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LONG-TERM NEUTRON ACTIVATION PRODUCTS

OF NICKEL-58

W. C. Morgan

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LONG-TERM NEUTRON ACTIVATION PRODUCTS OF NICKEL-58

Introduction

Certain advantages, such as higher strength at elevated temperature, have led to the use of alloys containing large percentages of nickel as a replacement for aluminum components. One example is the inconel sheathing of control rods for the Hanford reactors. Since some of these components may remain in a reactor several years prior to removal, the neutron-activation products resulting from multiple chain reactions may become important¹ in determining the radiation exposure levels at discharge. This paper outlines one of the activation chains which may contribute significantly to the γ -activity. Equations are developed from which the concentrations of the product isotopes emanating from nickel-58 can be calculated. "Best values" of the parameters used in these calculations are tabulated, and the conversion from isotope concentration to γ -dose rate is outlined.

Derivation of Equations

The long-term activation chain of nickel-58 is outlined as follows:

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Where: σ , σ_m , σ_1 , etc. are activation cross sections, and λ_1 , λ_a , etc. are decay constants.

The concentrations of the activation products, emanating from the original Ni^{58} , can be described by the following set of differential equations:

$$\frac{d [\text{Co}^{58m}]}{dt} = [\text{Ni}^{58}] \sigma_m \phi_f - [\text{Co}^{58m}] (\lambda_1 + \sigma_1 \phi) \quad (1)$$

$$\frac{d [\text{Co}^{58}]}{dt} = [\text{Ni}^{58}] \sigma \phi_f + [\text{Co}^{58m}] \lambda_1 - [\text{Co}^{58}] (\lambda_2 + \sigma_2 \phi) \quad (2)$$

$$\frac{d [\text{Fe}^{58}]}{dt} = [\text{Co}^{58}] \lambda_2 - [\text{Fe}^{58}] \sigma_3 \phi \quad (3)$$

$$\frac{d [\text{Fe}^{59}]}{dt} = [\text{Fe}^{58}] \sigma_3 \phi - [\text{Fe}^{59}] \lambda_3 \quad (4)$$

$$\frac{d [\text{Co}^{59}]}{dt} = \left([\text{Co}^{58m}] \sigma_1 + [\text{Co}^{58}] \sigma_2 \right) \phi + [\text{Fe}^{59}] \lambda_3 - [\text{Co}^{59}] \sigma_a \phi \quad (5)$$

$$\frac{d [\text{Co}^{60}]}{dt} = [\text{Co}^{59}] \sigma_a \phi - [\text{Co}^{60}] (\lambda_a + \sigma_c \phi) \quad (6)$$

Where, ϕ_f = Fast neutron flux

ϕ = Thermal neutron flux, and

$[\text{Ni}^{58}]$ indicates the concentration of the enclosed isotope, Ni^{58} , etc.

$$[\text{Ni}^{58}] = \frac{\text{CP N}_o}{10^4 \text{ A}}$$

C = per cent concentration of nickel in the alloy.

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P = per cent abundance of Ni^{58} in nickel

N_0 = Avogadro's Number, number of atoms per gram mole

A = Atomic Weight of nickel

Thermal activation, not shown in the preceding activation scheme, removes only about 0.8 per cent of the Ni^{58} per year at the highest flux positions in K reactor; therefore, it may be ignored for most calculations, or a simple reduction in $[\text{Ni}^{58}]$ can be made to compensate for the average reduction over the exposure range of interest.

The solution to equation 1 can be written:

$$[\text{Co}^{58m}] = \frac{[\text{Ni}^{58}] \sigma_m \phi_f [1 - e^{-(\lambda_1 + \sigma_1 \phi)t}]}{(\lambda_1 + \sigma_1 \phi)} \quad (7a)$$

For most positions in the Hanford reactors, equation (7a) reduces, for t more than 2 days, to:

$$[\text{Co}^{58m}] = \frac{[\text{Ni}^{58}] \sigma_m \phi_f}{(\lambda_1 + \sigma_1 \phi)} \quad (7b)$$

which can be written more compactly,

$$[\text{Co}^{58m}] = \alpha \sigma_m \quad (7c)$$

$$\text{Where, } \alpha = \frac{[\text{Ni}^{58}] \phi_f}{(\lambda_1 + \sigma_1 \phi)}$$

The solution to equation 2 is given by:

$$[\text{Co}^{58}] = [\text{Ni}^{58}] \phi_f \left\{ \left(\frac{1}{\lambda_2 + \sigma_2 \phi} \right) \left(\frac{\lambda_1 \sigma_m}{\lambda_1 + \sigma_1 \phi} + \sigma \right) \left[1 - e^{-(\lambda_2 + \sigma_2 \phi)t} \right] + \left(\frac{\lambda_1 \sigma_m}{\lambda_1 + \sigma_1 \phi} \right) \left[\frac{1}{\lambda_1 - \lambda_2 + (\sigma_1 - \sigma_2) \phi} \right] \left[e^{-(\lambda_1 + \sigma_1 \phi)t} - e^{-(\lambda_2 + \sigma_2 \phi)t} \right] \right\} \quad (8a)$$

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For most positions in the Hanford reactors, equation (8a) reduces, for t more than 15 days, to:

$$[Co^{58}] = \frac{[Ni^{58}]\phi_f}{(\lambda_2 + \sigma_2\phi)} \left(\frac{\lambda_1\sigma_m}{\lambda_1 + \sigma_1\phi} + \sigma \right) \left[1 - e^{-(\lambda_2 + \sigma_2\phi)t} \right] \quad (8b)$$

or, written more compactly,

$$[Co^{58}] = \beta [1 - e^{-(\lambda_2 + \sigma_2\phi)t}] \quad (8c)$$

$$\text{Where, } \beta = \frac{[Ni^{58}]\phi_f}{(\lambda_2 + \sigma_2\phi)} \left(\frac{\lambda_1\sigma_m}{\lambda_1 + \sigma_1\phi} + \sigma \right) = \alpha \left[\frac{\lambda_1\sigma_m + \sigma(\lambda_1 + \sigma_1\phi)}{\lambda_2 + \sigma_2\phi} \right]$$

Substituting (8) into equation 5, the solution reduces to:

$$[Fe^{58}] = \beta \frac{\lambda_2}{\sigma_3\phi} \left(1 - e^{-\sigma_3\phi t} \right) \quad (9)$$

Likewise the solution to equation 6 reduces to:

$$[Fe^{59}] = \beta \frac{\lambda_2}{\lambda_3} \left(1 - e^{-\sigma_3\phi t} \right) \quad (10)$$

The solution to equation 7 can be written:

$$[Co^{59}] = \left[\frac{\alpha\sigma_m\sigma_1}{\sigma_a} + \frac{\beta\sigma_2}{\sigma_a} + \frac{\beta\lambda_2}{\sigma_a\phi} \right] (1 - e^{-\sigma_a\phi t}) - \left[\frac{\beta\sigma_2\phi}{\lambda_2 + (\sigma_2 - \sigma_a)\phi} \right] \cdot \left[e^{-(\lambda_2 + \sigma_2\phi)t} - e^{-\sigma_a\phi t} \right] - \left[\frac{\beta\lambda_2}{(\sigma_a - \sigma_3)\phi} \left(e^{-\sigma_3\phi t} - e^{-\sigma_a\phi t} \right) \right] \quad (11a)$$

or, more compactly,

$$[Co^{59}] = \left(\frac{\gamma}{\sigma_a\phi} \right) \left(1 - e^{-\sigma_a\phi t} \right) - \delta \left[e^{-(\lambda_2 + \sigma_2\phi)t} - e^{-\sigma_a\phi t} \right] - \frac{\epsilon}{\phi} \left(e^{-\sigma_3\phi t} - e^{-\sigma_a\phi t} \right) \quad (11b)$$

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Where, $\gamma = [\alpha\sigma_m\sigma_1\phi + \beta(\lambda_2 + \sigma_2\phi)]$

$$\delta = \frac{\beta\sigma_2\phi}{\lambda_2 + (\sigma_2 - \sigma_a)\phi}$$

$$\epsilon = \frac{\beta\lambda_2}{(\sigma_a - \sigma_3)}$$

The solution to equation 6 can be written:

$$[Co^{60}] = (\gamma + \delta\sigma_a + \epsilon\sigma_a) \left[\frac{e^{-\sigma_a\phi t} - e^{-(\lambda_a + \sigma_c\phi)t}}{\lambda_a + (\sigma_c - \sigma_a)\phi} \right] + \gamma \left[\frac{1 - e^{-(\lambda_a + \sigma_c\phi)t}}{\lambda_a + \sigma_c\phi} \right] \\ + \delta\sigma_a\phi \left[\frac{e^{-(\lambda_a + \sigma_c\phi)t} - e^{-(\lambda_2 + \sigma_2\phi)t}}{\lambda_a - \lambda_2 + (\sigma_c - \sigma_2)\phi} \right] - \epsilon\sigma_a \left[\frac{e^{-\sigma_3\phi t} - e^{-(\lambda_a + \sigma_c\phi)t}}{\lambda_a + (\sigma_c - \sigma_3)\phi} \right] \quad (12)$$

Notes on Numerical Evaluation

It is suggested that the exposure (ϕt) and neutron fluxes (ϕ and ϕ_f) be entered in the forms $\left[(nvt/Mwd/At)_{\text{average}} \times (Mwd/At)_{\text{measured}} \right]$ and $\left[(nvt/Mwd/At)_{\text{average}} \times (Mw/At)_{\text{measured}} \right]$. Using this method, the calculations are not made obsolete by a change in power level, which changes the flux level but not the ratio of exposure per unit power. Mw/At can be routinely monitored; thus the activity can readily be calculated at any time.

Neutron exposures are dependent on position in the reactors. The fast-neutron cross sections ($\bar{\sigma}$) are given in terms of the neutron spectrum², and the neutron-flux energy range established by ϕ_f :

$$\bar{\sigma}(E_1) = \frac{\int_0^\infty \phi(E) \sigma(E) dE}{\int_{E_1}^\infty \phi(E) dE}$$

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Where, $\bar{\sigma}(E_1)$ = the neutron activation cross section averaged
over the reactor spectrum above energy E_1 .

$\phi(E)$ = the neutron energy spectrum for the reactor.

$\sigma(E)$ = the neutron-activation cross section as a function of neutron energy.

The neutron exposures per Mwd/At and fast-neutron activation cross sections are listed in Table I. The numbers given are valid only for irradiations made in the equivalent of hot-test-hole positions of the two reactors. "Best values" of the other constants are also tabulated.

TABLE I. "BEST VALUES" OF THE CONSTANTS

<u>Symbol</u>	<u>Quantity</u>	<u>Value</u>	<u>Ref.</u>
$\frac{\phi}{\text{Mwd/At}}$	Thermal exposure in:		(3,4)
	K Reactor	4.5×10^{17} nvt/Mwd/At	
	C Reactor	4.6×10^{17} nvt/Mwd/At	
	other old reactors	4.7×10^{17} nvt/Mwd/At	
$\frac{\phi_f}{\text{Mwd/At}}$	Fast exposure ($E > 1.0$ Mev) in:		(2)
	K Reactor	4.6×10^{16} nvt/Mwd/At	
	C Reactor	3.5×10^{16} nvt/Mwd/At	
σ	$\text{Ni}^{58} (n,p) \text{Co}^{58}$ activation		(2,4)
	cross section for ϕ_f ($E > 1.0$ Mev)		
	in:		
	K Reactor	7.4×10^{-26} cm ²	
σ_m	C Reactor	7.3×10^{-26} cm ²	
	$\text{Ni}^{58} (n,p) \text{Co}^{58m}$ activation		(2,4)
	cross section for ϕ_f ($E > 1.0$ Mev)		
	in:		
	K Reactor	3.2×10^{-26} cm ²	
	C Reactor	3.2×10^{-26} cm ²	

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- Table I continued -

<u>Symbol</u>	<u>Quantity</u>	<u>Value</u>	<u>Ref.</u>
σ_1	Co ^{58m} thermal neutron cross section	$1.70 \times 10^{-19} \text{ cm}^2$	(5)
σ_2	Co ⁵⁸ " " " "	$1.65 \times 10^{-21} \text{ cm}^2$	(6)
σ_3	Fe ⁵⁸ " " " "	$1.0 \times 10^{-24} \text{ cm}^2$	(7)
σ_a	Co ⁵⁹ " " " "	$3.8 \times 10^{-23} \text{ cm}^2$	(7)
σ_c	Co ⁶⁰ " " " "	$6.0 \times 10^{-24} \text{ cm}^2$	(8)
λ_1	Co ^{58m} decay constant ($t_{1/2} = 9.1 \text{ hrs}$)	1.83 day^{-1}	(5)
λ_2	Co ⁵⁸ " " ($t_{1/2} = 71.3 \pm 0.2 \text{ day}$)	$9.72 \times 10^{-3} \text{ day}^{-1}$	(9)
λ_3	Fe ⁵⁹ " " ($t_{1/2} = 44.3 \text{ days}$)	$1.56 \times 10^{-2} \text{ day}^{-1}$	(7)
λ_a	Co ⁶⁰ " " ($t_{1/2} = 5.28 \text{ years}$)	$3.60 \times 10^{-4} \text{ day}^{-1}$	(7)

To compute the disintegration rate per unit of material from the concentrations of the isotopes calculated from the equations derived in the preceding section, multiply the concentration of each isotope by its decay constant and weight (in grams) of alloy per unit of material (i.e.: per foot, 100 gm., etc. of component). The γ -yield per disintegration¹⁰ for the product isotopes is listed in Table II.

TABLE II. γ -YIELDS

<u>Isotope</u>	<u>Energy per γ, mev</u>	<u>Average number of γ's per disintegration</u>
Co ⁶⁰	1.17	1.00
	1.33	1.00
Co ⁵⁸	0.81	1.00
Fe ⁵⁹	1.29	0.46
	1.10	0.54

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