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ANL-5335

Physics and Mathematics

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THERMAL NEUTRON IRRADIATION OF NATURAL THORIUM

by

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Price \$ 0.35

Available from the
Office of Technical Services
Department of Commerce
Washington 25, D. C.

July 30, 1954

METALLURGY DIVISION

CLASSIFICATION CANCELLED
DATE 2-7-57
For The Atomic Energy Commission
H. H. Canale
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INTRODUCTION

In many experiments involving the thermal neutron irradiation of natural thorium, it is often necessary to estimate the magnitude of the following quantities:

1. The burnup of natural thorium resulting from the fission of uranium-233.
2. The heat generated in natural thorium as a result of the fission of uranium-233.
3. The concentration of thorium-233, protactinium-233, uranium-233 and uranium-234 in natural thorium after exposure to neutrons.

Because it is usually impractical to measure these quantities experimentally, it is frequently necessary to rely upon the results of a theoretical investigation. Therefore, an attempt has been made to evaluate quantitatively the principal nuclear reactions occurring in the thermal neutron irradiation of thorium.

A number of useful graphs and tables are presented to provide assistance in the planning and evaluating of experiments pertaining to the neutron irradiation of thorium.

DISCUSSION OF THEORY

If natural thorium is exposed to a source of thermal neutrons, a series of nuclear reactions occurs resulting in the formation of uranium-233 and a variety of other nuclear products. Uranium-233 is of considerable interest in that it possesses a relatively large nuclear cross section for fission with thermal neutrons.

Exposure of thorium-232 to thermal neutrons results in the formation of thorium-233. However, thorium-233 has a relatively short half-life for radioactive disintegration and decays quite rapidly to protactinium-233. Accordingly, protactinium-233 decays to the final product uranium-233. The principal nuclear reactions occurring in the thermal neutron irradiation of natural thorium are represented symbolically in Figure 1.

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The nuclear reactions described in Figure 1 are represented mathematically by a system of linear differential equations in Figure 2. The solutions of the system of equations are given in Figure 3. An equation for calculating the burnup of natural thorium resulting only from the fission of uranium-233 is presented in Figure 4. It is assumed for mathematical simplicity that the supply of thorium-232 remains constant throughout the irradiation.

DISCUSSION OF RESULTS

Table 1 lists values of the thermal neutron cross sections and nuclear half-lives used in solving the equations in Figures 3 and 4.

Figure 5 describes graphically the formation of thorium-233 and Figures 6 and 7 the formation of protactinium-233 during exposure of thorium to neutrons. The concentration of thorium-233 increases quite rapidly and approaches an equilibrium concentration after two hours of irradiation. The concentration of protactinium-233 increases much more slowly and approaches an equilibrium concentration after 160 days of irradiation.

Figure 8 illustrates the buildup of uranium-233 during exposure of thorium to various thermal neutron fluxes. The concentration of uranium-233 increases until it reaches an equilibrium concentration and is then burned up at a rate equal to that at which it is being produced. Although the equilibrium concentration of uranium-233 is lowered as a result of an increase in the neutron flux, equilibrium is approached much more rapidly.

After the thorium is removed from the source of neutrons, the concentration of uranium-233 again increases to another maximum because of the radioactive decay of protactinium-233 into uranium-233. Figure 9 illustrates the radioactive decay of protactinium-233 and the formation of uranium-233. These curves are of use in estimating the amount of uranium-233 formed by radioactive decay of protactinium-233 subsequent to irradiation.

The burnup of thorium resulting from the fission of uranium-233 is shown graphically in Figure 10. It is directly related to the formation and buildup of fission products and is not concerned by the disappearance of thorium by conversion to uranium-234. The curves are derived from the burnup equation for natural thorium, Figure 4.

Figure 11 describes the buildup of uranium-234 in thorium during exposure to thermal neutrons.

Table 2 lists the secular equilibrium concentrations of thorium-233, protactinium-233, uranium-233 and uranium-234.

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Figures 12 and 13 exhibit the specific power of natural thorium resulting only from the thermal neutron fission of uranium-233. The heat generated by other means, such as γ -rays, is not considered. Gamma-ray heat production depends only on location in a reactor and maximum γ -heating usually occurs in locations of maximum thermal neutron flux. For example, the specific power of thorium due to the absorption of reactor γ 's in the Materials Testing Reactor may be as great as 5 watts/gram.¹ Table 3 lists the maximum values of the specific power of thorium. These maxima are for secular equilibrium concentrations of uranium-233.

REFERENCES

- ¹Buck, J. M., and Leyse, C. F., Materials Testing Reactor Project Handbook, ORNL 963, section 4.5.2, May, 1951.

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FIG. 1

PRINCIPAL NUCLEAR REACTIONS OCCURRING IN THE THERMAL
NEUTRON IRRADIATION OF NATURAL THORIUM

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$$(1) \quad \frac{dN_1}{dt} = \alpha_0 N_0 - \alpha_1 N_1 \quad (t = 0, N_1 = 0)$$

$$(2) \quad \frac{dN_2}{dt} = \lambda_1 N_1 - \alpha_2 N_2 \quad (t = 0, N_2 = 0)$$

$$(3) \quad \frac{dN_3}{dt} = \lambda_2 N_2 - \alpha_3 N_3 \quad (t = 0, N_3 = 0)$$

$$(4) \quad \frac{dN_4}{dt} = \sigma_2 \phi N_2 + \sigma_3 \phi N_3 - \alpha_4 N_4 \quad (t = 0, N_4 = 0)$$

$N_0, N_1, N_2, N_3,$ and N_4 = Number of $\text{Th}^{232}, \text{Th}^{233}, \text{Pa}^{233}, \text{U}^{233}$ and U^{234} atoms per unit volume of thorium, respectively, at time, t .

$\lambda_0, \lambda_1, \lambda_2, \lambda_3,$ and λ_4 = Radioactive decay constant of $\text{Th}^{232}, \text{Th}^{233}, \text{Pa}^{233}, \text{U}^{233}$ and U^{234} , respectively.

$\sigma_0, \sigma_1, \sigma_2,$ and σ_4 = Thermal neutron absorption cross section of $\text{Th}^{232}, \text{Th}^{233}, \text{Pa}^{233}$ and U^{234} , respectively.

σ_3 = Non-Fission thermal neutron absorption cross section of U^{233} .

$\alpha_0, \alpha_1, \alpha_2, \alpha_3,$ and α_4 = Sum of radioactive decay constant, λ , and pseudo-decay constant, $\phi\sigma$, of $\text{Th}^{232}, \text{Th}^{233}, \text{Pa}^{233}, \text{U}^{233}$ and U^{234} , respectively. (i.e. $\alpha_1 = \lambda_1 + \phi\sigma_1$)

ϕ = Thermal neutron flux.

FIG. 2

MATHEMATICAL REPRESENTATION OF THE NUCLEAR REACTIONS OCCURRING IN THE NEUTRON IRRADIATION OF THORIUM

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$$(1) \quad N_1(t) = \frac{k}{\alpha_1} (1 - e^{-\alpha_1 t})$$

$$(2) \quad N_2(t) = \frac{k\lambda_1}{\alpha_1} \left[\frac{1}{\alpha_1 - \alpha_2} e^{-\alpha_1 t} - \frac{\alpha_1}{\alpha_2(\alpha_1 - \alpha_2)} e^{-\alpha_2 t} + \frac{1}{\alpha_2} \right]$$

$$(3) \quad N_3(t) = k\lambda_1\lambda_2 \left[-\frac{1}{\alpha_1(\alpha_1 - \alpha_2)(\alpha_1 - \alpha_3)} e^{-\alpha_1 t} + \frac{1}{\alpha_2(\alpha_1 - \alpha_2)(\alpha_2 - \alpha_3)} e^{-\alpha_2 t} - \frac{1}{\alpha_3(\alpha_1 - \alpha_3)(\alpha_2 - \alpha_3)} e^{-\alpha_3 t} + \frac{1}{\alpha_1\alpha_2\alpha_3} \right]$$

$$(4) \quad N_4(t) = \tau\phi^2 \left[\frac{1}{\alpha_1(\alpha_1 - \alpha_2)(\alpha_1 - \alpha_3)(\alpha_1 - \alpha_4)} e^{-\alpha_1 t} - \frac{1}{\alpha_2(\alpha_1 - \alpha_2)(\alpha_2 - \alpha_3)(\alpha_2 - \alpha_4)} e^{-\alpha_2 t} + \frac{1}{\alpha_3(\alpha_1 - \alpha_3)(\alpha_2 - \alpha_3)(\alpha_3 - \alpha_4)} e^{-\alpha_3 t} \right. \\ \left. - \frac{\alpha_1^2 - \alpha_1(\alpha_2 + \alpha_3) + \alpha_2\alpha_3}{\alpha_4(\alpha_1 - \alpha_2)(\alpha_1 - \alpha_3)(\alpha_1 - \alpha_4)(\alpha_2 - \alpha_4)(\alpha_3 - \alpha_4)} e^{-\alpha_4 t} + \frac{1}{\alpha_1\alpha_2\alpha_3\alpha_4} \right] + \rho\phi^2 \left[-\frac{1}{\alpha_1(\alpha_1 - \alpha_2)(\alpha_1 - \alpha_4)} e^{-\alpha_1 t} \right. \\ \left. + \frac{1}{\alpha_2(\alpha_1 - \alpha_2)(\alpha_2 - \alpha_4)} e^{-\alpha_2 t} - \frac{1}{(\alpha_1 - \alpha_4)(\alpha_2 - \alpha_4)\alpha_4} e^{-\alpha_4 t} + \frac{1}{\alpha_1\alpha_2\alpha_4} \right]$$

where $k = \sigma_0\phi N_0$, $\tau = \sigma_0\sigma_3\lambda_1\lambda_2 N_0$ and $\rho = \sigma_0\sigma_2\lambda_1 N_0$.

FIG. 3

SOLUTIONS OF THE SYSTEM OF EQUATIONS REPRESENTING THE NUCLEAR REACTIONS
OCCURRING IN THE NEUTRON IRRADIATION OF THORIUM

$$B(t) = \sigma_0 \sigma_f \lambda_1 \lambda_2 N_0 \phi^2 \left[\frac{-1}{\alpha_1^2 (\alpha_1 - \alpha_2) (\alpha_1 - \alpha_3)} (1 - e^{-\alpha_1 t}) + \frac{1}{\alpha_2^2 (\alpha_1 - \alpha_2) (\alpha_2 - \alpha_3)} (1 - e^{-\alpha_2 t}) \right. \\ \left. - \frac{1}{\alpha_3^2 (\alpha_1 - \alpha_3) (\alpha_2 - \alpha_3)} (1 - e^{-\alpha_3 t}) + \frac{1}{\alpha_1 \alpha_2 \alpha_3} t \right]$$

σ_0, σ_1 and σ_2 = Thermal neutron absorption cross section of Th^{232} , Th^{233} and Pa^{233} , respectively.

σ_3 = Non-fission thermal neutron cross section of U^{233} .

σ_f = Thermal neutron fission cross section of U^{233} .

$\lambda_0, \lambda_1, \lambda_2$, and λ_3 = Radioactive decay constant of Th^{232} , Th^{233} , Pa^{233} and U^{233} , respectively.

ϕ = Thermal neutron flux.

N_0 = Number of Th^{232} atoms per unit volume of thorium.

t = Time of irradiation.

$\alpha_i = \lambda_i + \phi \sigma_i$

FIG. 4

EQUATION FOR CALCULATING BURNUP OF NATURAL THORIUM
RESULTING FROM FISSION OF URANIUM-233

Table 1

THERMAL NEUTRON CROSS SECTIONS (IN BARNS) AND NUCLEAR
HALF-LIVES OF Th^{232} , Th^{233} , Pa^{233} , U^{233} AND U^{234}

Isotope	Half-Life	σ_{abs}	σ_{act}	σ_{f}	σ_{abs}^*	σ_{act}^*	σ_{f}^*
Th^{232}	$1.39 \times 10^{10} \text{ y}$	7.0 ± 0.4			6.2		
Th^{233}	23.3 m		1400 ± 200			1240	
Pa^{233}	27.4 d		150			133	
U^{233}	$1.62 \times 10^5 \text{ y}$	564 ± 10		514	500		456
U^{234}	$2.50 \times 10^5 \text{ y}$	89 ± 7	72 ± 10		79	64	

* Average cross section for thermal neutrons having a Maxwellian distribution

FIG. 5

FIG. 6

FIG. 8

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FIG. 9

FIG. 10

PRODUCTION OF URANIUM-234 BY NEUTRON
IRRADIATION OF NATURAL THORIUM

FIG. II

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Table 2

SECULAR EQUILIBRIUM CONCENTRATIONS OF Th^{233} , Pa^{233} ,
 U^{233} , AND U^{234} WITH NATURAL THORIUM

Thermal Neutron Flux, ϕ	$\text{Th}^{233}\% \times 10^{+3}$	$\text{Pa}^{233}\%$	$\text{U}^{233}\%$	$\text{U}^{234}\%$
1.0×10^{13}	.0125	.0211	1.23	.642
5.0×10^{13}	.0625	.104	1.21	.754
1.0×10^{14}	.125	.202	1.19	.888
2.0×10^{14}	.250	.388	1.14	1.14
5.0×10^{14}	.624	.862	1.01	1.79
1.0×10^{15}	1.25	1.45	0.850	2.59

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FIG. 12

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FIG. 13
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TABLE 3

MAXIMUM SPECIFIC POWER OF THORIUM RESULTING
FROM FISSION OF URANIUM-233*

Thermal Neutron Flux neut. / cm ² -sec	Specific Power watts / gram
1.0 x 10 ¹³	4.3
5.0 x 10 ¹³	21
1.0 x 10 ¹⁴	41
2.0 x 10 ¹⁴	79
5.0 x 10 ¹⁴	175
1.0 x 10 ¹⁵	295

*Heat absorbed in thorium per U²³³ fission including
fission product decay = 183 Mev

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