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Th-228 CONTAMINATION IN IRRADIATED THORIA

By

W. K. Woods
Research and Engineering
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March 8, 1965

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RICHLAND, WASHINGTON

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Th-228 CONTAMINATION IN IRRADIATED THORIA

Reactor irradiation of naturally-occurring thorium-232 produces both uranium-233 and the undesirable isotopic contaminant, uranium-232. Uranium-232 decays (74 year half-life) to thorium-228, which in turn decays with a two year half-life to a string of comparatively short-lived daughters some of which emit very highly energetic beta and gamma radiations.

It is generally desirable to accumulate a substantial inventory of irradiated thoria before diverting a plant such as Purex temporarily to chemical processing of the thoria. Especially during this waiting period significant amounts of thorium-228 can be formed from decay of uranium-232, thus causing isotopic contamination of the recovered thorium-232. The purpose of this report is to show how thorium-228 builds up in thoria which has been irradiated under several different conditions and then allowed to cool for a varying period of time. The results are shown in Figures 1 through 4. Derivation of the results is described in Appendix I.

It should be noted that thorium-228 is also formed naturally as a result of the very slow decay of thorium-232 to radium-228 and subsequent decay of radium-228 with a seven year half-life. At secular equilibrium the natural abundance of thorium-228 is 100 millicuries per ton of thoria. Chemical refining of thorium ores generally lowers the radium-228 concentration in the thoria, thus curtailing this source of supply of thorium-228 for a decade or so, so that ordinary thoria will usually contain less than 100 mc of thorium-228 per ton. The thorium-228 concentration prevailing at secular equilibrium, 100 mc per ton of thoria, is shown by dotted line on Figures 1 to 4 for reference purposes. If a ton of recovered thoria contains 100 mc of thorium-228 caused by uranium-232 decay, the total amount of thorium-228 will be at least twice as great as that in the thoria prior to irradiation.

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FIGURE 3

IRRADIATION FRINGE OF SMALL REACTOR

Th-228 FROM U-232 DECAY

rpm = parts U-232 / 10 parts U-233

g/r = gms U-233 / can thorium

secular equilibrium

IRRADIATION TIME PLUS COOLING TIME - days

2 rpm = 0.00 g/r

0.007

FIGURE 4

IRRADIATION FRINGE OF K REACTOR

Th-228 FROM U-232 DECAY

(for ppm and g/r, refer above)

secular equilibrium

IRRADIATION TIME PLUS COOLING TIME - days

1600 g/r

600 gm

500 gm

300 gm

1000 g/r

Th-228 CONCENTRATION
[milli-curies / Ton of Thorium]

Th-228 CONCENTRATION
[milli-curies / Ton of Thorium]

APPENDIX ITh-228 in Irradiated Thoria Caused by U-232 Decay1. Derivation of Relationships During Irradiation

The amount of U-233 in irradiated thoria is essentially linear with irradiation time for the time periods under discussion. Hence, we write that the amount of U-233 = At , where t is irradiation time in calendar days, and A is a constant which is proportional to neutron flux and inversely proportional to time operating efficiency.

The ratio of U-232 to U-233 increases linearly with irradiation time as affected by fast neutrons from adjacent fuel, but increases more rapidly than linearly as U-233 builds in and begins to contribute fast neutrons by fission. For conditions considered herein the ratio of U-232 to U-233 can be expressed approximately by the relation $Bt + Ct^2$, where B and C are empirical constants.

Hence, the amount of U-232, shown by the symbol U , is given by the relation:

$$U = At(Bt + Ct^2) \quad (1)$$

Let the amount of thorium-228 be expressed by the symbol T . Then:

$$dT/dt = \lambda_U U - \lambda t \quad (2)$$

where λ_U is the decay constant for U-232

and λ is the decay constant for Th-228.

Integrating Equation 2, and setting $T = 0$ when $t = 0$, yields:

$$T = A(\lambda_U/\lambda) [B\phi_1(t) + C\phi_2(t)] \quad (3)$$

$$\text{where } \phi_1(t) = t^2 - 2t/\lambda + (2/\lambda^2)(1 - e^{-\lambda t}) \quad (4)$$

$$\text{and } \phi_2(t) = t^3 - 3t^2/\lambda + 6t/\lambda^2 - (6/\lambda^3)(1 - e^{-\lambda t}) \quad (5)$$

Equations 4 and 5 can be put in more usable form by substituting the relation:

$$e^{-\lambda t} = 1 - \lambda t + (\lambda t)^2/2! - (\lambda t)^3/3! + \dots$$

to obtain:

$$\phi_1(t) = (\lambda t^3/3)(1 - \lambda t/4 + (\lambda t/4)(\lambda t/5) - \dots) \quad (4A)$$

$$\phi_2(t) = (\lambda t^4/4)(1 - \lambda t/5 + (\lambda t/5)(\lambda t/6) - \dots) \quad (5A)$$

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2. Derivation of Relationships After Irradiation

Let t_0 = irradiation time, and t represent the variable time after discharge from the reactor. At time of discharge the U-232 concentration and Th-228 concentration are U_0 and T_0 , respectively.

$$dT/dt = \lambda_u U_0 - \lambda t \quad (6)$$

Integrating Equation 6, with the initial conditions given above, yields:

$$T = (\lambda_u/\lambda)(U_0)(1 - e^{-\lambda t}) + T_0 e^{-\lambda t} \quad (7)$$

The abscissa of Figures 1-4 is the sum of $t_0 + t$. The ordinate of Figures 1-4 is obtained from Equations 3 and/or 7.

3. Assignment of Values to the Constants

$$\lambda = \ln(2)/1.91 \text{ years} \times 365 \text{ days/year} = 0.001/\text{day}$$

$$\lambda_u/\lambda = \text{ratio of half-lives} = 1.91/73.6 = 0.026$$

In evaluating constants A , B , and C it is first necessary to specify an assumed operating efficiency in order to arrive at average values per calendar day, rather than per reactor operating day. For purposes of this report it was assumed that the reactor was in operation at a specified power level for 83% of the time.

Information relative to the constants A , B , and C was obtained from R. H. Meichle for both core and fringe locations in both KE, KW and B, D Reactors. (Conditions in C Reactor are somewhat different than for B and D Reactors, but this difference was neglected.) The value of the constant, A , is dependent in part on the thorium density; a density of about 7.5 g/cc was assumed. The constants B and C are essentially independent of density.

In the expression for $U-233 = At$, where $U-233$ is expressed in grams per ton and time is expressed as calendar days, the following values of the constant A were selected, for the indicated tube power (kilowatts) specified as prevailing in the supporting enriched uranium fuel columns.

	<u>A, g/ton/day - (tube power, kw)</u>	
	<u>Small Reactors</u>	<u>K Reactors</u>
Core	17.1 (1250)	22.5 (1900)
Fringe	5.6 (650)	5.6 (845)

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The empirical values of B and C in the expression $(U-232/U-233) \times 10^6 = Bt + C(t)^2$ are shown in Figures 5 and 6, along with a comparison with the original information supplied by Mr. Meichle.

Substitution of the indicated values for the constants shown algebraically above would yield values of thorium-228 in terms of grams per ton of thoria. This is converted into millicuries per ton by noting that a gram of Th-228 is equivalent to 840 curies.

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FIGURE 5

U-232 FORMATION - REACTOR CORES

EMPIRICAL FIT TO CALCULATED DATA

Assumptions: 83% operating efficiency
 1900 KW tube power at K Reactor
 1250 KW " " "Small"

O, X = data points computed by R.H. Meikle

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FIGURE 6

U-232 FORMATION -- REACTOR FRINGES

EMPIRICAL FIT TO CALCULATED DATA

Assumptions: 83% operating efficiency

845 kW tube power at K Reactor

650 kW " " Small " "

X, O = data points compared by R & Heuble

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