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SUBJECT: DETERMINATION OF RESIDUAL ACTIVITIES
FROM LONG-LIVED FISSION PRODUCTS OF ²³⁵U

TO: F. L. Culler

FROM: E. J. Frederick

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1.0 ABSTRACT

A table of nuclear constants and saturation values for a thermal-neutron flux of 2×10^{13} n/cm²/sec or a power density of approximately 1 kw/g of initial U²³⁵ charged to the reactor is presented for all of the U²³⁵ fission products with half-lives greater than 8 days but less than 100 years and fission yields greater than 0.01%. Approach-to-saturation and decay of activity vs. time are plotted in a series of graphs for all the fission products included in the table. Several examples of practical application of these data are included.

2.0 INTRODUCTION

The continued need for determining residual activities of various fission products involved in the chemical processing of U²³⁵ heterogeneous reactor fuels has led to the tabulation of data from a rather complete series of calculations, thereby providing a quick and easy means of making future determinations. In this type of problem the chief variables are neutron flux, irradiation time, and decay time. Accordingly, the data given in this report present accurate determination of residual activities for a neutron flux of 2×10^{13} which can be linearly extrapolated to other fluxes from 10^{12} to 10^{14} n/cm²/sec, irradiation times of 0 to 14 months, and decay times from 2 weeks to 14 months. These data cannot be used for accurately determining total fission product activity for decay times of less than two weeks since all fission products with half-lives less than 8 days have been neglected. In the calculations the effect of neutron capture by individual fission products was neglected. At low neutron fluxes the error introduced by this assumption is of only minor importance for the fission products considered here.

Much more detailed calculations for all fission products, including the effects of neutron capture by fission products, are currently being

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made on the ORNL Oracle digital computer. Results of these calculations will be the subject of a subsequent report. The nuclear constants used in both the present and the forthcoming study are the same and are selected "best" values; therefore, in problems where the decay period is long, the data presented in this report will give an approximation to within a few percent of the values in the later report.

3.0 DISCUSSION

In Table 1 are summarized the fission product yields, half-lives, average β energies, and total γ energies of all fission products with yields greater than 0.01% and with half-lives greater than 8 days as well as saturation values in curies and watts for a reactor operating at an average thermal flux of 2×10^{13} n/cm²/sec. The percentage figures following the γ energies indicate the fraction of the total disintegration going to that particular emission. It is assumed that the average β energies are 33% of the maximum. Saturation values for reactor conditions other than the noted flux are obtained by multiplying the individual saturation values by the ratio of the fluxes. For example, the saturation value for 73-year Sm¹⁵¹ at a flux of 2×10^{13} n/cm²/sec is 4.03 β curies. The saturation value then for this same fission product at a flux of 2×10^{14} n/cm²/sec is $4.03 \times 2 \times 10^{14} / 2 \times 10^{13} = 40.3$ β curies. The same ratio factor applies for determining β , γ , and total watts.

The approach of the various fission product activities to their saturation values is shown graphically in Figs. 1-4 as a function of irradiation time. For instance, in Fig. 2, after 5.3 months of irradiation, 1.0-year Ru¹⁰⁶ is at 27.7% of its saturation value and has associated with it an equal amount of 30-sec Rh¹⁰⁶ activity, which is in equilibrium with the Ru¹⁰⁶ activity from which it is formed.

The various fission product activities are given as a function of decay time after reactor discharge in Figs. 5-8. It may be noted in Fig. 6 that 5.0 months after reactor discharge the activity of 11.3-day Nd¹⁴⁷ is reduced by a factor of 9.2×10^{-5} . Decay curves for Nb⁹⁵ after various irradiation times are shown in Fig. 8. These curves are typical of a decay chain where both the parent (Zr⁹⁵) isotope and the daughter (Nb⁹⁵) have fairly long half-lives of the same order of magnitude.

Table 1. Nuclear Constants and Saturation Values for Fission Products with Half-lives between 8 days and 100 years and a Fission Yield of $\geq 0.01\%$

Species	Yield, %	Avg. β Energy (0.33 Mev.), Mev	γ Energy, Mev	No. of γ 's per 100 β 's	Saturation Values/g Initial U^{235} for $\phi = 2 \times 10^{13}$ n/cm ² /sec			
					β Curies	β Watts	γ Watts	Total Watts
¹⁵¹ Ba	0.445	0.025	0.019	100	4.03	5.96×10^{-4}	4.53×10^{-4}	1.049×10^{-3}
¹³⁷ Cs	5.9	0.19	0	---	47.50	5.34×10^{-2}	---	5.34×10^{-2}
(2.60m) Ba ¹³⁹	5.4	0	0.661	100	43.47	---	1.70×10^{-1}	1.70×10^{-1}
⁹⁰ Sr	5.9	0.20	0	---	47.50	5.63×10^{-2}	---	5.63×10^{-2}
(64.5h) Y ⁹⁰	5.9	0.73	0	---	47.50	2.05×10^{-1}	---	2.05×10^{-1}
⁸⁵ Kr	0.3	0.232	0	---	2.44	3.35×10^{-3}	---	3.35×10^{-3}
¹²⁵ Sb	0.023	0.102	0.428	37	0.19	1.15×10^{-1}	1.78×10^{-4}	5.65×10^{-4}
			0.598	28			1.88×10^{-4}	
			0.638	7			0.50×10^{-4}	
			0.175	9			0.18×10^{-4}	
			0.035	9			0.04×10^{-4}	
			0.120	10			0.12×10^{-4}	
(59d) Te ¹²⁵	0.003	0	0.110	100	0.03	---	1.95×10^{-5}	1.95×10^{-5}
¹⁴⁷ Pu	2.6	0.074	0	---	20.93	9.17×10^{-3}	---	9.17×10^{-3}
¹⁵⁵ Ba	0.031	0.056	0.0187	100	0.25	8.28×10^{-5}	0.28×10^{-4}	8.76×10^{-4}
			0.0593	100			0.88×10^{-4}	
			0.0558	100			1.27×10^{-4}	
			0.1045	100			1.54×10^{-4}	
			0.1309	100			1.54×10^{-4}	
			0.1368	100			2.03×10^{-4}	
¹⁰⁶ Ba	0.38	0.0131	0	---	3.06	2.37×10^{-4}	---	2.37×10^{-4}
(30d) Ba ¹⁰⁶	0.38	1.05	0.513	25	3.06	1.90×10^{-2}	2.32×10^{-2}	4.44×10^{-2}
			0.624	12			0.14×10^{-2}	
			0.87	1			0.02×10^{-2}	
			1.045	2			0.04×10^{-2}	
			1.55	0.5			0.01×10^{-2}	
			2.41	0.25			0.01×10^{-2}	
¹⁴⁴ Ce	6.1	0.090	0.0337	10	49.11	2.61×10^{-2}	0.98×10^{-3}	3.78×10^{-2}
			0.054	10			1.57×10^{-3}	
			0.0807	10			2.35×10^{-3}	
			0.108	10			2.90×10^{-3}	
			0.134	10			3.09×10^{-3}	
(17.5m) Pr ¹⁴⁴	6.1	0.97	0.693	4	49.11	2.82×10^{-1}	8.08×10^{-3}	3.05×10^{-1}
			1.480	2			8.62×10^{-3}	
			2.165	1			6.35×10^{-3}	
¹¹⁹ Sn	<0.01	0	0.0653	100	0.08	---	3.09×10^{-5}	4.24×10^{-5}
			0.0642	100			1.15×10^{-5}	
⁹⁵ Zr	6.4	0.127	0.722	42	51.52	3.87×10^{-2}	0.93×10^{-1}	2.63×10^{-1}
			0.754	57			1.31×10^{-1}	
(35d) Ba ⁹⁵	6.4	0.0533	0.745	100	51.52	1.62×10^{-2}	2.27×10^{-1}	2.43×10^{-1}
⁹¹ Y	5.9	0.512	0	---	47.50	1.44×10^{-1}	---	1.44×10^{-1}
⁹⁹ Sr	4.8	0.487	0	---	38.64	1.11×10^{-1}	---	1.11×10^{-1}
¹⁰³ Ba	2.9	0.080	0.498	95	23.35	1.11×10^{-2}	6.54×10^{-2}	7.65×10^{-2}
(54m) Ba ¹⁰³	2.9	0	0.040	100	23.35	---	5.52×10^{-3}	5.52×10^{-3}
^{129m} Te	0.34	0	0.106	100	2.74	---	1.71×10^{-3}	1.71×10^{-3}
(72m) Te ¹²⁹	1.0	0.6	1.1	100	8.05	2.86×10^{-2}	5.23×10^{-2}	8.09×10^{-2}
¹⁴¹ Cs	6.0	0.163	0.145	67	48.30	4.64×10^{-2}	2.77×10^{-2}	7.41×10^{-2}
¹⁴³ Pr	6.2	0.31	0	---	49.91	9.16×10^{-2}	---	9.16×10^{-2}
¹⁴⁰ Ba	6.3	0.268	0.0296	100	50.72	8.04×10^{-2}	0.89×10^{-2}	1.52×10^{-1}
			0.132	Weak			---	
			0.162	10			0.49×10^{-2}	
			0.304	10			0.91×10^{-2}	
			0.537	30			4.83×10^{-2}	
(40d) La ¹⁴⁰	6.3	0.495	3.0	1	50.72	1.48×10^{-1}	0.90×10^{-2}	7.81×10^{-1}
			2.50	5.4			4.04×10^{-2}	
			1.596	94			45.0×10^{-2}	
			0.8191	20			7.05×10^{-2}	
			0.4867	39			5.70×10^{-2}	
			0.3286	5.4			0.53×10^{-2}	
			0.053	1.8			0.05×10^{-2}	
¹⁴⁷ Mo	2.6	0.220	0.092	60	20.93	2.82×10^{-2}	0.68×10^{-2}	5.71×10^{-2}
			0.318	15			0.59×10^{-2}	
			0.532	25			1.64×10^{-2}	
¹³¹ I	2.9	0.191	0.060	2.2	23.35	2.64×10^{-2}	0.24×10^{-3}	8.01×10^{-2}
			0.161	0.7			0.16×10^{-3}	
			0.284	5.3			2.08×10^{-3}	
			0.364	80			40.3×10^{-3}	
			0.637	9			7.9×10^{-3}	
			0.722	3			2.99×10^{-3}	

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Fig. 1.—Fractional Approach to Saturation vs. Irradiation Time at Constant Neutron Flux for Fission Products with Half-lives between 10 and 73 years.

Fig. 2.—Fractional Approach to Saturation vs. Irradiation Time at Constant Neutron Flux for Fission Products with Half-lives between 100 days and 10 years.

Fig. 3.— Fractional Approach to Saturation vs. Irradiation Time at Constant Neutron Flux for Fission Products with Half-lives between 13 and 100 days.

Fig. 4.—Fractional Approach to Saturation vs. Irradiation Time at Constant Neutron Flux for Fission Products with Half-lives between 8 and 13 days.

Fig. 5.—Fractional Decay after Reactor Discharge vs. Decay Time for Fission Products with Half-lives between 100 days and 73 years.

Fig. 6.—Fractional Decay after Reactor Discharge vs. Decay Time for Fission Products with Half-lives between 8 and 100 days.

Fig. 7.—Fractional Decay after Reactor Discharge vs. Decay Time for Ba^{140} and La^{140} . $T_1 = \infty$

Fig. 8.—Fractional Decay after Reactor Discharge vs. Decay Time for Nb^{95}

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4.0 PRACTICAL APPLICATIONS

Below are given several examples of practical applications of the data.

Example 1

Problem. A reactor fuel element initially containing 50 g of U^{235} is irradiated 20 days at a power level of 5 kw/g U^{235} and allowed to decay for 60 days. Determine the 3.0-, 2.5-, and 1.6-Mev La^{140} γ energies, in watts, for use in determining shielding requirements.

Solution. From Table 1 the saturation values for 3.0-, 2.5-, and 1.6-Mev γ energies from La^{140} at a flux of 2×10^{13} n/cm²/sec equivalent to a power level of 1 kw/g U^{235} are 0.90×10^{-2} , 4.0×10^{-2} , and 4.50×10^{-1} watts/g initial U^{235} , respectively. Therefore, at a power of 5 kw/g U^{235} the saturation values are

$$\begin{aligned} \text{For 3.0 Mev} & \quad 0.90 \times 10^{-2} \times 5/1 = 4.5 \times 10^{-2} \text{ watt/g } U^{235} \\ \text{For 2.5 Mev} & \quad 4.04 \times 10^{-2} \times 5/1 = 2.02 \times 10^{-1} \text{ watt/g } U^{235} \\ \text{For 1.6 Mev} & \quad 4.5 \times 10^{-1} \times 5/1 = 2.25 \text{ watt/g } U^{235} \end{aligned}$$

From Fig. 4 the approach-to-saturation factor for a 20-day irradiation time is 0.623. The decay factor from Fig. 7 for a 60-day decay time is 4.35×10^{-2} . The residual γ activities then are

$$\begin{aligned} \text{For 3.0 Mev} & \quad 4.5 \times 10^{-2} \times 0.623 \times 4.35 \times 10^{-2} = 1.22 \times 10^{-3} \text{ watt/g } U^{235} \\ & \quad = 6.1 \times 10^{-2} \text{ watt/50 g } U^{235} \\ \text{For 2.5 Mev} & \quad 2.02 \times 10^{-1} \times 0.623 \times 4.35 \times 10^{-2} = 5.47 \times 10^{-3} \text{ watt/g } U^{235} \\ & \quad = 2.74 \times 10^{-1} \text{ watt/50 g } U^{235} \\ \text{For 1.6 Mev} & \quad 2.25 \times 0.623 \times 4.35 \times 10^{-2} = 6.10 \times 10^{-2} \text{ watt/g } U^{235} \\ & \quad = 3.05 \text{ watts/50 g } U^{235} \end{aligned}$$

Example 2

Problem. The aqueous waste from the processing of 100 kg of U^{235} from a reactor operated 8 months at a power level of 400 watts/g initial U^{235} is 10,000 gal. After 14 months' decay, it is decided to process the waste stream to recover the Cs^{137} . Determine the total curies of Cs^{137} available for processing.

Solution. The saturation value from Table 1 for Cs^{137} at a power level of 1 kw/g U^{235} is 47.50 β curies. At a power level of 0.4 kw/g U^{235} the value is $47.5 \times 0.4/1 = 19.0 \beta$ curies/g U^{235} .

From Fig. 1 the approach-to-saturation factor for Cs^{137} after 8 months' reactor operation is 0.014. The decay factor for a 14-month decay period (Fig. 5) is $1 - (5.83 \times 10^{-5} \times 30 \times 14) = 1 - 2.45 \times 10^{-2} = 0.975$.

$$\begin{aligned} 19.0 \times 1.4 \times 10^{-2} \times 0.975 &= 0.259 \beta \text{ curies of } \text{Cs}^{137}/\text{g } \text{U}^{235} \\ &= 2.59 \times 10^4 \beta \text{ curies}/100 \text{ kg } \text{U}^{235}. \end{aligned}$$

Example 3

Problem. In a certain radiochemical processing operation an amount of fuel containing 10 kg of U^{235} before burnup is processed per day. The fuel is irradiated 50 days at a flux of $8 \times 10^{13} \text{ n/cm}^2/\text{sec}$ and decayed 15 days prior to processing. Determine the total curies of I^{131} evolved per day if 50% is released during dissolution. If only 10 curies is permitted to escape to the atmosphere per day, what must be the efficiency of an absorber to meet this requirement?

Solution. From Table 1 the saturation value for I^{131} at a reactor operating flux of $2 \times 10^{13} \text{ n/cm}^2/\text{sec}$ is 23.35 β curies. At a flux of $8 \times 10^{13} \text{ n/cm}^2/\text{sec}$ the value is $23.35 \times 8 \times 10^{13}/2 \times 10^{13} = 93.4 \beta$ curies/g U^{235} .

The approach-to-saturation factor for a 50-day irradiation (Fig. 4) is 0.987 and the decay factor for a 15-day decay time (Fig. 6) is 0.25.

$$\begin{aligned} 93.4 \times 0.987 \times 0.25 &= 23.1 \beta \text{ curies/g } \text{U}^{235} \\ &= 2.31 \times 10^5 \beta \text{ curies}/10 \text{ kg } \text{U}^{235} \\ &= 2.31 \times 10^5 \times 0.5 = 1.16 \times 10^5 \beta \text{ curies/day evolved} \\ &\quad \text{during dissolution} \end{aligned}$$

$100 \times 10/1.16 \times 10^5 = 8.63 \times 10^{-3}\%$ per day tolerated to escape; the absorber efficiency therefore must be $100.000 - 0.009 = 99.991\%$.

Example 4

Problem. In a certain radiochemical processing plant the capacity is 20 kg of U^{235} for fuel with a U^{235} depletion of 20%. This fuel has been irradiated 159 days and decayed 150 days prior to chemical processing. The reactor power is 2 kw/g U^{235} . The daily waste volume is 1200 gal. Determine the total heat (both β and γ) evolved in the waste in Btu/hr/gal after 150 days' decay.

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Solution. The amount of U^{235} charged to the reactor is $20/(1 - 0.2) = 25 \text{ kg } U^{235}$. Obtain the saturation value for the total fission products from a reactor operating at a thermal flux of $2 \times 10^{13} \text{ n/cm}^2/\text{sec}$ (Table 1) in total watts/g U^{235} and the appropriate factors for 5.3 months' irradiation (Figs. 1-4) and 5.0 months' decay (Figs. 5-8). The residual watts per gram of U^{235} as given in the following tabulation are the product of these factors.

Species	Saturation Values ($\phi = 2 \times 10^{13} \text{ n/cm}^2/\text{sec}$), total watts/g U^{235}	Irradiation (5.3 months)	Decay (5.0 months)	Residual $\beta + \gamma$ watts/g U^{235}
73y Sm ¹⁵¹	1.049×10^{-3}	4.2×10^{-3}	3.89×10^{-3}	1.72×10^{-8}
33y Cs ¹³⁷	5.34×10^{-2}	9.4×10^{-3}	8.75×10^{-3}	4.39×10^{-6}
(2.6m)Ba ^{137*}	1.70×10^{-1}	9.4×10^{-3}	8.75×10^{-3}	1.40×10^{-5}
28y Sr ⁹⁰	5.63×10^{-2}	1.1×10^{-2}	1.05×10^{-2}	6.50×10^{-6}
(64.5h) Y ⁹⁰	2.05×10^{-1}	1.1×10^{-2}	1.05×10^{-2}	2.37×10^{-5}
10.27y Kr ⁸⁵	3.35×10^{-3}	2.88×10^{-2}	2.73×10^{-2}	2.64×10^{-6}
2.7y Sb ¹²⁵	5.65×10^{-4}	1.01×10^{-1}	9.00×10^{-1}	5.13×10^{-5}
(59d)Te ^{125*}	1.95×10^{-5}	1.01×10^{-1}	9.00×10^{-1}	1.77×10^{-6}
2.6y Pm ¹⁴⁷	9.17×10^{-3}	1.01×10^{-1}	9.00×10^{-1}	8.33×10^{-4}
1.7y Eu ¹⁵⁵	8.76×10^{-4}	1.63×10^{-1}	8.46×10^{-1}	1.21×10^{-4}
1.0y Ru ¹⁰⁶	2.37×10^{-4}	2.64×10^{-1}	7.50×10^{-1}	4.70×10^{-5}
(30s)Rh ¹⁰⁶	4.44×10^{-2}	2.64×10^{-1}	7.50×10^{-1}	8.80×10^{-3}
290d Ce ¹⁴⁴	3.78×10^{-2}	3.15×10^{-1}	7.00×10^{-1}	8.34×10^{-3}
(17.5m)Pr ¹⁴⁴	3.05×10^{-1}	3.15×10^{-1}	7.00×10^{-1}	6.73×10^{-2}
~275d Sn ^{119*}	4.24×10^{-5}	3.29×10^{-1}	6.85×10^{-1}	9.57×10^{-6}
63d Zr ⁹⁵	2.63×10^{-1}	8.26×10^{-1}	1.92×10^{-1}	4.17×10^{-2}
(35d)Nb ⁹⁵	2.43×10^{-1}	6.49×10^{-1}	4.60×10^{-1}	7.26×10^{-2}
58d Y ⁹¹	1.44×10^{-1}	8.49×10^{-1}	1.64×10^{-1}	2.01×10^{-2}
54d Sr ⁸⁹	1.11×10^{-1}	8.68×10^{-1}	1.45×10^{-1}	1.40×10^{-2}
41d Ru ¹⁰³	7.65×10^{-2}	9.30×10^{-1}	8.00×10^{-2}	5.68×10^{-3}
(54m)Rh ^{103*}	5.52×10^{-3}	9.30×10^{-1}	8.00×10^{-2}	4.11×10^{-4}
33d Te ^{129*}	1.71×10^{-3}	9.63×10^{-1}	4.33×10^{-2}	7.13×10^{-5}
(72m)Te ¹²⁹	8.09×10^{-2}	9.63×10^{-1}	4.33×10^{-2}	3.38×10^{-3}
32d Ce ¹⁴¹	7.41×10^{-2}	9.69×10^{-1}	3.82×10^{-2}	2.74×10^{-3}

Species	Saturation Values ($\phi = 2 \times 10^{13}$ n/cm ² /sec), total watts/g U ²³⁵	Irradiation (5.3 months)	Decay (5.0 months)	Residual $\beta + \gamma$ watts/g U ²³⁵
17.3d Pr ¹⁴³	9.16×10^{-2}	1.00	5.00×10^{-4}	4.57×10^{-5}
12.8d Ba ¹⁴⁰	1.52×10^{-1}	1.00	2.85×10^{-4}	4.33×10^{-5}
(40h)La ¹⁴⁰	7.81×10^{-1}	1.00	3.43×10^{-4}	2.68×10^{-4}
11.3d Nd ¹⁴⁷	5.73×10^{-2}	1.00	1.02×10^{-4}	5.85×10^{-6}
8.05d I ¹³¹	8.01×10^{-2}	1.00	2.51×10^{-6}	2.01×10^{-7}
Total				2.463×10^{-1}

$$2.5 \times 10^4 \text{ g} \times 2.463 \times 10^{-1} = 6.15 \times 10^3 \text{ total watts/day}$$

$$6.15 \times 10^3 / 1.2 \times 10^3 = 5.08 \text{ total watts/gal of waste.}$$

Since there are 3.413 Btu/hr/watt, $5.08 \times 3.413 = 17.3$ Btu/hr/gal.

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