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August 17, 1954

H. F. Soule
Radiation Monitoring Sub-Section
Separations Section

ACTIVITY OF RU 103 AND RU 106 IN PILE FISSION PRODUCTS

Introduction

The purpose of this document is to provide a means for estimating the age of ruthenium ground contamination when the ratio of Ru 103 activity to Ru 106 activity in the contamination is known.

Summary

For a pile operating at a time-average power level of P megawatts per ton of uranium and discharging uranium at an exposure of M megawatt days per ton the ratio of Ru 103 activity to Ru 106 activity in the fission products at the time of discharge can be calculated by the following equation:

$$R = \frac{0.35\ M \times 10^{-3} + (3.00 - .020\ P)\ (1 - e^{-0.0174\ M/P})}{0.77\ M \times 10^{-3} + (0.466 - 0.405\ P)\ (1 - e^{-0.0019\ M/P})}$$

For 600 MWD/t uranium the value of R can be calculated to within ±10% of the value given by the above equation by the simple approximation:

$$R = 590\ T^{-0.712} \qquad (100 < T < 600 \text{ days})$$

where: T is the average uranium residence time in the pile in days, or $T = M/P$.

For 900 MWD/t uranium,

$$R = 510\ T^{-0.712} \qquad (100 < T < 600 \text{ days})$$

For 200 MWD/t uranium,

$$R = 92\ T^{-0.246} \qquad (20 < T < 80 \text{ days})$$

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After the uranium is discharged from the pile, the activity ratio decays exponentially with a half life of 45 days. Thus, for 600 MWD/t uranium irradiated 200 days, the activity ratio at time of discharge is calculated to be 13.6 whereas after 90 days of cooling it is calculated to be 3.4.

Basis

The constants used in deriving the equations presented here are as follows:

Fission yield Ru 103 from U-235 -- 2.85 ± 0.16 per cent (1)

Fission yield Ru 106 from U-235 -- 0.38 ± 0.03 per cent (1)

$$\frac{\text{Fission yield Ru 103 from Pu-239}}{\text{Fission yield Ru 103 from U-235}} = 1.49 \quad (2)$$

$$\frac{\text{Fission yield Ru 106 from Pu-239}}{\text{Fission yield Ru 106 from U-235}} = 9.04 \quad (2)$$

$$\frac{\text{Fission yield Ru 103 from U-238}}{\text{Fission yield Ru 103 from U-235}} = 2.00 \quad (2)$$

$$\frac{\text{Fission yield Ru 106 from U-238}}{\text{Fission yield Ru 106 from U-235}} = 5.19 \quad (2)$$

Half life of Ru 103 = 39.8 days (3)

Half life of Ru 106 = 1.0 years (3)

Derivation of Equations

The differential equation describing the net rate of formation of a radioactive fission product A in uranium under irradiation is as follows:*

$$\frac{dA}{dt} = R_1Y_1 + R_2Y_2 + R_3Y_3 - \lambda A \quad (1)$$

where: A represents the concentration of isotope A
λ represents the radioactive decay constant of isotope A
R_1 represents the U-235 fission rate per unit volume
R_2 represents the U-238 fission rate per unit volume
R_3 represents the Pu-239 fission rate per unit volume
Y_1, Y_2, Y_3 are the fission yields from U-235, U-238 and Pu-239 respectively.

* Note: This equation applies to the case where the isotope is formed either directly from fission or from a very short half life "mother" fission fragment.

1. Hardwicke, W.H., Phys. Rev. 92, 1072-73 (Nov. 1953)
2. Steinberg, E.P. and Freedman, M.S., NNES-PPR 9, 1381 (1951)
3. Hollander, J.M. Perlman, I., and Seaborg, G.T., "Table of Isotopes," UCRL 1928 Revised (1952).

[redacted]

In order to express the concentration of isotope A in terms of curies per unit volume, both sides of equation (1) are multiplied by λ/d where d signifies the disintegration rate per curie, yielding:

$$\frac{dC}{dt} = \frac{\lambda}{d}\left(R_1Y_1 + R_2Y_2 + R_3Y_3\right) - \lambda C \qquad (2)$$

C = curies of isotope A per unit volume

If the heat generation rate of each of the three fissionable isotopes under consideration is known, then the R_1, R_2, and R_3 factors can be easily calculated by dividing the respective heat generation rates by the heat of fission. The per cent of the total heat generation due to U-238 fissions, essentially independent of exposure, is approximately 5.39 per cent; the distribution of the remaining heat generation between U-235 fissions and Pu-239 fissions has been shown[4] to depend on the metal exposure as follows:

$$P_1/P = 0.9461 - 2.52\ M \times 10^{-4}$$

$$P_2/P = 0.0539$$

$$P_3/P = 2.52\ M \times 10^{-4}$$

where: P_1, P_2, P_3 are the megawatts per ton generated by U-235, U-238 and Pu-239 respectively
P is the total power generated
M is the metal exposure (MWD/ton)

By noting that M, the exposure per ton, equals Pt, the product of the power per ton and the in-pile time, and by substituting P_1/H_f, P_2/H_f for R_1, R_2, and R_3 respectively in equation (2) one obtains:*

$$\frac{dC}{dt} = \frac{\lambda}{d}\frac{Y_1}{H_f}\left(0.9461\ P - 2.52 \times 10^{-4}\ P^2t\right) \qquad (3)$$

$$+ \frac{\lambda}{d}\frac{Y_2}{H_f}\left(0.0539\ P\right) + \frac{\lambda}{d}\frac{Y_3}{H_f}\left(2.52 \times 10^{-4}\ P^2t\right) - \lambda C$$

where: H_f is the heat of fission (in MWD/fission)

The solution of the above equation can be expressed in the following form:

$$C = P\left[\lambda DT + (B-D)(1 - e^{-\lambda T})\right] \qquad (4)$$

where: $B = \dfrac{0.9461\ Y_1 + 0.0539\ Y_2}{d\ H_f}$

$D = \dfrac{2.52\ P\ (Y_3 - Y_1) \times 10^{-4}}{\lambda\ d\ H_f}$

T = Residence time of uranium in pile

*Note: The dimensions employed here are time in days, power in megawatts per ton and activity in curies per ton since the factor 2.52×10^{-4} has the dimension $(MWD/t)^{-1}$.

4. Gumprecht, R.O., HW-32753 "Production of Kr 85 in the Present Hanford Reactors," 8-13-54.

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In the present case where it is desired to compare the activities of Ru 103 and Ru 106 by means of a ratio, the power term before the bracket and the dH_f factors defining B and D cancel out. When the values of the fission yields and decay constant of Ru 103 and Ru 106 previously given are substituted in equation (4) and the ratio of their activities is taken, the result is as follows:

$$R = \frac{C\ (\text{Ru } 103)}{C\ (\text{Ru } 106)} = \frac{0.35\ M \times 10^{-3} + (3.00 - .020\ P)(1 - e^{-.0174T})}{0.77\ M \times 10^{-3} + (0.466 - 0.405\ P)(1 - e^{-.0019T})} \qquad (5)$$

Table I illustrates the values of R computed from equation (5) for several exposures and pile residence times.

TABLE I

M (MWD/t)	T (days)	R
200	20	44
200	40	38
200	60	33.6
200	80	29.8
600	100	21.2
600	200	13.6
600	300	10.1
600	400	8.2
600	500	7.1
600	600	6.3
900	100	18.5
900	200	11.9
900	500	6.2
900	800	4.7

When the points of Table I are plotted, it is observed that values of R can be calculat to within about ±10 per cent of the value given by equation (5) by the simple approxi-mations:

for 200 MWD/t uranium $R = 92\ T^{-0.246}$ $(20 < T < 80)$

for 600 MWD/t uranium $R = 590\ T^{-0.712}$ $(100 < T < 600)$

for 900 MWD/t uranium $R = 510\ T^{-0.712}$ $(100 < T < 600)$

After the uranium is discharged from the pile, the Ru 103 activity will decay exponentially by the factor $e^{-0.0174\ t}$ and the Ru 106 activity will decay by the factor $e^{-0.0019\ t}$. Consequently, the ratio of their activities will decay by the factor $e^{-0.0155\ t}$ where t is the cooling time in days.

Discussion

It must be recognized that equations (4) and (5) are approximate in that they rely on the assumption that the uranium is irradiated at a constant rate which, of course, is not true in the Hanford piles which must shut down periodically to discharge metal.

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For the relatively long half-lives involved here, however, the assumption of a constant average irradiation rate causes an error in the calculated value of R of less than 2 per cent. A far more likely source of error occurs in determining the average residence time of the uranium in the pile since each discharge is composed of uranium from various tubes operating at various powers for various periods of time. Going back to the general formula, equation (4), the total curies of isotope A in the push should be expressed as follows:

$$\text{Total curies of isotope A} = \sum_{i} X_i P_i \left[\lambda D_i T_i + (B-D_i)(1 - e^{-\lambda T_i}) \right]$$

where X_i = tons of uranium exposed at P_i megawatts per ton for T_i days

A tube-by tube summation of ruthenium activities for the B Pile discharge of August 11, 1953 gave a value of R about 6 1/2 per cent greater than that obtained from equation (5 using the average metal exposure of the push and the average residence time of the uranium in the pile. However, even if all uranium were exposed at a fixed power for a fixed time, the value of R given by equation (5) would have an uncertainty of at leas ±10 per cent due to the uncertainties associated with the values given for the fission yields. An error of 10 per cent in R results in an error of about 7 days in estimating the age of the observed ground contamination.

R.O.Gumprecht

Pile Physics Sub-Unit
Technical Section
ENGINEERING DEPARTMENT

R.O. Gumprecht:mbs

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