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# ISOTOPIC ABUNDANCE AND THE VARIATION OF $\eta$ IN IRRADIATED NUCLEAR FUELS CONSISTING INITIALLY OF COMBINATIONS OF Th^{232}, U^{238}, and U^{235}

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# ISOTOPIC ABUNDANCE AND THE VARIATION OF $\eta$ IN IRRADIATED NUCLEAR FUELS CONSISTING INITIALLY OF COMBINATIONS OF Th^{23\,2}, U^{238}, and U^{235}

by

J. C. Carter and R. C. Howard

#### ABSTRACT

The abundances of the significant isotopes and the variation of  $\eta$  during the irradiation of a nuclear fuel, which initially consists of combinations of U<sup>235</sup>, U<sup>238</sup>, and Th<sup>232</sup>, is presented.

#### INTRODUCTION

This paper on the irradiation of a nuclear fuel consisting initially of combinations of the isotopes  $U^{235}$ ,  $U^{238}$ , and  $Th^{232}$  has been prepared in connection with studies of reactor economics. The characteristics of the fuel as a function of irradiation time are presented. It is intended that the designer apply the information to specific reactor designs.

The principal products of a reactor are thermal energy and isotopes resulting from the irradiation of the fuel. Since the attainment of the optimum production of each of these is not necessarily concomitant, successful reactor operation will depend upon a carefully planned fuel-loading program, taking into consideration the characteristics of the fuel during irradiation in the specific reactor and the current prices of power and of isotopes.

It would be quite a difficult task to determine the optimum initial combination of isotopes for a specific reactor without exploring an area of interest encompassing the combinations which are possible. The preparation of such areas of interest involves the solution of the equations for isotopic abundance and reactivity as a function of time and presenting the results in parametric form.

Economical use of nuclear fuel depends upon the achievement of high fuel burnup and this, in turn, depends upon the maintenance of a critical condition. Initial combinations of isotopes which, by means of conversion and breeding, maintain reactivity nearly constant for a long time are most desirable. As the fissionable isotopes resulting from the irradiation of the initial fuel build up, their fissions contribute to the supply of fast neutrons. At any time (t) there is a continuous evolution (growth and burnup) for each of the numerous isotopes involved. The rate of change in abundance of each is determined by its own particular nuclear constants. The net amounts of each at any time is found by a simultaneous solution of the equations of isotopic abundance as a function of irradiation time.

It is assumed that the resonance escape probability (p) and the fast fission factor ( $\epsilon$ ) do not change during the fuel cycle, since the cross sections do not vary appreciably over the range of irradiation studies. Therefore, the change in reactivity is expressed in terms of the ratio  $\eta_f$  to  $\eta_0 f_0$ .

The number of fast fission neutrons emitted per thermal neutron absorbed,  $\eta$ , is a function only of the initial fuel composition and its resultant products, but the thermal utilization (f) is dependent upon reactor structural materials and can be varied by design.

The assumption was made that, for a given resonance escape probability, the resonance absorption by the  $U^{238}$  and  $Th^{232}$  was divided according to the fraction of each present in the fuel. This, of course, is not accurate for a homogeneous mixture, because the thorium resonance integral is lower than the uranium resonance integral. For a heterogeneous system, the resonance absorption of a given amount of  $U^{238}$  or  $Th^{232}$  is a function of geometry as well as of concentration and, therefore, depends on the design of each individual reactor. The choice made for dividing the resonance capture is an arbitrary one which is probably adequate for the survey purposes that this analysis is supposed to serve. It is likely that this error has no greater effect than arises from the error in fission product and plutonium cross sections.

The studies have been concerned with irradiation up to 40,000 MWD/ ton at neutron fluxes of the order of  $10^{13}$  cm<sup>-2</sup>sec<sup>-1</sup>. The range of enrichment is from natural uranium to enriched thorium: Th<sup>232</sup>/U<sup>235</sup> = 70.

#### CALCULATIONS

All calculations were made for a point in the reactor at which the fuel and moderator were considered homogeneous, and the neutron temperature was related to moderator temperature through hardening typical of slightly enriched reactors.

The power was considered to be held constant throughout; therefore, the flux was treated as a function of time in the solution of the simultaneous differential equations that define isotopic abundance. The chain of isotopes considered is that shown in the accompanying design.



The variation in reactivity of the fuel and the abundance of an isotope at any time t is found from the simultaneous solution of the following equations. Here  $\eta$  is defined as the number of neutrons produced per thermal neutron absorbed in the fuel:

The product of the two variables  $\Sigma_f$  and  $\phi(t)$  was held constant. The variation of  $\Sigma_a$ ,  $\Sigma_f$ ,  $\phi$  and  $\eta$  as a function of irradiation time was determined from the following differential equations which define the rate of change of isotopes due to absorption, decay, fission, leakage, and resonance capture:

$$\frac{\mathrm{dTh}^{232}}{\mathrm{dt}} = -\left[\mathrm{Th}^{232}\sigma_{\mathrm{a}} + \mu(1-W)\left\{\mathrm{U}^{233}\sigma_{\mathrm{a}}\eta + \mathrm{U}^{235}\sigma_{\mathrm{a}}\eta + \mathrm{Pu}^{239}\sigma_{\mathrm{a}}\eta + \mathrm{Pu}^{241}\sigma_{\mathrm{a}}\eta\right\}\right]\phi(t) \tag{1}$$

$$\frac{\mathrm{d}\mathrm{Pa}^{233}}{\mathrm{d}t} = \left[ \mathrm{Th}^{232} \sigma_{\mathrm{a}} + \mu (1-W) \left\{ \mathrm{U}^{233} \sigma_{\mathrm{a}} \eta + \mathrm{U}^{235} \sigma_{\mathrm{a}} \eta + \mathrm{Pu}^{239} \sigma_{\mathrm{a}} \eta + \mathrm{Pu}^{241} \sigma_{\mathrm{a}} \eta \right\} \right] \phi(t) - \mathrm{Pa}^{233} \lambda - \mathrm{Pa}^{233} \sigma_{\mathrm{a}} \phi(t) \quad (2)$$

$$\frac{dU^{233}}{dt} = Pa^{233}\lambda - U^{233}\sigma_a \phi(t)$$
(3)

$$\frac{dU^{234}}{dt} = U^{233}\sigma_{c} \phi(t) + Pa^{233}\sigma_{a} \phi(t) - U^{234}\sigma_{a} \phi(t)$$
(4)

$$\frac{\mathrm{d}U^{235}}{\mathrm{d}t} = U^{234}\sigma_{c} \phi(t) - U^{235}\sigma_{a} \phi(t)$$
(5)

$$\frac{\mathrm{d}U^{238}}{\mathrm{d}t} = -\left[U^{238}\sigma_{a} + \mu W \left\{U^{233}\sigma_{a}\eta + U^{235}\sigma_{c}\eta + \mathrm{Pu}^{239}\sigma_{a}\eta + \mathrm{Pu}^{241}\sigma_{a}\eta\right\}\right]\phi(t)$$
(6)

$$\frac{\mathrm{dNp}^{239}}{\mathrm{dt}} = \left[ U^{238} \sigma_{a} + \mu W \left\{ U^{233} \sigma_{a} \eta + U^{235} \sigma_{a} \eta + \mathrm{Pu}^{239} \sigma_{a} \eta + \mathrm{Pu}^{241} \sigma_{a} \eta \right\} \right] \phi(t) - \mathrm{Np}^{239} \lambda$$
(7)

$$\frac{\mathrm{dPu}^{239}}{\mathrm{dt}} = \mathrm{Np}^{239}\lambda - \mathrm{Pu}^{239}\sigma_{\mathrm{a}}\phi(t)$$
(8)

$$\frac{\mathrm{d}\mathrm{Pu}^{240}}{\mathrm{d}t} = \mathrm{Pu}^{239}\sigma_{\mathrm{c}}\phi(t) - \mathrm{Pu}^{240}\sigma_{\mathrm{a}}\phi(t)$$
(9)

$$\frac{dPu^{241}}{dt} = Pu^{240}\sigma_{c}\phi(t) - Pu^{241}\sigma_{a}\phi(t)$$
(10)

$$\frac{\mathrm{d}\,\mathrm{B}^{10}}{\mathrm{d}t} = -\,\mathrm{B}^{10}\sigma_{\mathrm{a}}\phi(t) \tag{11}$$

$$\frac{\mathrm{dF}}{\mathrm{dt}} = \mathbf{N} - \mathbf{F}\sigma_{\mathbf{a}}\phi(\mathbf{t}) \tag{12}$$

$$\phi(t) = \frac{K}{U^{233}\sigma_{f} + U^{235}\sigma_{f} + Pu^{239}\sigma_{f} + Pu^{241}\sigma_{f}}$$
(13)

The symbols employed are as follows:

${ m Th}$	Thorium atoms per cubic centimeter
U	Uranium atoms per cubic centimeter
$\mathbf{Pu}$	Plutonium atoms per cubic centimeter
Pa	Protoactinium atoms per cubic centimeter
Np	Neptunium atoms per cubic centimeter
В	Boron atoms per cubic centimeter
N	Integrated fissions per ton of fissionable material
F	Total number of fission product nuclei

- $\phi$  Neutron flux
- $\sigma_a$  Microscopic cross section for absorption
- $\sigma_c$  Microscopic cross section for capture
- $\sigma_{f}$  Microscopic cross section for fission
- $\Sigma_{\mathbf{f}}$  Macroscopic cross section for fission
- $\Sigma_{a}$  Macroscopic cross section for absorption
  - $\lambda$  Decay constant
  - $\nu$  Neutrons produced per thermal neutron fission
  - $\eta$  The average number of neutrons liberated for each neutron absorbed in the fuel
  - $\mu \in (1-p) L$
  - $\epsilon$  Fast fission factor
  - p Resonance escape probability
- L Neutron leakage
- W Per cent of uranium by weight (it is assumed that resonance captures in  $U^{238}$  and  $Th^{232}$  are proportional to the weight per cent of  $U^{238}$  and  $Th^{232}$  respectively)
- K A constant proportional to specific power
- t Time

 $\mu$  is taken as a parameter, since the combination of terms which it represents are singular to each reactor. The size, geometry, fuel arrangement, and moderator characteristics all affect its value.

Since the cross section of the aggregate fission products is questionable, the results with and without this cross section are presented to give some idea of how much the fission products can affect the results.

The equations for isotopic abundances and  $\eta$  were solved with the aid of an analog computer. The analog diagram of the equations is shown on the following page.



#### NUCLEAR CONSTANTS

The nuclear constants used in the computations are those generally acceptable at the time (1956) the work was done and are given in the table below. The value for the aggregate cross section of the fission products was obtained by combining fission yields with cross sections.

	U <sup>233</sup>	U <sup>234</sup>	U <sup>235</sup>	U <sup>238</sup>	Th <sup>232</sup>	Pa <sup>233</sup>	Pu <sup>239</sup>	Pu <sup>240*</sup>	Pu <sup>241</sup>	Np	Fission Products
σ <sub>a</sub>	349	55	403	1.64	4.17	150	1103	400	1360		50
$\sigma_{\mathbf{f}}$	318	-	340				733		960		
σc	31	55	63	1.64	4.17	150	370	400	400		
ν	2.31		208				1 912		2.06		
η	2.54		2.46				2.88		2.91		
λ						2.94				3.42	

# NUCLEAR CONSTANTS AT A NEUTRON TEMPERATURE OF 650°K (This allows for some hardening)

#### RESULTS

The results of this work are presented as curves of  $\eta$  and isotopic abundance versus the irradiation time of fuel in the reactor, with  $\mu$  serving as a parameter.

The isotopes which were considered to be significant are shown on Figures 1 through 9a. The fuel was assumed to be initially composed of Th<sup>232</sup>, U<sup>235</sup>, and U<sup>238</sup>, but provision was made on the analog computer to study the effect of a burnable poison (B<sup>10</sup>) if so desired. In the thorium-uranium system, U<sup>235</sup> is the primary source, and Th<sup>232</sup> and U<sup>238</sup> are secondary sources of neutrons. There are many branching reactions during the irradiation, but only those which lead to a significant production of Pu<sup>241</sup> in the interval of 40,000 MWD/ton are considered.

The variation of  $\eta$  with irradiation time is shown on Figures 10 to 30a.

These two sets of curves are intended to be used as an aid in deciding which combination of isotopes initially in the fuel will most likely give the desired reactivity and isotopic content at some time (t) or during some interval ( $\Delta t$ ).

Certain combinations of enrichment and  $\mu$  shown on the graphs are incompatible in the critical core; however, these combinations may occur in a blanket.

The calculations were based on the assumption of a constant specific power. In the event that the neutron flux is other than that shown in the parametric presentation, the irradiation time scale may be varied inversely as the flux ratio. This procedure is not strictly correct because the variation with time of two different initial fluxes is not necessarily in phase. For small variations in flux, the discrepancy is considered negligible. Interpolation between enrichments gives satisfactory results. A list of values obtained in the parametric study follows.

Atom % Th <sup>232</sup> /U <sup>235</sup> /U <sup>238</sup>	Atom Ratio $\frac{\text{Th}^{232}}{\text{U}^{235}}$	Wt % NAT U	Fig. No.
0/0.73/99.27	0	100	l-la
39.61/1.41/58.98	40	60	2-2a
39.82/1.21/58.98	50	60	3-3a
39.93/1.09/58.98	60	60	4-4a
40.03/0.99/58.98	70	60	5-5a
78.50/2.10/19.40	40	20	6-6a
78.82/1.71/19.47	50	20	7-7a
79.12/1.46/19.42	60	20	8-8a
79.27/1.27/19.46	70	20	9-9a

#### ISOTOPIC ABUNDANCE

Atom $\frac{C_0}{10}$ Th <sup>232</sup> /U <sup>235</sup> /U <sup>238</sup>	Variations in $\eta$	Wt % NAT U	Fig. No.
0/0 73/99 27	0	100	10-10a
97 56/2.44/0	40	0	ll-lla
78.20/2.40/19.40	40	20	12-12a
59.11/1.76/39.13	40	40	13 <b>-</b> 13a
39.61/1.41/58.98	40	60	14-14a
19 90/1.06/79.09	40	80	15 <b>-</b> 15a
98.04/1.96/0	50	0	16-16a
78.82/1.71/19.97	50	20	17-17a
59.28/1.47/39.25	50	40	18-18a
39.80/1.22/58.98	50	60	19 <b>-</b> 19a
20.00/0.96/79.04	50	80	20-20a
98.36/1.64/0	60	0	21-21a
79.12/1.46/19.42	60	70	22-22a
	Atom Ratio $\frac{\mathrm{Th}^{232}}{\mathrm{U}^{235}}$		
59.60/1.27/39.13	60	40	23-23a
39.93/1.09/58.98	60	60	24-24a
20.07/0.90/79.03	60	80	25-25a
98.59/1.41/0	70	0	26-26a
79.27/1.27/19.46	70	20	27-27a
59.75/1.13/39.12	70	40	28 <b>-</b> 28a
40.03/0.99/58.98	70	60	29 <b>-</b> 29a
20.12/0.85/79.03	70	80	30-30a

#### CONCLUSIONS

1. By varying the atom ratio of a fuel consisting of  $\mathrm{Th}^{232}$ ,  $\mathrm{U}^{235}$  and  $\mathrm{U}^{238}$  it is possible to attain, for a specific reactor, an efficient use of the original fuel.

2. Removal of fission products during the operation greatly increases the lifetime of a fuel.

3. For systems using Th<sup>232</sup> as fertile material, very large reactivity lifetimes are possible for an initial conversion ratio significantly less than the theoretical maximum ( $\eta$ -1) as compared with systems using U<sup>238</sup> only as fertile material.

4. The characteristics of the  $\eta$  variations in a Th<sup>232</sup>-U<sup>235</sup> fuel are almost a mirror image of the  $\eta$  variations in U<sup>238</sup>-U<sup>235</sup> fuel. Therefore, it is concluded that a reactor which is loaded with a mixture of natural uranium and thorium can have a flat curve of reactivity versus irradiation time over its lifetime. The lifetime would then be determined theoretically by irradiation damage.

5. It is desirable to have as low an enrichment and as high a value of  $[\mu = \epsilon(1-p)L]$  as is consistent with good design, where L is the neutron leakage probability.



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