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BUILDUP OF Cf^{252} AND INTERMEDIATE ISOTOPES FROM
 Cm^{244} IN A HIGH FLUX

by

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INTRODUCTION

The possibility that a reactor with a high neutron flux [10^{16} n/(cm^2)(sec)] will be available within a few years made it desirable to calculate the amount of Cf^{252} and intermediate isotopes which could be made from Cm^{244} in such a reactor. In order to make such calculations, it was necessary to evaluate measured cross sections and half-lives and, in some cases, to estimate cross sections which have not been measured. The amounts of isotopes produced relative to the amount of the starting material (Cm^{244}) are presented in tables and graphs.

BUILDUP PATH

BUILDUP EQUATIONS

The buildup of each isotope is represented by an equation below. The following abbreviations are used:

- N ratio of the number of atoms to the initial number of atoms of Cm^{244}
- F flux [in neutrons/(cm^2)(sec)]
- t time of irradiation in seconds
- σ_c capture cross section
- σ_T total cross section, that is, the sum of the capture cross section and the fission cross section.

Isotope	Symbols	Isotope	Symbols
Cm ²⁴⁴	64	Cm ²⁴⁹	69
Cm ²⁴⁵	65	Bk ²⁴⁹	79
Cm ²⁴⁶	66	Cf ²⁵⁰	80
Cm ²⁴⁷	67	Cf ²⁵¹	81
Cm ²⁴⁸	68	Cf ²⁵²	82

The variations of the different isotopes formed in the bombardment as a function of time can be described by a series of linear differential equations. For example, the destruction of Cm²⁴⁴ is described by the equation

$$\frac{dN^{64}}{dt} = -\sigma_T^{64} F N^{64} \quad . \quad (1)$$

The solution of this equation then gives the amount of Cm²⁴⁴ at any time t:

$$N^{64} = e^{-\sigma_T^{64} F t} \quad . \quad (2)$$

Similarly, the variation of Cm²⁴⁵ with time can be represented by the differential equation

$$\frac{dN^{65}}{dt} = \sigma_C^{64} F N^{64} - (\sigma_C^{65} + \sigma_f^{65}) F N^{65} \quad ; \quad (3)$$

the first term represents the rate of formation of Cm²⁴⁵ from Cm²⁴⁴, and the second term represents the rate of destruction of Cm²⁴⁵. The solution of equation 3 is

$$N^{65} = \frac{\sigma_C^{64}}{(\sigma_T^{65} - \sigma_T^{64})} e^{-\sigma_T^{64} F t} + \frac{\sigma_C^{64}}{(\sigma_T^{64} - \sigma_T^{65})} e^{-\sigma_T^{65} F t} \quad . \quad (4)$$

The production of all the isotopes in the buildup chain can be represented by the following general equation:

$$N_i = (\sigma_C^1 \sigma_C^2 \sigma_C^3 \dots \sigma_i^{i-1}) F^{i-1} \sum_{n=1}^i \frac{e^{-T_n t}}{\prod_{\substack{s=1, \\ s \neq n}}^i (T_s - T_n)} F^{(i-1)} \quad (5)$$

where T_i is the sum of processes removing N_i atoms, such as by decay, neutron capture and fission.

As an example, consider the number of atoms of Cf^{252} formed per initial atom of Cm^{244} . Now Cf^{252} is the eighth product formed in the buildup sequence; hence $i = 9$, and n , the running index, will have a maximum value of 9, i.e., there are 9 terms. All 9 terms have the common term $(\sigma_C^1 \sigma_C^2 \dots \sigma_C^{i-1}) F^{(i-1)}$ in their numerator, that is, $\sigma_C^{64} \sigma_C^{65} \sigma_C^{66} \sigma_C^{67} \sigma_C^{68} \sigma_C^{79} \sigma_C^{80} \sigma_C^{81}$, the last σ_C being σ_C^{i-1} . The nine exponential terms are the terms shown below and the product of differences in the denominator will each have 8 terms. The first denominator consists of $\sigma_T^1 (= \sigma_T^{64})$ terms subtracted from all other σ_T terms, except for the term $(\sigma_T^{64} - \sigma_T^{64})$ which is not allowed, i.e., $s \neq n$. The second term contains the denominator in which all the terms consist of subtracting $\sigma_T^2 (= \sigma_T^{65})$ from all other σ_T terms. The terms involving decay have been neglected in this calculation. This permits cancellation of the flux in the numerator and denominator terms.

$$N_{\text{Cf}^{252}} = \sigma_C^{64} \sigma_C^{65} \sigma_C^{66} \sigma_C^{67} \sigma_C^{68} \sigma_C^{79} \sigma_C^{80} \sigma_C^{81} \left[\begin{aligned} & \frac{e^{-\sigma_T^{64} Ft}}{(\sigma_T^{65} - \sigma_T^{64})(\sigma_T^{66} - \sigma_T^{64})(\sigma_T^{67} - \sigma_T^{64})(\sigma_T^{68} - \sigma_T^{64})(\sigma_T^{79} - \sigma_T^{64})(\sigma_T^{80} - \sigma_T^{64})(\sigma_T^{81} - \sigma_T^{64})(\sigma_T^{82} - \sigma_T^{64})} \\ & + \frac{e^{-\sigma_T^{65} Ft}}{(\sigma_T^{64} - \sigma_T^{65}) \dots (\sigma_T^{82} - \sigma_T^{65})} \\ & + \frac{e^{-\sigma_T^{66} Ft}}{(\sigma_T^{64} - \sigma_T^{66}) \dots (\sigma_T^{82} - \sigma_T^{66})} \\ & + \frac{e^{-\sigma_T^{67} Ft}}{(\sigma_T^{64} - \sigma_T^{67}) \dots (\sigma_T^{82} - \sigma_T^{67})} \\ & + \frac{e^{-\sigma_T^{68} Ft}}{(\sigma_T^{64} - \sigma_T^{68}) \dots (\sigma_T^{82} - \sigma_T^{68})} \\ & + \frac{e^{-\sigma_T^{79} Ft}}{(\sigma_T^{64} - \sigma_T^{79}) \dots (\sigma_T^{82} - \sigma_T^{79})} \\ & + \frac{e^{-\sigma_T^{80} Ft}}{(\sigma_T^{64} - \sigma_T^{80}) \dots (\sigma_T^{82} - \sigma_T^{80})} \\ & + \frac{e^{-\sigma_T^{81} Ft}}{(\sigma_T^{64} - \sigma_T^{81}) \dots (\sigma_T^{82} - \sigma_T^{81})} \\ & + \frac{e^{-\sigma_T^{82} Ft}}{(\sigma_T^{64} - \sigma_T^{82}) \dots (\sigma_T^{81} - \sigma_T^{82})} \end{aligned} \right] \quad (6)$$

CROSS SECTIONS

The cross sections used in these calculations were either weighted measured values or values estimated from the amounts of curium and californium isotopes formed in irradiations of heavy elements. Many of these are not accurate and will need to be revised when more data are available. Since the calculated values of amounts of Cf^{252} and intermediate isotopes formed in Cm^{244} irradiations depend in an exponential way on the cross sections used, it is apparent that the calculated values should not be taken literally. Table I lists the cross sections used.

Table I

CAPTURE AND TOTAL CROSS SECTIONS

Isotope	σ_c (barns)	σ_T (barns)	Isotope	σ_c (barns)	σ_T (barns)
Cm^{244}	32	32	Bk^{249}	800	800
Cm^{245}	135	2150	Cf^{250}	2000	2000
Cm^{246}	80	80	Cf^{251}	3800	7500
Cm^{247}	35	70	Cf^{252}	30	30
Cm^{248}	6	6			

CALCULATION OF THE BUILDUP

Because the total cross sections for Cm^{245} , Bk^{249} , Cf^{250} , and Cf^{252} are large, the exponential terms involving these cross sections become small and can be neglected in equations (7) through (14). At the high fluxes considered, the decay of Bk^{249} and Cf^{252} can be ignored. With these assumptions, the buildup equations can be rewritten as follows:

$$N_{\text{Cm}^{245}} = k_1 e^{-\sigma_T^{64} Ft} \quad (7)$$

$$N_{\text{Cm}^{246}} = k_2 e^{-\sigma_T^{64} Ft} + k_3 e^{-\sigma_T^{66} Ft} \quad (8)$$

$$N_{\text{Cm}^{247}} = k_4 e^{-\sigma_T^{64} Ft} + k_5 e^{-\sigma_T^{66} Ft} + k_6 e^{-\sigma_T^{67} Ft} \quad (9)$$

$$N_{\text{Cm}^{248}} = k_7 e^{-\sigma_T^{64} Ft} + k_8 e^{-\sigma_T^{66} Ft} + k_9 e^{-\sigma_T^{67} Ft} + k_{10} e^{-\sigma_T^{68} Ft} \quad (10)$$

$$N_{\text{Bk}^{249}} = k_{11} e^{-\sigma_T^{64} Ft} + k_{12} e^{-\sigma_T^{66} Ft} + k_{13} e^{-\sigma_T^{67} Ft} + k_{14} e^{-\sigma_T^{68} Ft} \quad (11)$$

$$N_{Cf^{250}} = k_{15} e^{-\sigma_T^{64} Ft} + k_{16} e^{-\sigma_T^{66} Ft} + k_{17} e^{-\sigma_T^{67} Ft} + k_{18} e^{-\sigma_T^{68} Ft} \quad (12)$$

$$N_{Cf^{251}} = k_{19} e^{-\sigma_T^{64} Ft} + k_{20} e^{-\sigma_T^{66} Ft} + k_{21} e^{-\sigma_T^{67} Ft} + k_{22} e^{-\sigma_T^{68} Ft} \quad (13)$$

$$N_{Cf^{252}} = k_{23} e^{-\sigma_T^{64} Ft} + k_{24} e^{-\sigma_T^{66} Ft} + k_{25} e^{-\sigma_T^{67} Ft} + k_{26} e^{-\sigma_T^{68} Ft} \\ + k_{27} e^{-\sigma_T^{82} Ft} \quad (14)$$

Using these simplified equations it was possible to write a program for an IBM-610 computer. The computer was used to calculate accurate values for the coefficients in these equations and also to solve the equations as a function of total flux (Ft).

RESULTS

Table II lists the number of atoms of the heavy curium isotopes per initial Cm^{244} atom as a function of total flux (Ft) intervals. Similarly, Table III shows the buildup of Bk^{249} and the californium isotopes. Figure 1 is a linear plot of the values listed in Table II and Table III. In Figure 2 the initial part of the buildup is plotted on a logarithmic scale.

Table II

NUMBER OF ATOMS OF THE HEAVY CURIUM ISOTOPES PER INITIAL Cm^{244} ATOM

Flux (ft)	Days at 3×10^{15} Flux	$N_{\text{Cm}^{244}}$	$N_{\text{Cm}^{245}}$	$N_{\text{Cm}^{246}}$	$N_{\text{Cm}^{247}}$	$N_{\text{Cm}^{248}}$
6×10^{21}	23	.825306	.012468	.008164	.001764	.0000887
1.2×10^{22}	46	.681131	.010290	.012294	.005346	.000813
1.8×10^{22}	69	.562142	.008492	.013584	.008669	.002246
2.4×10^{22}	93	.463940	.007009	.013339	.011003	.004216
3.0×10^{22}	116	.382892	.005784	.012325	.012261	.006484
3.6×10^{22}	139	.316004	.004774	.010986	.012612	.008834
4.2×10^{22}	162	.260800	.003940	.009571	.012296	.011099
4.8×10^{22}	185	.215240	.003251	.008211	.011542	.013171
5.4×10^{22}	208	.177639	.002683	.006969	.010537	.014985
6.0×10^{22}	231	.146606	.002214	.005871	.009420	.016514
6.6×10^{22}	255	.120995	.001828	.004919	.008286	.017755
7.2×10^{22}	278	.099858	.001508	.004106	.007198	.018722
7.8×10^{22}	301	.082413	.001245	.003417	.006190	.019439
8.4×10^{22}	324	.068016	.001027	.002837	.005282	.019932
9.0×10^{22}	347	.056134	.000848	.002352	.004478	.020231
1.02×10^{23}	394	.038235	.000577	.001612	.003173	.020360
1.14×10^{23}	440	.026043	.000393	.001101	.002218	.020026
1.26×10^{23}	486	.017738	.000267	.000751	.001536	.019386
1.38×10^{23}	532	.012082	.000182	.000512	.001058	.018558
1.50×10^{23}	579	.008229	.000124	.000349	.000726	.017625
1.62×10^{23}	625	.005605	.000084	.000238	.000497	.016645
1.74×10^{23}	671	.003818	.000057	.000162	.000339	.015656
1.86×10^{23}	718	.002600	.000039	.000110	.000231	.014683
1.98×10^{23}	764	.001771	.000026	.000075	.000158	.013741
2.10×10^{23}	810	.001206	.000018	.000051	.000107	.012839
2.22×10^{23}	856	.000821	.000012	.000034	.000073	.011983
2.34×10^{23}	903	.000559	.000008	.000023	.000050	.011176
2.46×10^{23}	949	.000381	.000005	.000016	.000034	.010416
2.58×10^{23}	995	.000259	.000003	.000011	.000023	.009703
2.70×10^{23}	1042	.000176	.000002	.000007	.000015	.009038
2.82×10^{23}	1088	.000120	.000001	.000005	.000010	.008415
2.94×10^{23}	1134	.000082	.000001	.000003	.000007	.007834

Table III

NUMBER OF ATOMS OF Bk^{249} , Cf^{250} , Cf^{251} AND Cf^{252} PER INITIAL Cm^{244} ATOM

Flux (ft)	Days at 3×10^{15} Flux	$N_{\text{Bk}^{249}}$	$N_{\text{Cf}^{250}}$	$N_{\text{Cf}^{251}}$	$N_{\text{Cf}^{252}}$
6×10^{21}	23	0.002×10^{-4}	0.005×10^{-5}	0.001×10^{-5}	0.000×10^{-3}
1.2×10^{22}	46	0.046×10^{-4}	0.16×10^{-5}	0.04×10^{-5}	0.000×10^{-3}
1.8×10^{22}	69	0.14×10^{-4}	0.53×10^{-5}	0.14×10^{-5}	0.01×10^{-3}
2.4×10^{22}	93	0.28×10^{-4}	1.08×10^{-5}	0.29×10^{-5}	0.05×10^{-3}
3.0×10^{22}	116	0.45×10^{-4}	1.74×10^{-5}	0.46×10^{-5}	0.12×10^{-3}
3.6×10^{22}	139	0.63×10^{-4}	2.45×10^{-5}	0.65×10^{-5}	0.22×10^{-3}
4.2×10^{22}	162	0.80×10^{-4}	3.13×10^{-5}	0.83×10^{-5}	0.34×10^{-3}
4.8×10^{22}	185	0.96×10^{-4}	3.78×10^{-5}	1.00×10^{-5}	0.48×10^{-3}
5.4×10^{22}	208	1.09×10^{-4}	4.34×10^{-5}	1.16×10^{-5}	0.62×10^{-3}
6.0×10^{22}	231	1.22×10^{-4}	4.83×10^{-5}	1.28×10^{-5}	0.78×10^{-3}
6.6×10^{22}	255	1.31×10^{-4}	5.22×10^{-5}	1.39×10^{-5}	0.93×10^{-3}
7.2×10^{22}	278	1.39×10^{-4}	5.53×10^{-5}	1.48×10^{-5}	1.07×10^{-3}
7.8×10^{22}	301	1.45×10^{-4}	5.77×10^{-5}	1.53×10^{-5}	1.21×10^{-3}
8.4×10^{22}	324	1.49×10^{-4}	5.94×10^{-5}	1.58×10^{-5}	1.34×10^{-3}
9.0×10^{22}	347	1.51×10^{-4}	6.05×10^{-5}	1.61×10^{-5}	1.45×10^{-3}
1.02×10^{23}	394	1.53×10^{-4}	6.11×10^{-5}	1.63×10^{-5}	1.64×10^{-3}
1.14×10^{23}	440	1.51×10^{-4}	6.03×10^{-5}	1.61×10^{-5}	1.76×10^{-3}
1.26×10^{23}	486	1.46×10^{-4}	5.85×10^{-5}	1.56×10^{-5}	1.84×10^{-3}
1.38×10^{23}	532	1.40×10^{-4}	5.61×10^{-5}	1.50×10^{-5}	1.87×10^{-3}
1.50×10^{23}	579	1.33×10^{-4}	5.33×10^{-5}	1.42×10^{-5}	1.86×10^{-3}
1.62×10^{23}	625	1.26×10^{-4}	5.04×10^{-5}	1.34×10^{-5}	1.83×10^{-3}
1.74×10^{23}	671	1.18×10^{-4}	4.74×10^{-5}	1.26×10^{-5}	1.77×10^{-3}
1.86×10^{23}	718	1.11×10^{-4}	4.45×10^{-5}	1.19×10^{-5}	1.71×10^{-3}
1.98×10^{23}	764	1.03×10^{-4}	4.16×10^{-5}	1.11×10^{-5}	1.63×10^{-3}
2.10×10^{23}	810	0.97×10^{-4}	3.89×10^{-5}	1.04×10^{-5}	1.55×10^{-3}
2.22×10^{23}	856	0.91×10^{-4}	3.6×10^{-5}	0.97×10^{-5}	1.46×10^{-3}
2.34×10^{23}	903	0.84×10^{-4}	3.4×10^{-5}	0.90×10^{-5}	1.38×10^{-3}
2.46×10^{23}	949	0.79×10^{-4}	3.2×10^{-5}	0.84×10^{-5}	1.30×10^{-3}
2.58×10^{23}	995	0.73×10^{-4}	2.9×10^{-5}	0.79×10^{-5}	1.21×10^{-3}
2.70×10^{23}	1042	0.68×10^{-4}	2.7×10^{-5}	0.73×10^{-5}	1.14×10^{-3}
2.82×10^{23}	1088	0.64×10^{-4}	2.5×10^{-5}	0.68×10^{-5}	1.06×10^{-3}
2.94×10^{23}	1134	0.59×10^{-4}	2.4×10^{-5}	0.63×10^{-5}	0.99×10^{-3}

Figure 1 Buildup of Cf²⁵² and Intermediate Isotopes from Cm²⁴⁴.

Figure 2 Buildup of Cf²⁵² and Intermediate Isotopes from Cm²⁴⁴, Plotted on a Semi-logarithmic Scale.