

Elements in Region of Platinum Formed by Fusion in Fission Explosions*

L. MARSHALL†

New York University, New York, New York

and

Brookhaven National Laboratory, Upton, New York

(Received 21 August 1962)

The fission products in a fission explosion at the time of the last few generations before explosion are sufficiently numerous that the probability of reaction with each other becomes measurable. In this paper the probability of fusion is calculated. It is found that the fusion of two light fission fragments is the only possible process of this type. For explosion of a mass of 14 kg of uranium or plutonium having a 25% efficiency the yield of platinum group elements is estimated as 1.2×10^{14} atoms. Although the kind of nucleus produced instantaneously with highest probability has $Z=75$ and $A=190$, the excess of kinetic energy is such as to boil off between 9 and 18 neutrons forming elements $A=175$ to 182, on the low side of the stability curve. In view of the recently developed theories of supernovae explosion, involving production of Cf^{254} and other fissile nuclei, it may be important to consider the subsequent fusion of fission fragments in supernovae. If this process is sufficiently important, it may affect the astrophysical abundances of elements in the region of platinum.

INTRODUCTION

THE fission products in a bomb at the time of the last few generations before explosion are sufficiently numerous so that the probability of reaction with each other becomes measurable. In this paper the probability of fusion is calculated. It is found that the fusion of two light fragments is the only possible process of this type, the fusion of two heavy fragments or of a light and a heavy fragment being improbable because in these cases the Coulomb barrier is much higher. For a bomb containing 14 kg of uranium or plutonium and having a 25% efficiency the yield of platinum group elements is estimated as 1.2×10^{14} atoms. Although the nucleus produced instantaneously with highest probability has $Z=75$, $A=190$, the excess of kinetic energy is such as to boil off between 9 and 18 neutrons forming a spectrum of nuclei having $A=175$ –182. Therefore, the nuclei formed will be on the low side of the stability curve. Most nuclei in this region decay by K capture although two positron emitters, ${}_{71}\text{Lu}^{170}$ and ${}_{69}\text{Tm}^{166}$, are known and perhaps others may be discovered.

Consider an average light fragment, $Z=37$, $A=96$, and an average heavy fragment, $Z=55$, $A=138$, which have just separated from each other. The average kinetic energy of the light fragment is 99 MeV, and of the heavy fragment is $168-99=69$ MeV (168 MeV being the total kinetic energy of the two fragments on the average).

A freshly formed light and heavy fragment have almost enough energy to recombine to form the original nucleus $Z=92$, $A=235$. But two heavy fragments are unable to combine to form a transuranic element, e.g., $Z=110$, $A=276$, for the reason that the reaction is endothermic by more than the maximum relative kinetic energy of the two, e.g., 138 MeV.

The situation is more favorable for the light fragments. Two freshly formed light elements which collide

may react to form such an element as $Z=74$, $A=192$. The mass difference between nucleus $Z=74$, $A=192$ and two nuclei $Z=37$, $A=96$, calculated from the semi-empirical atomic mass formula has the value 89 MeV. The height of the Coulomb barrier¹ is 143 MeV. The initial relative kinetic energy for a head on collision of two average light fragments is $2 \times 99 = 198$ MeV, which is greater than both the mass difference and the barrier. The barrier height, being greater than the mass difference, is taken here as the limiting factor; we assume that a light fragment may react in fusion with other light fragments until it has lost of the order of $\frac{1}{2}(198-143)=27.5$ MeV, i.e., until it has energy 71.5 MeV.

Estimate of the Probability of Fusion of Two Light Fragments

In order to take into account the collisions of two fragments of unequal energy in the energy range in which fusion may occur, we define a quantity q as the total number of light fission fragments per cubic centimeter and per unit volume of velocity space having velocity between v and $v+dv$.

(1) Calculation of q : Let N be the total number of light fission fragments produced during the duration T of fission in the bomb ($T \sim 2 \times 10^{-8}$ sec). For 25% yield in a bomb containing 14 kg of Pu or U,

$$N = (1.4 \times 10^4 / 235) \times \frac{1}{4} \times 6 \times 10^{23} = 8.91 \times 10^{24}.$$

Let Δt be the time for a light fission fragment to lose 1 cgs unit of velocity. Then at any instant, $n(v)dv$, the total number of light fission fragments with velocity between v and $v+dv$, is $N\Delta t/T$.

To estimate Δt , the rate of energy loss dE/dx of a light fission fragment to a 10-keV Maxwellian distribution of

* Work performed under the auspices of the U. S. Atomic Energy Commission.

† Based on a Los Alamos Report LA-1299, 1951 (unpublished).

¹ Computed on the assumption that the nuclear radius is $1.5 \times 10^{-13} A^{1/3}$ cm.

electrons is needed. Assume the reaction to occur mainly at normal density, N_1 .

$$-\frac{dE}{dx} \cong N_1 \frac{4(\pi)^{1/2} Z_1^2 e^4}{(ET)^{1/2}} \left(\frac{1}{1830A} \right)^{1/2} \left[\frac{2E}{3T} \right] \ln \frac{P_1}{P_0}$$

$$N_1 = 92 \times 19 / 235 \times 6 \times 10^{23} = 4.46 \times 10^{24} \text{ cm}^{-3}$$

= density of electrons,

(where 92 is the number of electrons per nucleon, and 19 g/cm³ is the density of uranium).

We use these quantities together with the values

$$E = 99 \text{ MeV}, \quad T = 10 \text{ keV}, \quad A = 96, \quad Z = 37,$$

and find

$$-dE/dx = 0.0926 \text{ erg/cm.}$$

The time to lose 1 cgs unit of velocity for a light fission fragment is

$$\Delta t = 1/(dv/dt) = M/(dE/dx) = 1.76 \times 10^{-21} \text{ sec,}$$

from which it follows that

$$n(v)dv = N(\Delta t/T) = 7.85 \times 10^{11}.$$

The quantity q is defined per unit volume of velocity space and per cm³ and so contains the reaction volume

$$V = \frac{1.4 \times 10^4 \text{ g}}{19 \text{ g/cm}^3} = 736 \text{ cm}^3.$$

It is evaluated from the relationship

$$q = [n(v)/4\pi v^2](1/V) = 4.31 \times 10^{-11}.$$

(2) Calculation of total velocity space from which collisions effecting fusion can occur against a given element of velocity space. For an average light fission fragment the maximum velocity is computed from its average kinetic energy or

$$v_0 = 1.41 \times 10^9 \text{ cm/sec.}$$

The minimum relative velocity at which a fusion can occur is computed from the height of the corresponding Coulomb barrier, 143 keV, as

$$V_1 = 2.39 \times 10^9 \text{ cm/sec.}$$

Consider a fission fragment of velocity v in a particular increment of velocity space d^3v . It may produce a fusion by collision with any particle lying in the velocity space inside the sphere of radius v_0 about the origin and outside the sphere of radius V_1 circumscribed about d^3v . This volume between the two spheres is given by

$$\int_0^{[2(v_0+v-V_1)/(1/v_0-1/V_1)]^{1/2}} \left[(v_0+v-V_1) + \frac{x^2}{2V_1} - \frac{x^2}{2v_0} \right] 2\pi x dx$$

$$= \pi \frac{(v_0+v-V_1)^2}{(1/v_0) - (1/V_1)}$$

(3) Calculation of total number of collisions of light fission fragments. A simplifying approximation follows from the fact that $V_1 \sim 2v_0$. Since the upper limit on the velocity v of a fission fragment is v_0 , the lower limit is $V_1 - v_0$ which is not greatly different from v_0 . On this account, in the integral below, v may be replaced by $V_1/2$, and V_1 may be substituted for v_{rel} . Then the number of collisions per sec cm³ are given by

$$\frac{1}{2} \int_{V_1-v_0}^{v_0} \sigma v_{rel} q^2 \left[\frac{\pi(v_0+v-V_1)^2}{(1/v_0) - (1/V_1)} \right] 4\pi v^2 dv$$

$$\sim \frac{1}{2} \sigma V_1 q^2 4\pi^2 \left(\frac{V_1}{2} \right)^2 \frac{V_1 v_0}{V_1 - v_0}$$

$$\times \int_{V_1-v_0}^{v_0} (v_0+v-V_1)^2 d(v_0+v-V_1)$$

$$\sim 2\pi^2 q^2 \sigma \frac{V_1^4 (2v_0 - V_1)^3}{4 \cdot 3}.$$

In this expression, σ is the cross section for fusion of two light fission fragments. It is not known, but is assumed to be 10^{-24} cm² for two fragments having relative energy greater than the Coulomb barrier.

The total number of collisions is

$$TV \left[2\pi^2 q^2 \sigma \frac{V_1^4 (2v_0 - V_1)^3}{4 \cdot 3} \right],$$

where

$$(2v_0 - V_1) = 0.43 \times 10^9 \text{ cm/sec.}$$

One finds with these assumptions that the total number of fusions is 1.2×10^{14} for light fission fragments.

Distribution of Fusion Nuclei

This number of atoms is distributed among several nuclear species. In order to get an idea of this distribution we may fit the light fission fragment yield curve roughly by a Gaussian (where A is the mass number of a chain

$$\exp \left[- \left(\frac{96 - A_1}{8} \right)^2 \right].$$

The probability of formation of element $A = A_1 + A_2$ is proportional to

$$\int \exp \left[- \left(\frac{96 - A_1}{8} \right)^2 \right] \exp \left[- \left(\frac{96 - A_2}{8} \right)^2 \right] dA_1$$

$$= \exp \left[- \left(\frac{192 - A}{11.3} \right)^2 \right],$$

except for the effect of variation in the Gamow barrier, and of increase in initial kinetic energy of a fission fragment as its mass decreases. These two factors affect the solid angle over which collisions may occur to form a fusion.

In general, most collisions are not head on. The relative kinetic energy of a collision of two particles of equal energy depends on the angle at which they collide. The relative velocity must be averaged over all angles for which E_{rel} is greater than the Coulomb barrier. This

TABLE I. Probability of formation of instantaneous elements by fusion (not corrected for boiling off of neutrons).

Instantaneous A	Instantaneous Z	Gaussian factor	Gamow factor	Relative yield
202	79	0.46	0.17	0.24
200	79	0.61	0.20	0.38
198	78	0.75	0.23	0.54
196	77	0.88	0.26	0.72
194	76	0.97	0.28	0.85
192	75	1.00	0.31	0.97
190	75	0.97	0.33	1.00
188	74	0.88	0.35	0.96
186	73	0.75	0.37	0.87
184	72	0.61	0.38	0.72
182	71	0.46	0.40	0.57
180	71	0.32	0.42	0.42
178	70	0.22	0.43	0.30
176	69	0.13	0.44	0.18

correction may be made crudely as follows. Let the angle between the two colliding fragments in the lab system be θ , and assume for simplicity that the two fragments have equal energy E . Then

$$v_{rel} = 2v \sin(\theta/2),$$

$$E_{rel} = \frac{1}{2}(M/2)v_{rel}^2 = 2\frac{1}{2}Mv^2 \sin^2(\theta/2) = 2E \sin^2(\theta/2).$$

For each value of E the limiting angle θ such that E_{rel} is not less than E_B , the height of the barrier, is given by

$$\sin(\theta/2) = (E_B/2E)^{1/2}.$$

The average value of the relative velocity times the effective solid angle is given by

$$\begin{aligned} (\Delta\Omega)v_{rel} &= 2v \int_{\theta}^{\pi} \frac{1}{2} \sin(x/2) \sin x dx \\ &= \frac{4}{3}v [1 - \sin^3(\theta/2)] = \frac{4}{3}v [1 - (143/2E)^{3/2}]. \end{aligned}$$

For example, consider the element $Z=82$, $A=208$ to be made by fusion of two atoms $Z=41$, $A=104$. The barrier is 171 MeV. The kinetic energy of each fragment is 94 MeV. The maximum relative kinetic energy of the two fragments is 188 MeV. Therefore, fusion can occur until each fragment has lost $\frac{1}{2}(188-171)=8.5$ MeV. The effective mean energy of the fragment is $94-8.5/2=90$ MeV. The effective value of $(\Delta\Omega)v_{rel}$ is $0.093v$.

For the formation of a nucleus $Z=76$, $A=192$ from two atoms having $Z=36$, $A=96$ we have already found the barrier to be 143 MeV and the mean energy 85.3 MeV. One obtains

$$(\Delta\Omega)v_{rel} = 0.31v.$$

At the light end of the spectrum we now need the correction for formation of a nucleus $Z=70$, $A=176$ from two fragments having $Z=35$, $A=88$. The Gamow barrier is 133 MeV. The initial kinetic energy is 105 MeV. The maximum kinetic energy is 210 MeV, therefore, each fragment can lose no more than $\frac{1}{2}(210-133)=38.5$ MeV. The effective energy of each fragment is $105-\frac{1}{2}38.5=86$ MeV. The effective correction factor is

$$(\Delta\Omega)v_{rel} = 0.43v.$$

A probable distribution of fusion products is shown in Table I. The relative probability of formation has been computed for the instantaneous A value. However, for all these nuclei there is enough kinetic energy in excess of the energy needed for the reaction so that many neutrons will boil off.

The average binding energy of a neutron in this region is 6 MeV. At the heavy end, e.g., Au, the available KE ranges from 171 to 188 MeV, and the endothermic energy of fusion is 101 MeV. Therefore, between 12 and 14 neutrons will boil off the nucleus $Z=79$, $A=202$ producing nuclei having $Z=79$ and having a spectrum of mass values, $A=190-188$. Since these elements are far below the curve of stability they will decay rapidly by K capture or by positron emission. The significant quantity will then be not Z but A .

Similarly, in the region of the most probable instantaneous element $Z=75$, $A=190$ the available KE is 198 to 143 MeV and the mass difference is 89 MeV. There will be from 9 to 18 neutrons boiled off producing mass chains of 172 to 181. And at the light end of the distribution, e.g., $Z=70$, $A=78$, the available KE ranges from

TABLE II. Probability of formation of chains of mass A (after consideration of the effect of boiling off of neutrons).

A	Relative probability of formation	Known activity at end of chain
190	0.15	Unknown
188	0.42	Unknown
186	0.60	Unknown
184	0.73	Unknown
182	0.83	${}^{76}\text{Re}^{182}$ (13 h, 64 h) K , e^-
180	0.94	Unknown
178	1.00	${}^{78}\text{Ta}^{178}$ (15.4 day) K , e^-
176	0.97	${}^{78}\text{Ta}^{176}$ (8 h) K , e^-
174	0.88	Unknown
172	0.80	${}^{71}\text{Lu}^{172}$ (>100 day) K , e^-
170	0.70	${}^{71}\text{Lu}^{170}$ (2.1 day) β^+
168	0.58	Unknown
166	0.47	${}^{69}\text{Tm}^{166}$ (7.7 h) β^+
164	0.32	Unknown
162	0.23	Unknown
160	0.16	${}^{67}\text{Ho}^{160}$ (20 m)

210 to 133 MeV, while mass difference is 80 MeV. One expects, therefore, masses ranging from 169 to 156 corresponding to loss of 22 to 9 neutrons. For the sake of brevity, the probability of formation is computed for every other mass number only, and is listed in Table II.

The effect of boiling off of neutrons is to produce the modified distribution of nuclei shown in Table II. Of these elements, two at least are positron emitters and so more easily detected. It is possible that there are others among the unknown isotopes which are positron active. The activity which can be collected is somewhat marginal. Consider for example the collected activity to be expected for the positron emitter ${}_{69}\text{Tm}^{166}$; assume 10^{-10} of the bomb is collected. Then the collected radio activity of ${}_{69}\text{Tm}^{166}$ produces one decay per minute.

The yield of fusion elements is proportional to

$$(\sigma/v_{\text{rel}})\theta^3\epsilon^2(V/T),$$

where θ is the electron temperature, ϵ is the efficiency, and V is the reaction volume. In the case of the hydro-

gen bomb both the volume and the electron temperature are expected to be larger than in an ordinary fission explosion. The hydrogen bomb, therefore, offers an opportunity for an increased yield of nuclear species produced by fission fragment fusion.

In view of the recently developed theories of supernovae explosion,² involving production of Cf^{254} and other fissile nuclei, it may be important to consider the subsequent fusion of fission fragments in supernovae. If this process is sufficiently important, it may affect the astrophysical abundances of elements in the region of platinum.

² G. R. Burbidge, F. Hoyle, E. M. Burbidge, R. F. Christy, and W. A. Fowler, *Phys. Rev.* **103**, 1143 (1956).

Elastic and Inelastic Scattering of 31.1-MeV Protons by Carbon-12*

J. KIRK DICKENS,[†] DAVID A. HANER, AND CHARLES N. WADDELL

University of Southern California, Los Angeles, California

(Received 17 August 1962)

The angular distributions of 31.1-MeV protons scattered by the ground state and the 4.4-, 7.7-, 9.6-, 12.7-, 14.0-, 15.1-, and 16.1-MeV excited states have been measured. The differential cross sections for the elastic scattering have been analyzed using the diffuse-surface optical model of the nucleus for a wide range of parameters. This analysis indicates that at this energy the best fit requires a potential characterized by volume, as well as surface, absorption. The angular distributions of the inelastically scattered protons are peaked forward and have been compared to predictions of direct interaction theories.

I. INTRODUCTION

THE recent optical-model analysis by Nodvik, Duke, and Melkanoff of the elastic scattering of 12- to 19-MeV protons by carbon has shown that excellent fits to the experimental data can be obtained for a light nucleus.¹ The most striking features of the results are the thin absorptive shell and the absence of volume absorption that characterize the optical-model potential over most of the studied energy range. However, for $17.8 \leq E_p \leq 18.9$ MeV the analysis indicates either a broadening of the absorptive part of the potential or the necessity for including volume absorption, or, possibly, both.

This work also emphasizes the need for accurate experimental data at small energy intervals over a sizeable range of incident energies. As the first step in an experimental program to extend the measurements of elastic scattering from carbon to the 20- to 30-MeV

range, the results obtained at the full energy of the U.S.C. Linac are presented in this paper.

The angular distributions of protons inelastically scattered from various excited states of C^{12} were measured simultaneously. These differential cross sections are peaked forward and are not symmetric about 90° , indicating the presence of a direct interaction mechanism rather than the formation of a compound nucleus.² The measured angular distributions are compared to the predictions of several existing direct interaction theories.³⁻⁵

II. EXPERIMENTAL DETAILS

A. General

The University of Southern California proton linear accelerator has been described by Alvarez *et al.*,⁶ and

* Work supported in part by the U. S. Atomic Energy Commission.

[†] Present address: Oak Ridge National Laboratory, Oak Ridge, Tennessee.

¹ J. S. Nodvik, C. B. Duke, and M. A. Melkanoff, *Phys. Rev.* **125**, 975 (1962).

² W. Tobocman, *Theory of Direct Interactions* (Oxford University Press, New York, 1961).

³ N. Austern, S. T. Butler, and H. McManus, *Phys. Rev.* **92**, 350 (1953).

⁴ J. S. Blair, *Phys. Rev.* **115**, 928 (1959).

⁵ J. D. Templin, thesis, University of California at Los Angeles, 1961 (unpublished).

⁶ L. W. Alvarez, H. Bradner, J. V. Franck, H. Gordon, J. D.