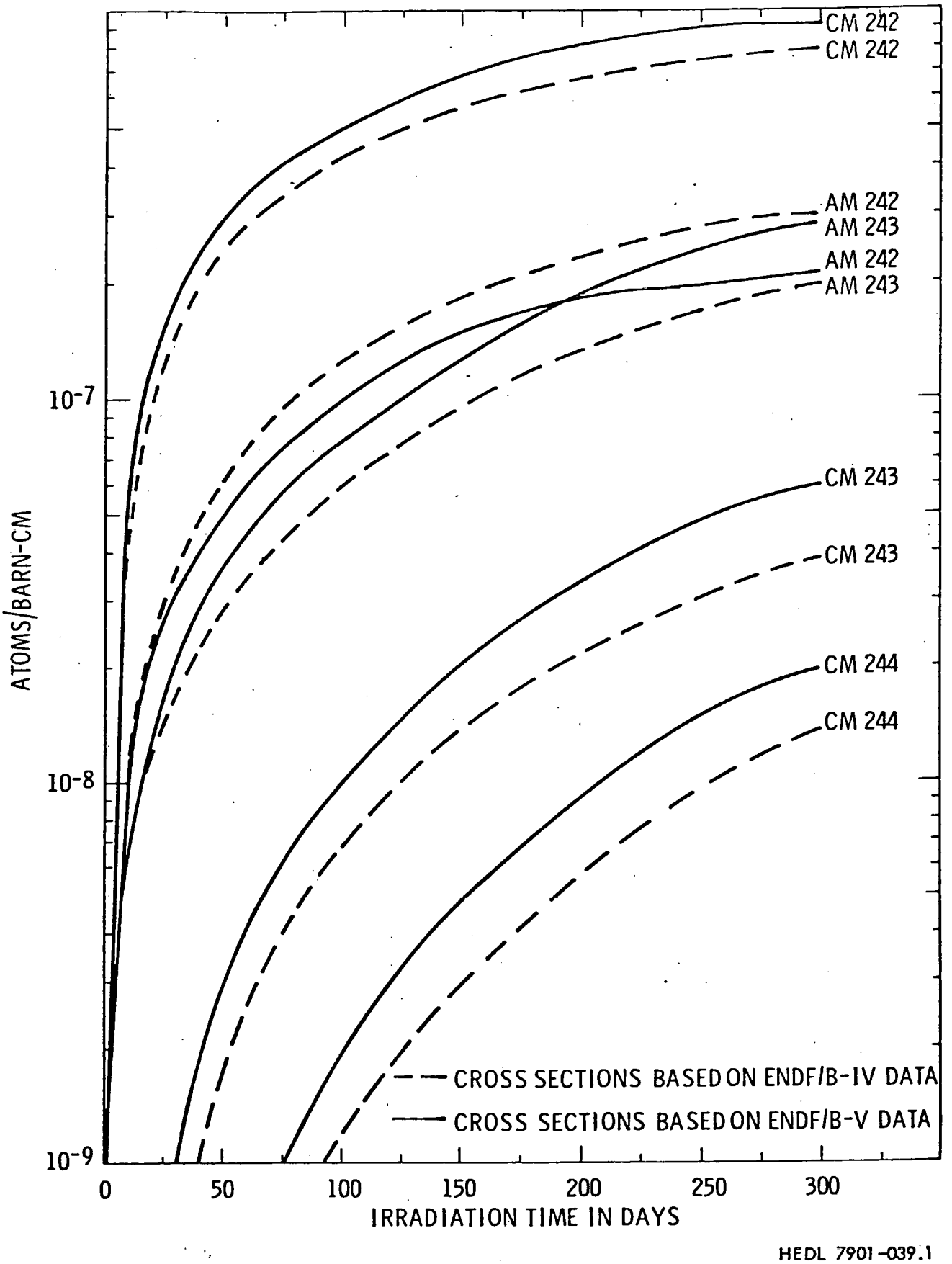


Figure 3. Plots of Buildup of Some Important Isotopes in FFTF Inner Driver Fuel Exposed to $7X(10^{15})$ n/cm^2-sec Flux.

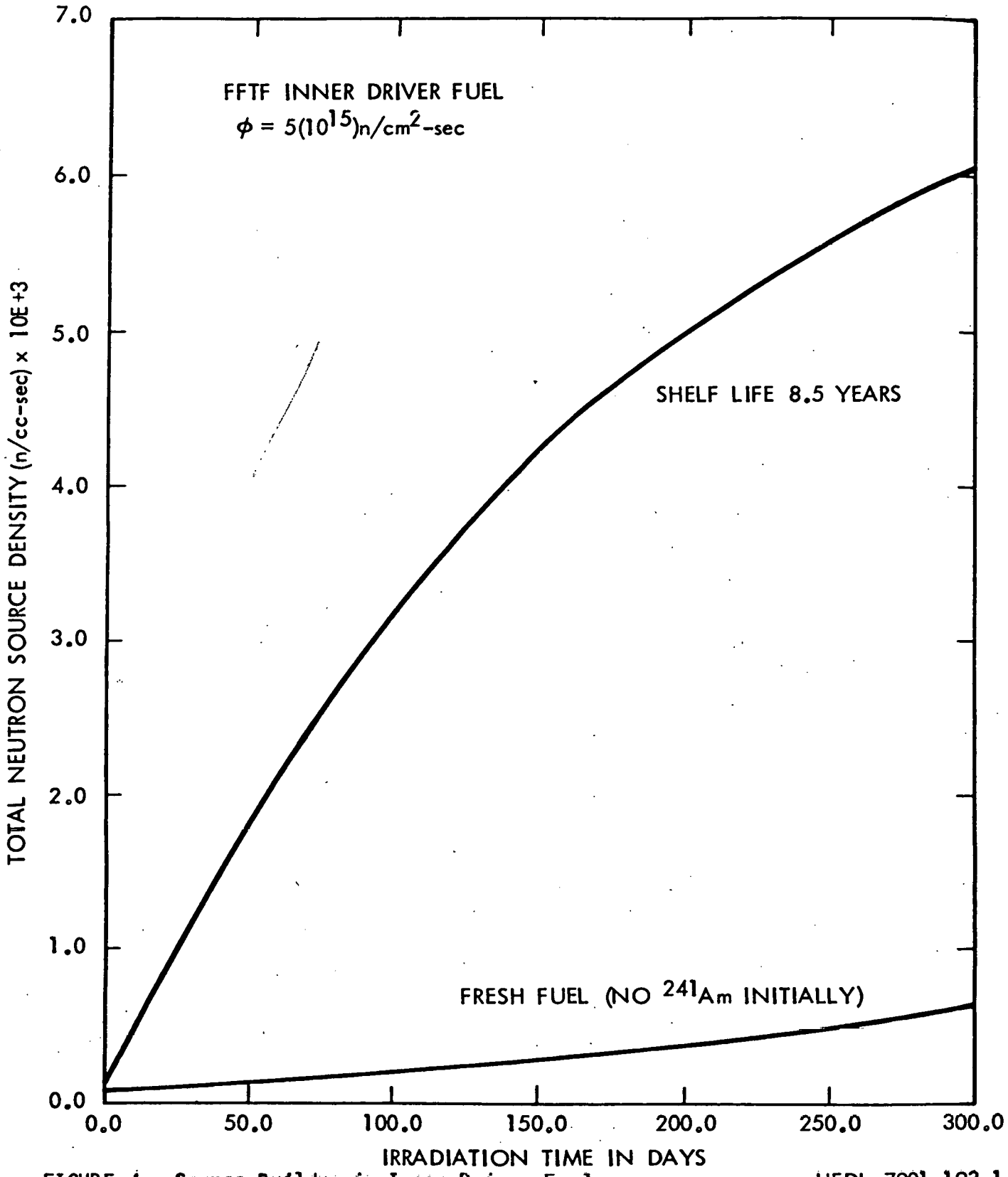


FIGURE 4. Source Buildup in Inner Driver Fuel. HEDL 7901-193.1

The two curves differ in shelf life only. The lower curve represents the source buildup for a fuel composition immediately after chemical separation; the top curve corresponds to the same composition having a shelf life of 8.5 years. The 8.5 year life corresponds closest to that expected for the actual FFTF fuel at startup.

Note that the increase in source density rate as a function of shelf life is dependent on the amount of ^{241}Am present at the beginning of the irradiation. The source increases much faster when ^{241}Am is present initially because ^{242}Cm is produced rapidly by $^{241}Am(n,\gamma)^{242g}Am$ reaction and ^{242g}Am subsequently decays by β^- emission to ^{242}Cm. The evaluations of ENDF/B-V cross sections by Schenter show that the most significant change from ENDF/B-IV cross sections occurs in ^{241}Am, which is important to the build up of ^{242}Cm.

The build up of ^{242}Cm dominates the build up of the neutron sources as shown by Figure 5 where the percent contribution to the total source strength by each of seven isotopes is plotted as a function of time for FFTF inner driver fuel in a flux of $5x10^{15}$ n/cm^2-sec.

It is noted that 30 days after irradiation has begun the source contribution from ^{242}Cm increases by more than 90% of the total source. In Figure 5, the ^{244}Cm also builds up rapidly, but not as rapidly as ^{242}Cm. Thus, of particular interest in the build up of heavy actinides are the isotopes of ^{242}Cm and ^{244}Cm because of their extremely high spontaneous fission and (α,n) activity.

VII. NEUTRON SOURCE BUILDUP VERSUS IRRADIATION

Figure 6 shows the inherent neutron source rate in FFTF driver fuel after three full (100 day) exposures. The depletion and buildup calculations were performed as follows. For the first cycle, 0 to 100 days, BOL cross sections were used. For the second cycle (100-200 days) the BOC-4 cross sections were used. Finally, in the third cycle (200-300 days) the EOC-4 cross sections were used.

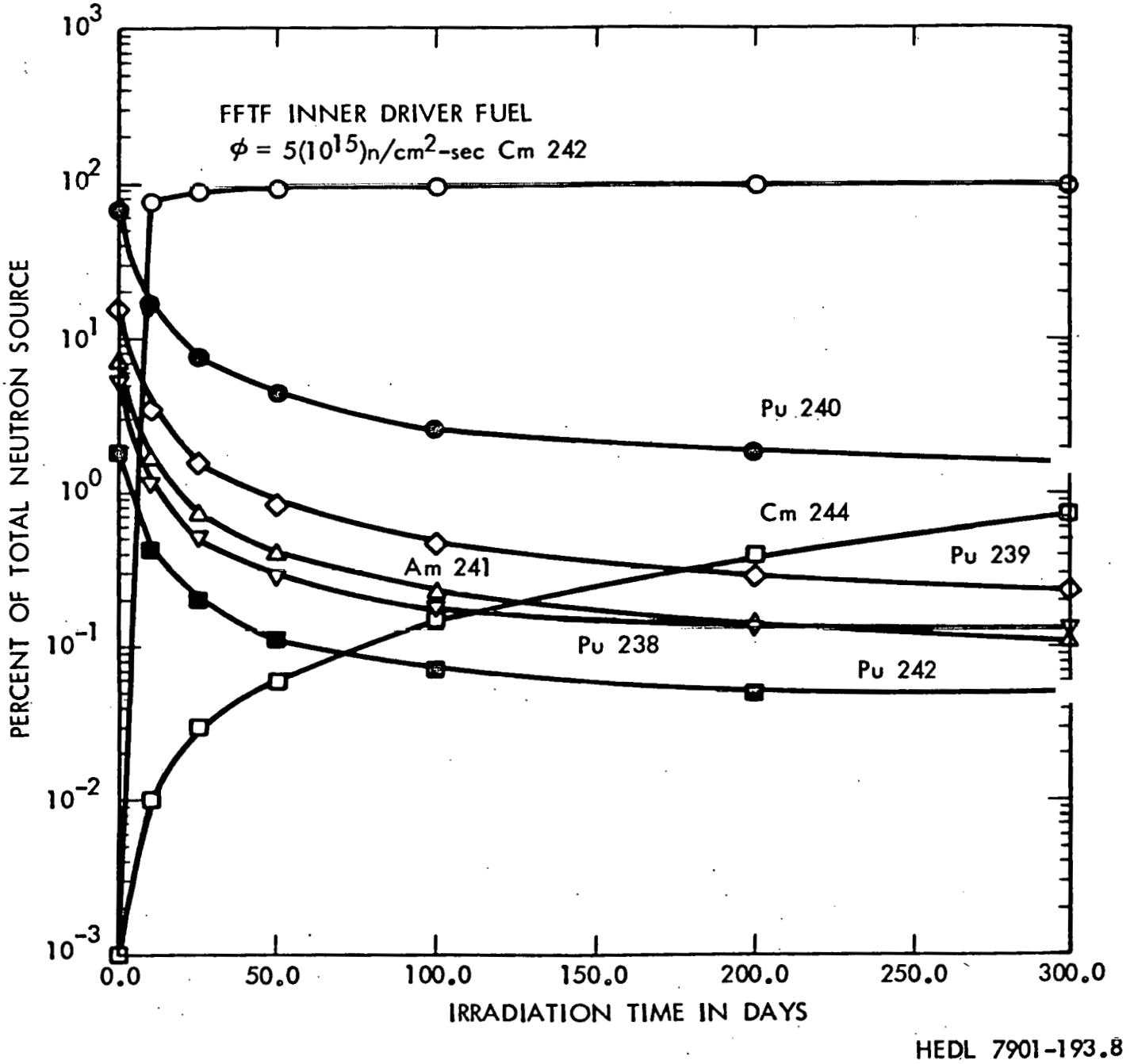


FIGURE 5. Percent Contribution of Isotope Neutron Source to the Total Neutron Source Strength Versus Irradiation Time.

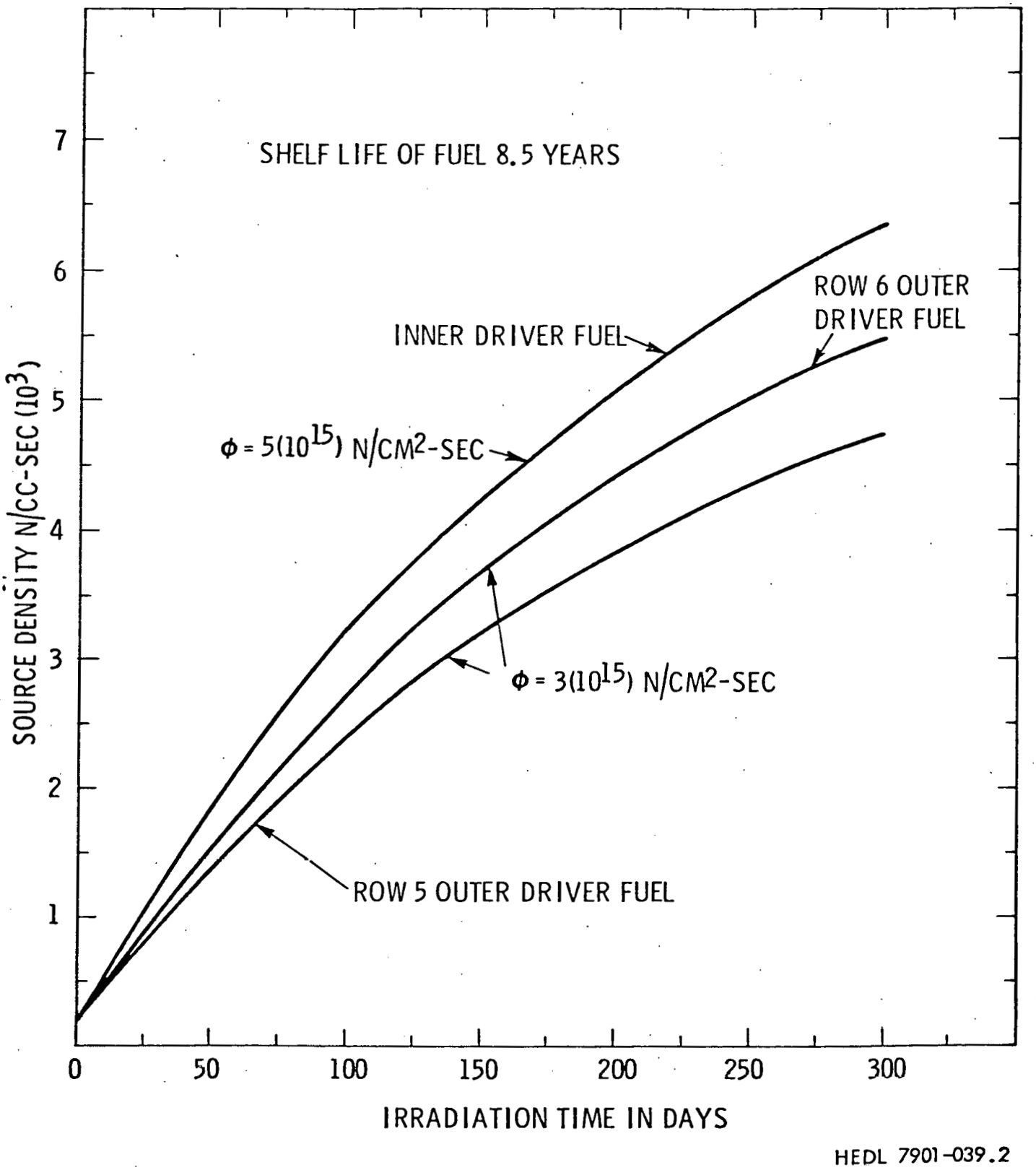


Figure 6. Source Buildup Versus Irradiation Time.

The results of these calculations show that the inherent neutron source in an FFTF inner driver after full exposure (100 days) is approximately six times larger than that of a fresh driver fuel element. The total core source density at the end of three cycles of operation prior to refueling is approximately two times that at the end of cycle one (100 days).

In summary, the important improvements which have been made here have been in obtaining improved values of cross sections for the less common isotopes and accounting for fuel temperature effects, energy self-shielding effects and the effect of space-energy flux variations during irradiation on the effective cross sections. Except for the concentrations of ^{242}Am, the cross sections generally predict larger isotopic concentrations in fuel than those calculated with an earlier set of effective cross sections based on ENDF/B-IV data. The evaluation of ENDF/B-V cross sections shows that the most significant change from ENDF/B-IV cross sections concerns the reactions in ^{241}Am. The changes are important for buildup calculations since ^{241}Am produces ^{242}Am by neutron capture which lies in the direct path to the production of high inventories of ^{242}Cm.