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NUCLEAR TRANSMUTATIONS USING ACCELERATED CARBON IONS

Jack Marvin Hollander

July 1951

Berkeley, California

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NUCLEAR TRANSMUTATIONS USING ACCELERATED CARBON IONS

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ABSTRACT

As part of a general heavy ion acceleration program, this work has been the investigation of some nuclear reactions induced by carbon nuclei accelerated in the Berkeley 60-inch cyclotron.

Excitation experiments have been done with the $\text{Au}^{197}(\text{C},4n)\text{At}^{205}$ and $\text{Au}^{197}(\text{C},6n)\text{At}^{203}$ reactions using the stacked foil technique, and the results are discussed in terms of the statistical theory of nuclear reactions. Cross sections for these two reactions are also estimated.

The fission of uranium with carbon ions has been studied, and the observed cross section compared with that estimated from yields in the $\text{U}^{238}(\text{C},6n)\text{Cf}^{244}$ reaction. The general theory of carbon ion induced fission is discussed.

Bombardments of copper with carbon ions have resulted in the production of a new isotope of bromine, tentatively assigned to Br^{74} produced by a $\text{Cu}^{65}(\text{C},3n)\text{Br}^{74}$ reaction. Also observed was 1.6 hour Br^{75} . The mass assignments of Br^{76} and Br^{77} have been verified by bombarding As_2O_3 with helium ions of various energies.

To the extent that these novel reactions have been studied, the indications are that carbon ions behave as "normal" nuclear projectiles, and that their reactions can be described adequately in terms of more or less "standard" nuclear models.

NUCLEAR TRANSMUTATIONS USING ACCELERATED CARBON IONS

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I. GENERAL CONSIDERATIONS

The cyclotron has been a very useful device for accelerating positive ions to high energies, where they may be used in the study of nuclear reactions. Hydrogen, deuterium, and helium nuclei have thus far been successfully accelerated, and have assumed the role of commonplace research tools in nuclear physics. However, a glance at the equations of the cyclotron will show that, in theory at least, many other nuclei could be accelerated in the same machines.

The centripetal force on a particle in a magnetic field (directed perpendicular to its path) is given by:

$$F = B \cdot e \cdot v$$

in which B = magnetic field intensity

e = charge on the particle

v = velocity of the particle.

The path of a particle traveling at constant velocity in a uniform magnetic field is a circle, and one may state that the centrifugal force and the magnetic force are equal, so that:

$$\frac{m v^2}{r} = B e v$$

$$\text{or } v = B \frac{e}{m} r$$

in which r is the radius of the circular path. The period of revolution:

$$\tau = \frac{2 \pi r}{v} = \frac{2 \pi}{B \frac{e}{m}}$$

which is independent of the radius, and (for a given field) depends only upon the charge to mass ratio of the particle. Thus, a given cyclotron with a frequency of oscillation set to be in resonance for a certain e/m ratio should be able to accelerate any ion of that ratio.

The Berkeley 60-inch cyclotron normally accelerates ions with

$$\frac{e}{m} = \frac{1}{2} \frac{e}{\mu}$$

where e is the electronic charge and μ the mass unit. Small variation in mass of the several ions is compensated for by appropriate adjustment of the magnetic field, B . The following ions have $e/m = 1/2$, and should be in resonance: H_2^{1+} , H^{2+} , He^{4++} , Li^{6+++} , B^{10++++} , $Cl^{12+++++}$, N^{14+7} , O^{16+8} , etc. Currently, the feasibility of using all of the above ions in transmutation work is being studied, and the present research was initiated as a phase of that program.

Since these ions would emerge from the cyclotron with the same velocity, their kinetic energies would be in the ratio of their masses. This can be seen, since for ions in resonance

$$v = \frac{2 \pi r}{\tau}$$

$$v_R = \frac{2 \pi R}{\tau} \quad \text{where } R = \text{radius of the dee}$$

$$E_R = \frac{2 m \pi^2 R^2}{\tau^2}$$

$$= (\text{constant}) m \quad (\text{for a given machine}).$$

The 60-inch cyclotron produces deuterons of 19 Mev, so the following energies would be available from these heavy ions:

$$\text{Li}^6(+3) = 57 \text{ Mev}$$

$$\text{B}^{10}(+5) = 95 \text{ Mev}$$

$$\text{C}^{12}(+6) = 114 \text{ Mev}$$

$$\text{N}^{14}(+7) = 133 \text{ Mev}$$

$$\text{O}^{16}(+8) = 152 \text{ Mev.}$$

It should also be possible to keep certain sub-multiples of the above charged ions in resonance. For example, an ion having $1/3(e/m)_{\text{std.}}$, such as $\text{Li}^6(+1)$, would travel at one-third the velocity of an ion with $e/m = 1/2$. Once in resonance, this ion would feel an attractive impulse once in every third reversal of polarity of the oscillator; the rest of the time it would spend in the field-free region inside the dees. The $\text{Li}^6(+1)$ would emerge with one-third the velocity, or one-ninth the energy of the $\text{Li}^6(+3)$ accelerated in the same machine.

An ion of $1/5(e/m)_{\text{std.}}$ could be accelerated in the $1/5$ harmonic, but would attain only $1/25$ the energy of its corresponding ion with $e/m = 1/2$. It should not be possible to utilize the even harmonics, for any ion would be out of resonance after only half a revolution.

Thus, if their charges could be maintained, one would also expect to find the following ions accelerated:

$$\text{Li}^6(+1) = 6.3 \text{ Mev}$$

$$\text{C}^{12}(+2) = 12.7 \text{ Mev}$$

$$\text{B}^{10}(+1) = 3.8 \text{ Mev}$$

$$\text{N}^{14}(+1) = 2.7 \text{ Mev.}$$

Acceleration of such ions would be feasible, though, only in a cyclotron whose dees are fairly close, and which is operating at a high dee voltage, so that on the first revolution the ion is already circulating at a large enough radius to ensure that it will be effectively out of the r.f. field. Otherwise, the ions would be immediately thrown out of phase.

The first successful acceleration of heavy ions was recorded by C. A. Tobias,¹ who used CO_2 as the source of ions in the 60-inch cyclotron. He was able to identify high energy carbon ions in the external beam in the presence of residual helium ions and protons by means of cloud chamber collision photographs, and also by the height of the carbon pulses in an oscilloscope. Because of the very small carbon ion current obtained (around 10^{-6} microamperes) he did not attempt to identify whether nuclear transmutations could actually be initiated by these carbon ions.

At the start of this work, the external carbon ion beam was investigated in the same manner as that of Tobias, and the similar conclusion was reached that the intensity of the beam was far too low for meaningful transmutation studies to be carried on. At best, 10^6 ions/second were being collected, which corresponds to 10^{-6} microamperes.

Judging from the fact that the internal deuteron or helium ion beam current is usually ten to twentyfold greater than the corresponding external beam, one was naturally encouraged to hope for similar enrichment of the carbon ion beam. Since at first no very sensitive current measuring device was available for use with the

internal beam, radioactivation was selected as the means of beam detection, and this has continued to be the most reliable monitor.

Selection of Monitor

The utilization of gold as a monitor for the carbon ion beam was prompted by the following considerations:

(a) (C,xn) reactions on gold would lead to the production of recently studied² neutron deficient isotopes of astatine ($_{85}\text{At}$) whose identification could be effected by analysis of their half-lives and alpha particle energies. The particular astatine isotopes sought could not be formed by helium ions on bismuth impurities or by helium ions or deuterons on heavier elements...

(b) The fact that these isotopes are alpha radioactive would serve to distinguish them even in the presence of a great excess of beta radioactivity (products of deuteron and helium ion reactions on gold due to the large amounts of residual beams in the cyclotron), both by simple counting in an alpha scaler and by pulse height analysis.³

(c) The physical properties of gold seemed to be well suited for use as a monitor: it is readily available in the form of very thin foils; it is ductile, fairly high melting (1060°C), and is obtainable in very high purity.

(d) Gold has only one stable isotope, $_{79}\text{Au}^{197}$, presenting an ideal situation for beam monitoring and for excitation function experiments.

Several trial bombardments of gold using the internal carbon beam had yielded a fair amount of alpha activity, which was identified as being due to astatine.⁴ With this encouragement, gold was selected as the monitor, and studies were begun on the Au(C,xn)At reactions.

Experimental Technique

It had been found by tracer experiments that astatine can be quickly and quantitatively separated from gold (with which it had been coprecipitated by SO₂ reduction of the gold) by heating the gold to melting in a quartz cup and collecting the volatilized astatine on the tip of a cold "finger" suspended in the cup. Melting the gold was sufficient but not necessary for this separation, because the coprecipitated astatine tracer could also be volatilized out of the solid at considerably lower temperatures. However, the rate of this alternate process was much slower and quite temperature sensitive, for it is essentially a process of diffusion of a gas through a solid. Thus the yields of astatine recovered in the latter procedure were quite variable, depending upon the length of time and the temperature of heating. Results of a typical experiment are given to illustrate the variation of yield with temperature at constant length of heating.

<u>Condition of Quartz</u>	<u>Percent Activity Collected</u>
first sign of redness	6
dull redness	36
bright redness	56
dull white	61
melting point of gold	~100

On the other hand, when the gold was melted, the yield of astatine out of the liquid was always virtually quantitative, even if the burner was removed from the gold immediately after melting took place; thus this latter procedure was followed in all subsequent experiments.

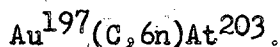
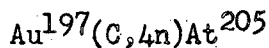
The apparatus used in these experiments was a modification of a stainless steel distiller which had been used² to separate astatine from bismuth targets. The bismuth had been melted by heating with a blast lamp, and the volatilized astatine collected on a platinum disk suspended in a cold "finger" about half an inch above the bismuth.

Stainless steel would not be suitable for use with molten gold, so a quartz bottom section was designed to clip on to the existing condenser. A diagram of this boiler is shown in Figure 1.

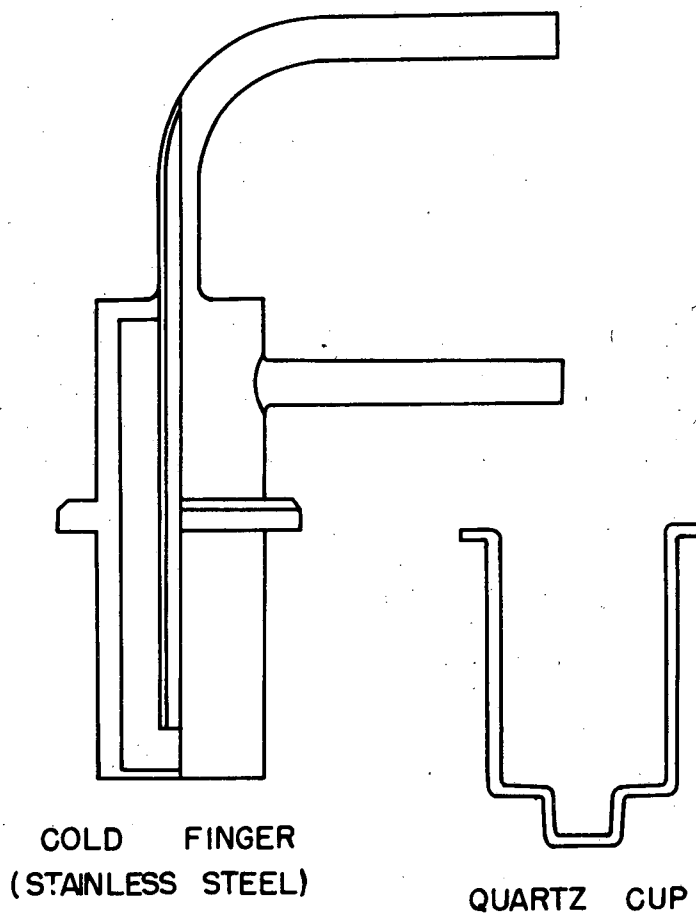
The resonance position of the carbon beam was estimated from the fact that

$$\left(\frac{e}{m}\right)_a - \left(\frac{e}{m}\right)_{C^{12(+6)}} = 0.07\%$$

so the first few bombardments were made with a magnetic field very close to that for the helium ion resonance. These resulted in ample alpha activity for identification; At²⁰⁵ and At²⁰³ were shown to be present by alpha pulse analysis, thus establishing that the following reactions had taken place:



No other alpha activities were observed in comparable yield. It should be noted, however, that At²⁰⁴ and At²⁰⁶ would probably have



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Fig. 1
Astatine Boiler

been missed in the above analysis because of their low alpha branching ratios, and At²⁰¹ or At²⁰² because of their short half-lives; there is no doubt that other reactions than the (C,4n) and (C,6n) occurred. The growth of a small amount of Po²⁰⁴ alpha activity into the sample was taken as evidence for the (C,5n) reaction, also.

The astatine fractions also were extremely beta radioactive; this high electron background made accurate alpha pulse analysis very difficult. One suspected immediately that the excess of Geiger counter detectable activity in the volatilized fraction was due to the presence of thallium isotopes produced from (α,xn) reactions on the gold. This was verified in the following way: The activity was leached off the platinum counting disk, and was shown to carry almost completely upon a precipitate of thalious iodide. The activity so carried showed two distinct periods of approximately 2 hours and 7 hours; it was therefore concluded that 1.8 hour Tl¹⁹⁸ and 7 hour Tl¹⁹⁹ had been produced in high yield by (α,3n) and (α,2n) reactions on the gold.

The Anomalous Carbon Ion Resonance; Cyclotron Parameters

It was noted in several bombardments that the amount of helium ion contamination in the carbon ion beam, radioassayed with gold as above, was not constant, but seemed to depend somewhat on the magnetic field strength. Some effect in this direction might have been anticipated from the fact that the e/m of C¹²⁽⁺⁶⁾ is 0.07 percent less than that of He⁴⁽⁺²⁾, but because the half-widths of the resonance peaks are so much broader than such a spacing, it should not have been possible to note so significant a resolution. This being the

case, the procedure in locating the internal carbon beam had been to adjust the magnet current regulator a certain prescribed amount lower than the position at which the external helium ion beam was located (the latter determination is a daily routine of the cyclotron operators). When this method failed to give reproducible results, several careful experiments were done in which the beam current was studied as a function of various adjustable cyclotron parameters, as:

- (a) Horizontal and vertical position of target probe
- (b) Radial distance of probe from ion source
- (c) Position of ion source
- (d) Oscillator power
- (e) Tank pressure
- (f) Magnet current.

The $Au^{197}(C,4n)At^{205}$ and $Au^{197}(C,6n)At^{203}$ reactions were again used to monitor the relative beam current. It was found that of the above parameters, only the variation of the magnet current proved critical, and this surprisingly so. The peak of the carbon ion resonance was observed at a magnet current position which was 1.1 percent lower than the helium ion resonance; this is fifteen times the expected shift as calculated from the ratios of the e/m . The experimental results are shown in Figure 2.

Perhaps the most interesting consequence of this experiment was the discovery of what was called the "burn-out resonance" - an intense beam of particles of very short range which actually melted the gold target foils. This "burn-out" beam has been very reproducible, always occurring at a magnet current ~ 0.2 percent lower than the carbon

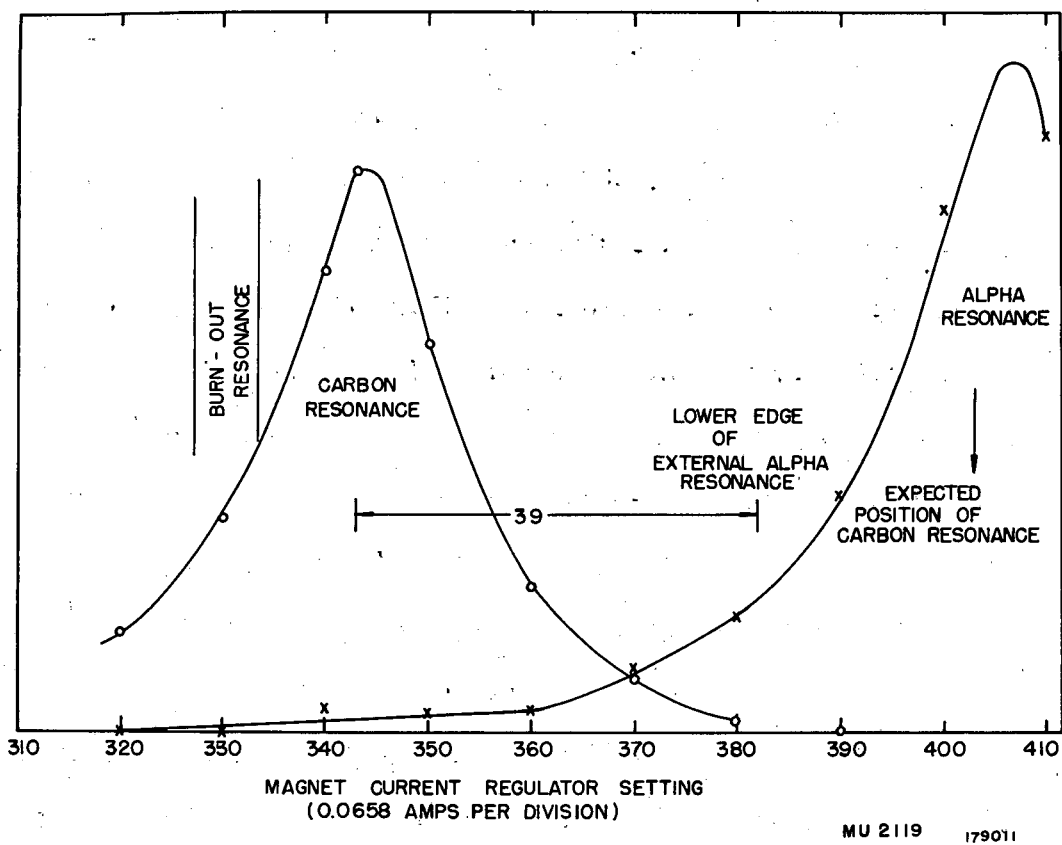


Fig. 2

Position of Carbon Resonance

resonance. The range of these particles corresponded roughly to carbon ions with $E \leq 30$ Mev, and was much too short to be attributed to a beam of deuterons, protons, or helium ions.

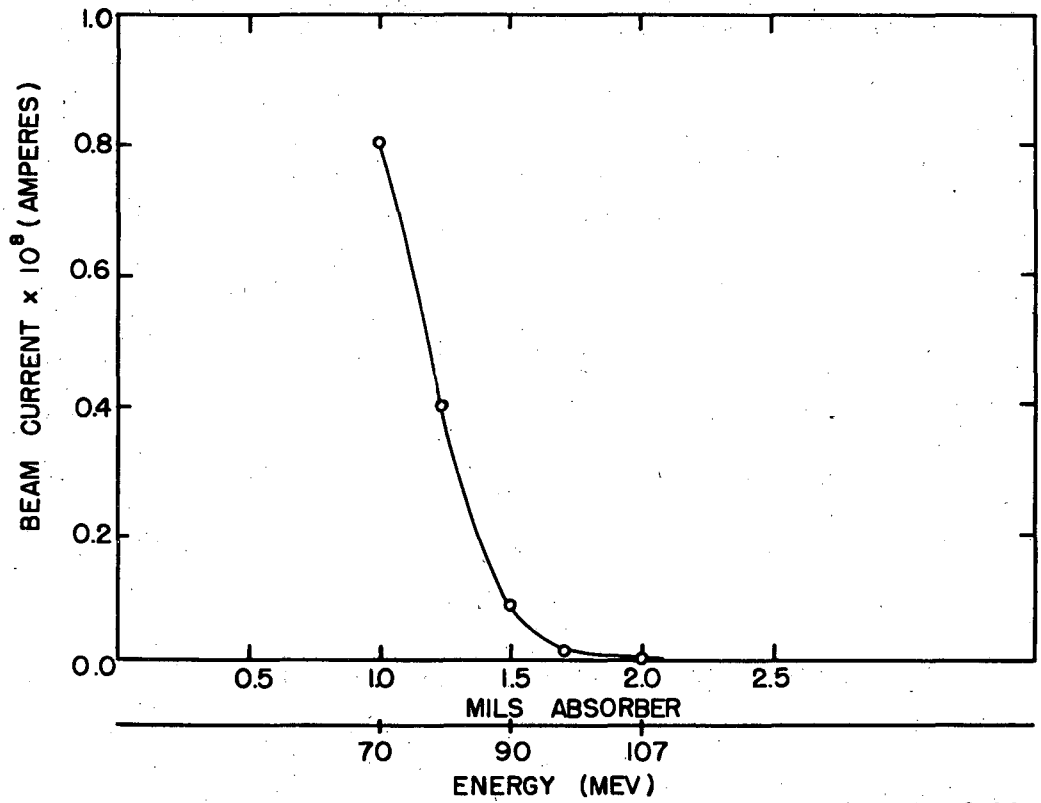
The latter point was further ruled out by the acceleration of $C^{13}(+6)$, using CO_2 enriched in that isotope and employing a higher (13/12) field strength to compensate for the lower e/m of C^{13} . These conditions are far off resonance for deuterons, protons, or helium ions, and yet the intense but useless beam was again found at the same place relative to the $C^{13}(+6)$ beam. Because of their low energy, these particles cannot penetrate the nuclear coulombic potential barrier, so no transmutations have been observed which could be correlated with them. The intensity of this beam has been verified visually by its intense heating effect on the target foils (which are not heated to incandescence by the $C(+6)$ beam) and by actual current measurements, indicating currents of the order of 1-100 microamperes. From these and other observations, it has been concluded that this beam is composed of $C(+2)$ ions of approximately 12 Mev energy which are accelerated in the 1/3 harmonic.

A discussion of the theories of beam formation will be given in a later section after the results of some more recent experiments have been described.

An attempt was made to evaluate a mean energy of the internal carbon (+6) ion beam at full radius by taking a rough absorption curve in tantalum. Although lack of thin foils at the time made it impossible to carry the curve into the lower energy region, the results were definite enough to establish that the most probable energy

is far below 114 Mev, as calculated from the cyclotron constants, and is in reality somewhere between 75 and 85 Mev. These data are shown in Figure 3. This unexpectedly low value was in contrast to the range measurement (in aluminum) of the external carbon ion beam by J. F. Miller, which indicated that the energy of the external beam is close to the calculated value of 114 Mev.

A sensitive current measuring probe, designed by A. Ghiorso, was used in conjunction with a vibrating reed electrometer to measure the beam current directly. An average value of the $C^{12}(+6)$ current over the period of most of these experiments was about 10^{-8} amperes. Recalling that the intensity of the external beam is usually approximately 10^{-12} amperes, one sees that the ratio of internal/external beam current is of the order of 10^4 , in contrast to a ratio of 10 to 20 found with standard bombarding particles. However, in view of the energy measurements this ratio is understandable, because the externally deflected beam will pass only ions of the Np corresponding to "full" energy, and these are seen from the range curve to constitute only a very small "tail" of the distribution curve, which is probably a very broad Gaussian.



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Fig. 3

Range of Internal Carbon Beam in Tantalum

II. EXCITATION FUNCTION EXPERIMENTS ON THE (C,4n) AND (C,6n) REACTIONS

An insight into the mechanisms of nuclear processes can often be gained by studying the energy dependence of reaction probabilities, and by correlating this dependence with predictions based on current nuclear models. Besides shedding light on the validity of the models, such a study also provides sound basis for confidence in planning future experiments in which these reactions may be utilized.

It is at once recognized that at the present stage of the heavy ion program, the uncertainty in experimental parameters will render a precise interpretation of excitation curves quite difficult. Nonetheless, such studies have been begun on the two reactions which have proved most accessible, the (C,4n) and (C,6n) reactions on gold.

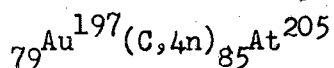
The stacked foil technique was used in making these bombardments, each foil having a thickness of 0.2 mil. Separation of the astatine activity from the bulk of the gold was done by the method described earlier. Identification of the At²⁰³ and At²⁰⁵ was made both by alpha pulse analyses and by direct half-life measurements with an alpha counter.

In order to facilitate the interpretation of these excitation experiments, a group of range-energy curves for carbon ions has been prepared by conversion of known data⁵ for other ions of the same e/m ratio, using the following approximate rule: A C¹²⁽⁺⁶⁾ has 1/N the range of particle X of 1/N the energy,

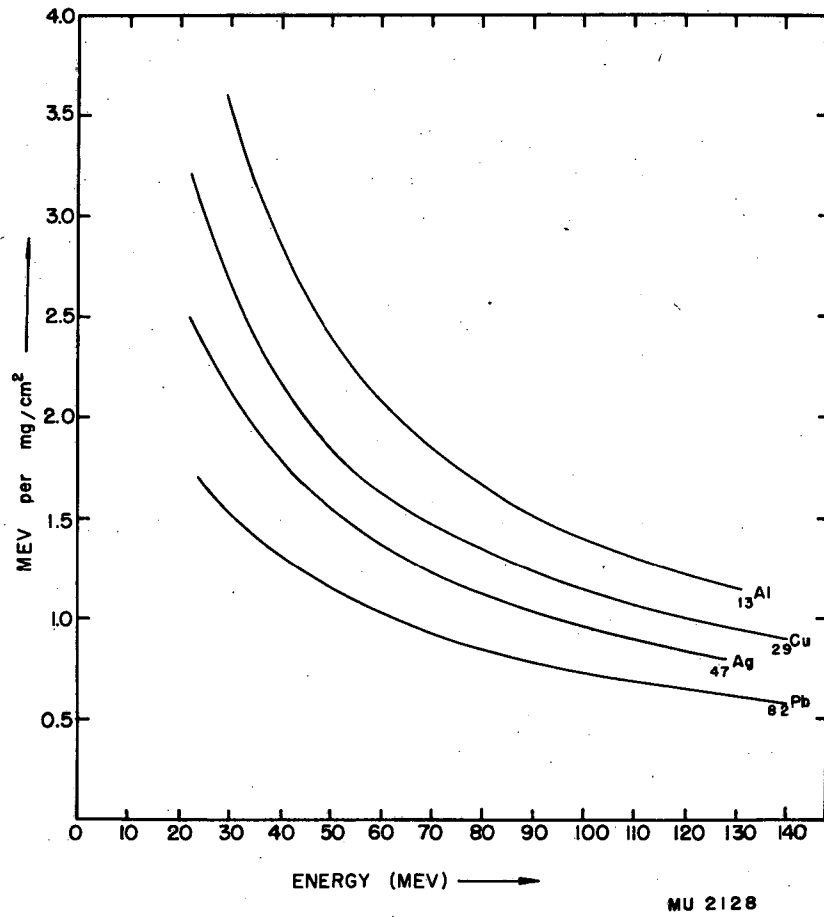
$$\text{where } N = \frac{\text{mass of C}^{12}}{\text{mass of X}} = \begin{cases} 3 & \text{for alpha particles} \\ 6 & \text{for deuterons} \end{cases}$$

This rule may also be stated as: A $C^{12}(+6)$ has N^2 the rate of energy loss of particle X of $1/N$ the energy. These curves are given in Figures 4, 5, and 6.

The results of two 5 minute bombardments are given in the following summary:



	Bombardment A	Bombardment B	Average
Activity in first foil	4.6×10^4 c/m	1.18×10^4 c/m	
Activity in first foil normalized	1	1	1
Activity in second foil normalized	0.913	0.483	0.698
Activity in third foil normalized	0.139	0.313	0.226
Activity in fourth foil normalized		0.040	0.040
Activity in fifth foil normalized		0.013	0.013

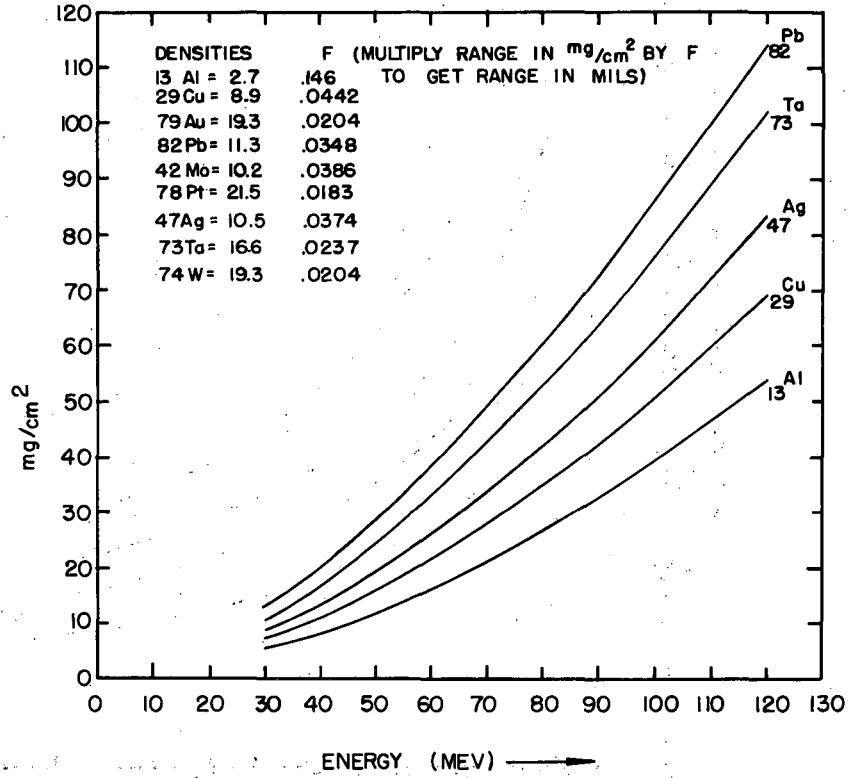


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Fig. 4

Rate of Energy Loss of Carbon C¹² (+6)
Ions in Various Absorbers



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Fig. 5
Range of Carbon C¹²⁽⁺⁶⁾ Ions
in Various absorbers

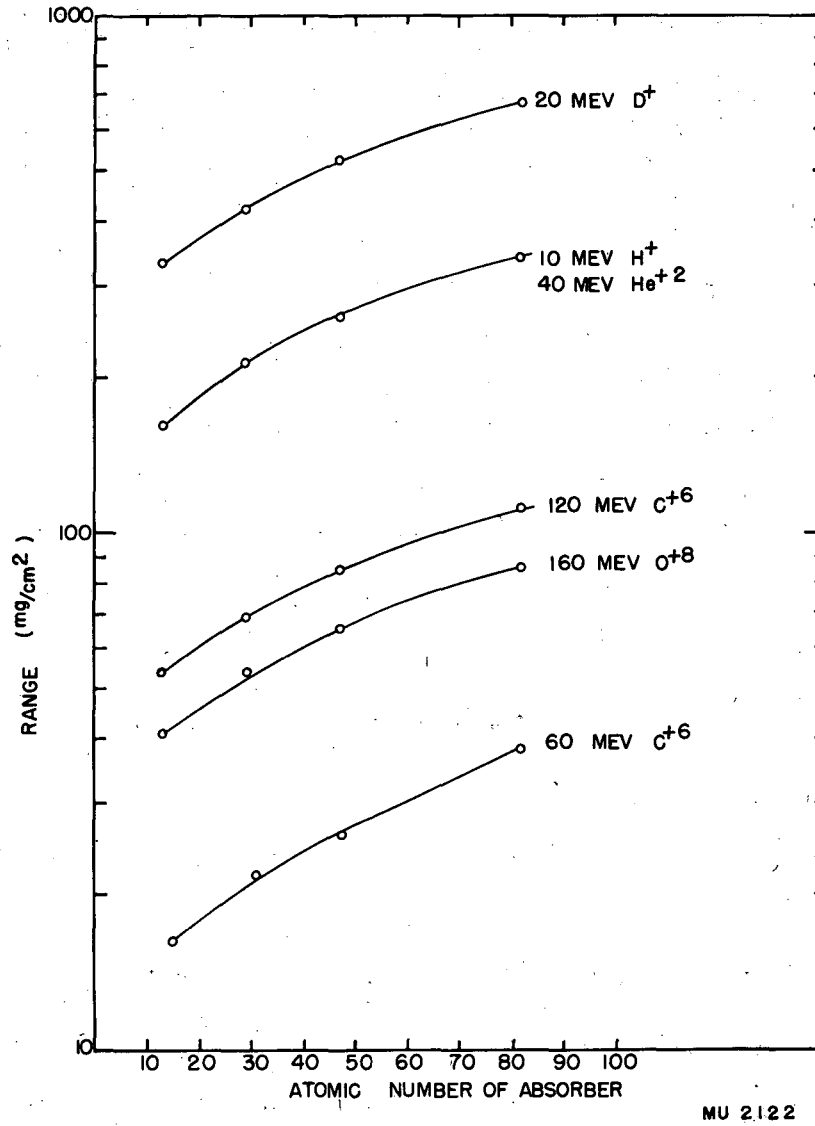
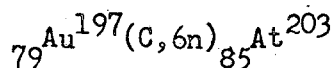


Fig. 6

Ranges of 60-inch Cyclotron
Particles in Various Absorbers



	Bombardment A	Bombardment B	Average
Activity in first foil	1.5×10^5 c/m	3.4×10^4 c/m	
Activity in first foil normalized	1	1	1
Activity in second foil normalized	0.246	0.165	0.205
Activity in third foil normalized	0.043	0.062	0.052

These data are shown graphically in Figure 7, from which the qualitative conclusion is drawn that at the highest energies, the yield of the (C,4n) reaction is approaching its maximum value, while the (C,6n) yield is still increasing.

In order to consider the excitation energies responsible for these reactions, an average energy of the beam must be specified. From Figure 3, a very approximate value of 80 Mev has been selected; because of the unusually wide energy distribution encountered with the carbon ion beam, the error introduced at this point is probably serious.

Computing the energy loss in each 0.2 mil foil, the data are tabulated as a function of energy.

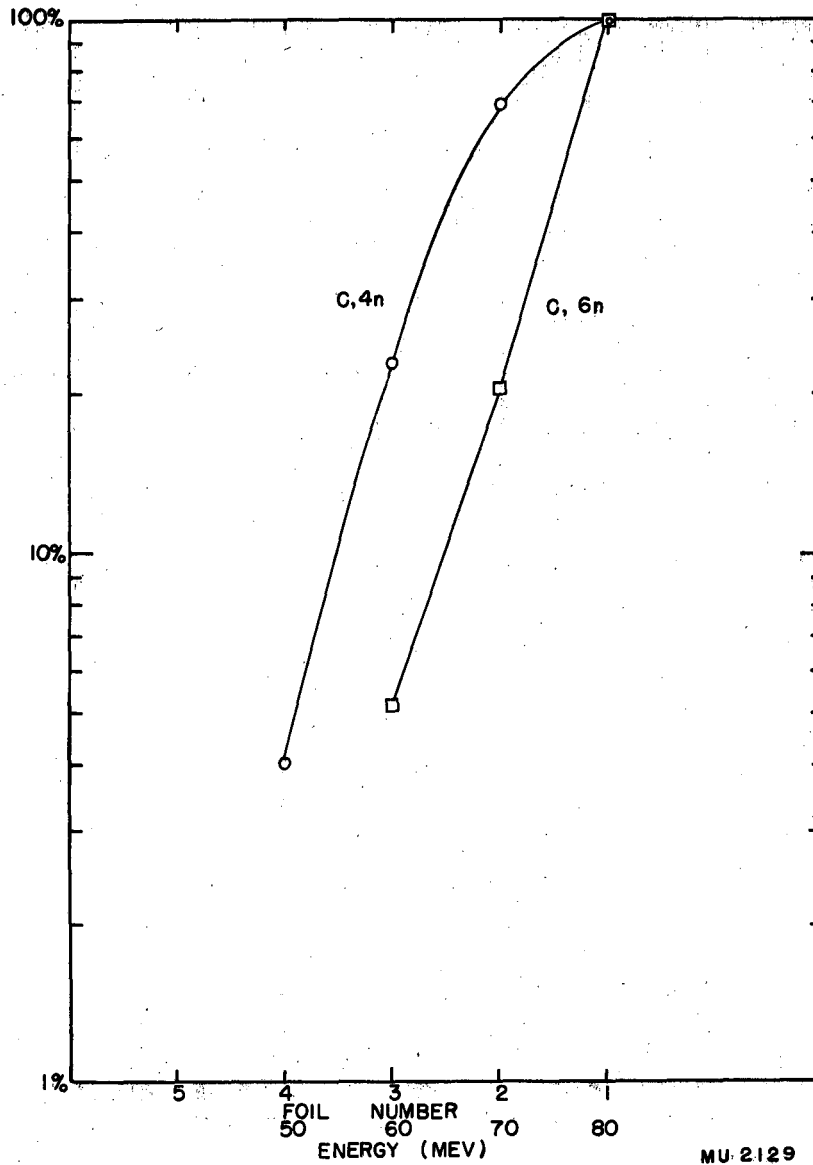


Fig. 7

Relative yields of (C,4n)
and (C,6n) Reactions on Gold

Foil Number	Average Carbon Energy	Yield (C,4n)	Yield (C,6n)
1	71-80	1	1
2	62-71	0.7	0.2
3	52-62	0.2	0.05

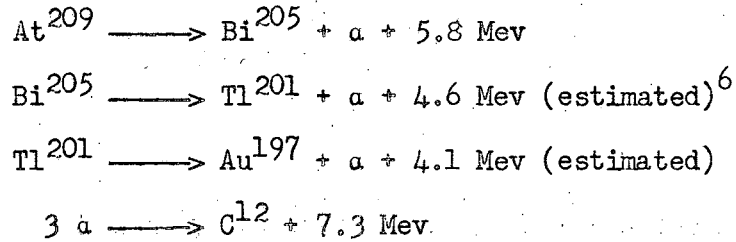
Carbon ions of energy less than 55-60 Mev should not contribute appreciably, since they do not have enough energy to overcome the potential barrier, which is given by:

$$\begin{aligned}
 V_0 &= \frac{Z_1 Z_2 e^2}{r} + \frac{A_1}{A_1 + A_2} E_1 \\
 &= \frac{0.96 Z_1 Z_2}{A_1^{1/3} + A_2^{1/3}} + \frac{A_1}{A_1 + A_2} E_1 \\
 &= 58 + 0.057 E_1 \text{ (Mev)}
 \end{aligned}$$

The fact that appreciable yields of the (C,4n) reaction are found in the fourth and fifth foils cannot be explained on the basis of barrier penetration, which will be shown to be quite unimportant. Reactions in these foils must have been caused by the smaller number of particles which constitutes the high energy "tail" of the energy distribution. An ion of 110 Mev, for example, will still have approximately 70 Mev when entering the fifth foil.

Consider the excitation energy given by a carbon ion of 80 Mev to a gold nucleus. First, the available kinetic energy in the center

of mass system will be $197/209 \times 80 \approx 76$ Mev. The binding energy of C^{12} to Au^{197} is approximately -22 Mev, as can be seen from the following scheme:



Therefore, the excitation energy will be $\approx (76-22) \approx 54$ Mev.

One can use the statistical treatment of Weisskopf⁷ to estimate the most probable number of neutrons emitted from a nucleus of At^{209} excited to 54 Mev.

Considering the nucleus as a degenerate Fermi gas of neutrons and protons, the total energy of excitation (measured above the $T = 0$ level) is given by:

$$E = a(kT)^2 + b(kT)^4 + \dots$$

The second term is generally neglected. The constant, a, can be evaluated from experimental data on nuclear level densities, and is given, for $A > 60$:

$$a = 0.84(A-40)^{1/2}$$

$$\text{or } kT = \sqrt{\frac{E}{0.84(A-40)^{1/2}}}$$

For At²⁰⁹, at an excitation of 54 Mev,

$$kT = \sqrt{\frac{54}{0.84(169)^{1/2}}}$$

$$kT = 2.2 \text{ Mev .}$$

The treatment further develops that the neutrons are emitted with greatest probability around an energy kT . So we may now estimate the most probable number of neutrons emitted by the compound nucleus At²⁰⁹, assuming an average binding energy for the last neutron as 8 Mev.

Nucleus	E _{excitation} (Mev)	KE _{neutron} (Mev)
At ²⁰⁹	54	2.2
At ²⁰⁸	43.8	2.0
At ²⁰⁷	33.8	1.7
At ²⁰⁶	22.1	1.4
At ²⁰⁵	10.7	1.0
At ²⁰⁴	1.7	--

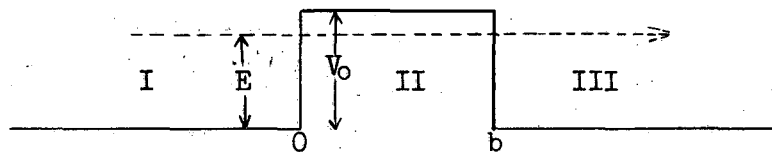
This approximate analysis shows, at least, that reactions of the order of (C,5n) should be exhibiting their peak yields under the conditions of the gold bombardments. Such an interpretation would predict that the (C,4n) curve should be bending over at this point, while that for the (C,6n) should still be rising. This is the situation which is observed experimentally.

Some "average" energy has been assumed for the above considerations, perhaps the only justification being simplicity of calculation. But one must add to this the complication resulting from the broad energy spread of the carbon beam. Since the (C,6n) reaction requires more excitation than the (C,4n), it is reasonable to assume that in the second and third foils, an increasing fraction of the (C,6n) processes would have to be initiated by high energy carbons from the tail of the distribution curve. This in itself would serve to decrease the ratio of (C,6n)/(C,4n) in the inner foils.

Leakage of Carbon Ions Through Potential Barrier, where $E < V_0$

It can be shown that the quantum mechanical phenomenon of barrier tunneling is far less important in the case of carbon ions than with other cyclotron accelerated particles; the heavier a body becomes, the closer it approaches classical behavior.

Consider first a one dimensional rectangular barrier.⁸



One represents the beam of incident particles as a plane wave, traveling to the right, so that:

$$\psi_{\text{inc.}} = ae^{ikx}$$

$$\text{where } k = \frac{p}{\hbar} = \sqrt{\frac{2\mu E}{\hbar^2}}$$

and the reflected wave is:

$$\psi_{\text{ref.}} = \beta e^{-ikx}$$

The transmitted wave is:

$$\psi_{\text{III}} = \gamma e^{ikx}$$

Inside the barrier, the wave function becomes

$$\psi_{\text{II}} = Ke^{\sqrt{\frac{2\mu}{\hbar^2} (V_0 - E) x}} + Le^{-\sqrt{\frac{2\mu}{\hbar^2} (V_0 - E) x}}$$

The transparency of the barrier is defined as

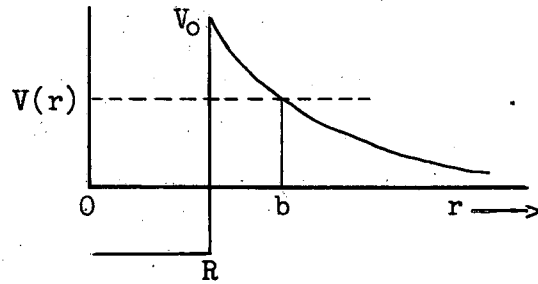
$$T = \frac{\text{transmitted intensity}}{\text{incident intensity}} = \frac{|\psi_{\text{III}}|^2}{|\psi_{\text{inc.}}|^2} = \frac{|\gamma|^2}{|a|^2}$$

The constants are determined by making use of the usual continuity conditions across the two boundaries. The transparency is found to be:

$$T = e^{-2\sqrt{\frac{2\mu}{\hbar^2} (V_0 - E) b}}$$

The order of magnitude of the transparency of a barrier of another shape can be obtained by finding the average height and treating as a rectangular barrier.

Then, for a Coulomb barrier:



$$T = e^{-2} \int_R^b \sqrt{\frac{2\mu}{\hbar^2} (V_0 - E)} dr = e^{-G}$$

$$\text{Now, } V_0 = \frac{Z_1 Z_2 e^2}{r}$$

$$\therefore G = 2 \sqrt{\frac{2\mu}{\hbar^2}} \int_R^b \sqrt{\frac{Z_1 Z_2 e^2}{r} - E} dr$$

$$= \sqrt{\frac{8\mu Z_1 Z_2 e^2}{\hbar^2}} \int_R^b \sqrt{\frac{1}{r} - \frac{E}{Z_1 Z_2 e^2}} dr$$

The integration gives:

$$G = \sqrt{\frac{8\mu Z_1 Z_2 e^2}{\hbar^2}} \left[\cos^{-1} \sqrt{\frac{R}{b}} - \sqrt{\frac{R}{b} - \frac{R^2}{b^2}} \right]$$

The penetration curve for carbon ions on gold has been calculated, using:

$$R = 1.5 \times 10^{-13} \left[197^{1/3} + 12^{1/3} \right]$$
$$= 1.2 \times 10^{-12} \text{ cm}$$

$$\text{at } r = b, E = V_0$$

$$b = \frac{Z_1 Z_2 e^2}{E}$$

Figure 8 illustrates the extent of leakage for carbon ions on gold. For comparison, a similar curve has been included showing proton leakage through the potential barrier of uranium, which is quite appreciable.

The above calculation, when considered in the light of a three dimensional barrier, refers only to those particles which approach the target head-on, (S-wave collision). When there is relative angular momentum between the two nuclei, the effective barrier is increased by the "centrifugal potential" term; the result of this additional force is to decrease the barrier penetration even further for those particles which hit off center. A further result is that even at $E = V_0$, only those particles which approach head-on will penetrate to the range of nuclear forces; others are scattered off because of the higher barrier. Classically, this is equivalent to a Rutherford (coulomb) scattering process, which will keep the effective cross section of the target less than πR^2 , approaching this value at the limit of very high energies. Such an argument is in agreement with the sharp rise of experimental yield curves

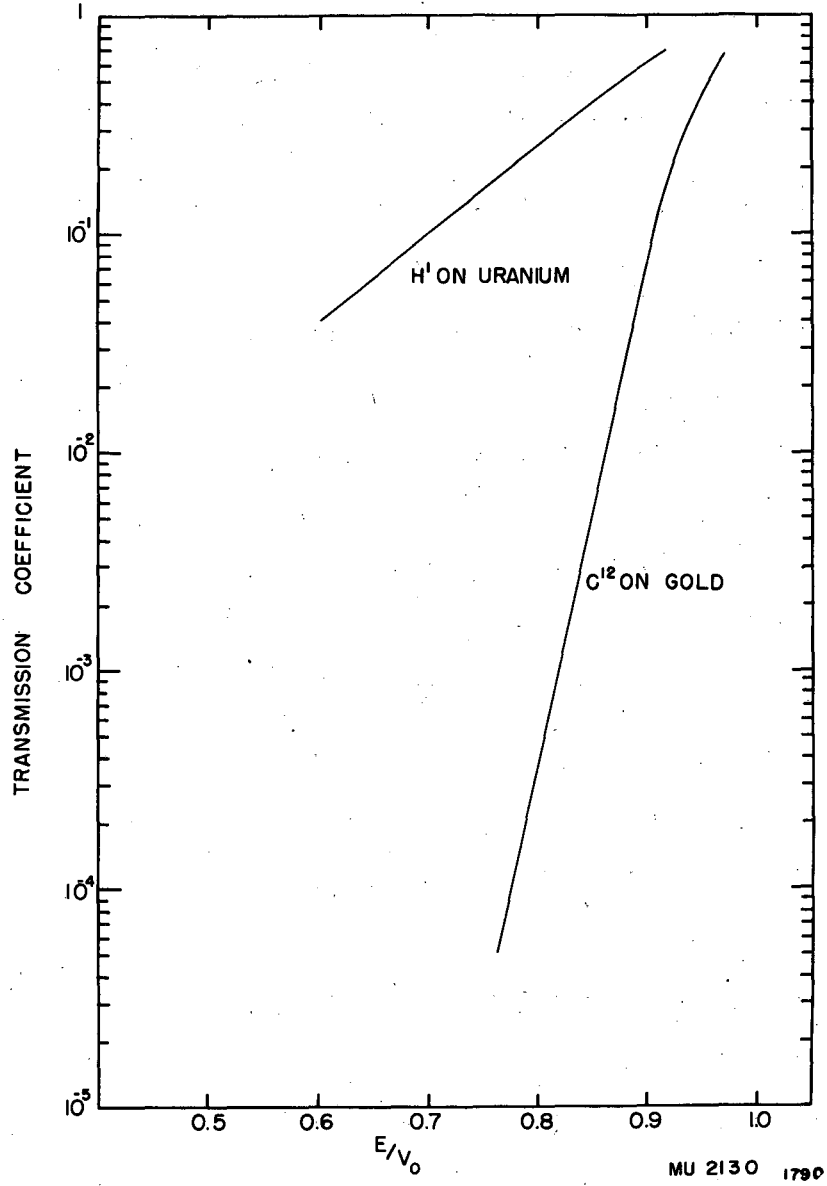


Fig. 8

Leakage of Carbon Ions Through Potential Barrier

for reactions which are limited by a barrier higher than the energetic threshold, such as the (C,4n) and (C,6n) reactions. Coulomb scattering will be discussed further in a future section.

Estimate of the (C,4n) and (C,6n) Cross Sections

The following estimate is made on the basis of the amount of astatine produced in the first gold foil during two average 5 minute bombardments. It is admittedly rough, but as good as can be done at the present time.

The experimental data are summarized below:

Yield (α c/m) of At ²⁰⁵ T _{1/2} = 26 min	Yield (α c/m) of At ²⁰³ T _{1/2} = 7 min	Average Ion Current (amperes)	Thickness of Foil (atoms/cm ²)	Length of Bombardment (minutes)
4.5 x 10 ⁴	1.5 x 10 ⁵	10 ⁻⁸	4 x 10 ¹⁹	5
1.2 x 10 ⁴	3.3 x 10 ⁴	10 ⁻⁸	4 x 10 ¹⁹	75

(C,4n) Cross Section.-- The average yield of At²⁰⁵, correcting for geometry of the alpha counter, was approximately 5 x 10⁴ alpha disintegrations per minute. At²⁰⁵ is a nuclide which decays predominantly by electron capture, but the exact branching ratio has not yet been determined, so it is difficult to convert alpha disintegrations into total decay rate. An extrapolation may be made, however, on the basis of the known systematics of alpha decay.⁶ These data would predict an alpha half-life of the order of three hours for At²⁰⁵,

which would establish its alpha branching ratio as $26/180 \approx 15$ percent.

Using this figure, we may calculate a cross section from the relation:

$$N(t) = I n \sigma t$$

where N = total number of atoms produced (neglecting decay during bombardment) = $\frac{dn}{dt/\lambda}$

I = flux = 10^{-8} amperes = 10^{10} carbon ions/second

n = number of target nuclei = 4.5×10^{19} nuclei/cm²

σ = cross section

t = length of bombardment = 300 seconds

$$\text{therefore } \sigma = \frac{5 \times 10^4}{0.15} \cdot \frac{26}{0.693} \cdot \frac{1}{10^{10}} \cdot \frac{1}{4 \times 10^{19}} \cdot \frac{1}{300}$$

$$\approx 10^{-25} \text{ cm}^2$$

$$\approx 0.1 \text{ barn}$$

(C,6n) Cross Section.--- An average of 9×10^4 alpha disintegrations per minute of At²⁰³ was produced in these irradiations. Similarly, estimating an alpha half-life for At²⁰³ to be roughly 30 minutes, the branching ratio becomes $\approx 7/30 \approx 20$ percent. Because of the short half-life of At²⁰³, we use the following relation:

$$N(t) = \frac{In\sigma}{\lambda} (1 - e^{-\lambda t})$$

In this case:

$$(1 - e^{-\lambda t}) \approx 0.5$$

$$\sigma = \frac{2 \lambda N(t)}{I_n}$$

$$= \frac{2 \frac{dN}{dt}(t)}{I_n}$$

$$\therefore \sigma \approx 2 \cdot \frac{9 \times 10^4}{0.2} \cdot \frac{1}{10^{10} \times 60} \cdot \frac{1}{4 \times 10^{19}}$$

$$\approx 4 \times 10^{-26} \text{ cm}^2$$

$$\approx 0.04 \text{ barn.}$$

The absolute magnitude of these figures is, of course, no more precise than the parameters involved in computing them; the actual beam current may have been different from the figure given by a factor of three or four, so these cross sections must be accepted accordingly.

The relative magnitude of the cross sections should be more reliable, for the validity of this ratio depends mainly upon the predictions of alpha decay systematics, which in the past have proved surprisingly accurate. One notices that this ratio is in agreement with that which would have been predicted from the shape of the excitation curves, since the (C,4n) reaction seems to be near its maximum value, while the slope of the (C,6n) yield curve is still quite steep at this point.

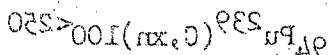
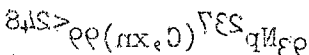
Because of the wide spread in carbon ion energies, it would be more precise to express the observed cross sections as:

A great stimulus toward the study of carbon ion reactions has

been the hope that the synthesis of transactinoid elements will

$$\sigma(\text{observed}) = \int_{E_{\text{min}}}^{E_{\text{max}}} I(E) \sigma(E) dE$$

be made possible by such reactions as:



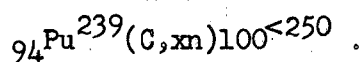
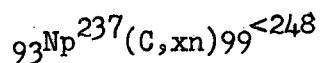
Accordingly, a knowledge of the (C, xn) cross sections in the region of uranium would be of great help in the design of future experiments with heavy element production in mind. Of comparable interest is the fission cross section of uranium for carbon ions, since the competition of the fission process with (C, xn) reactions in the region $E = 9 \text{ MeV}$ is likely to be serious, or even prohibitive. Also of interest in its own right is the shape of the fission yield curves; from this datum some clue to the mechanism of the

fission process may be found.

Several bombardments of uranium with carbon ions have been made, in which the production of U^{236} and U^{238} has been verified, representing the (C, xn) and (C, dn) reactions. From an aliquot of the original target solution of one of these bombardments, an analysis was made for radioactivities of silver, strontium, and barium. The chemical separations proceeded as follows:

III. THE FISSION OF URANIUM WITH CARBON IONS

A great stimulus toward the study of carbon ion reactions has been the hope that the synthesis of transcalifornium elements will be made possible by such reactions as:



Accordingly, a knowledge of the (C,xn) cross sections in the region of uranium would be of great help in the design of future experiments with heavy element production in mind. Of comparable interest is the fission cross section of uranium for carbon ions, since the competition of the fission process with (C,xn) reactions in the region $Z = 94$ is likely to be serious, or even prohibitive.

Also of interest in its own right is the shape of the fission yield curve; from this datum some clue to the mechanism of the fission process may be found.

Several bombardments of uranium with carbon ions have been made, in which the production of Cf^{246} and Cf^{244} has been verified, representing the (C,4n) and (C,6n) reactions.⁹ From an aliquot of the original target solution of one of these bombardments, an analysis was made for radioactivities of silver, strontium, and barium. The chemical separations proceeded as follows:

A. (1) To the target solution, 10 mg of standardized silver carrier, and 20 mg of standardized barium and strontium carriers (as nitrates) were added.

(2) Fifteen ml fuming nitric acid was added, and the solution cooled for several minutes while stirring.

(3) Precipitate was centrifuged, and supernatant (Ag^+) set aside.

B. Precipitates of $\text{Ba}(\text{NO}_3)_2$ and $\text{Sr}(\text{NO}_3)_2$ from A.

(1) Precipitates were dissolved in 2 ml H_2O .

(2) $\text{Ba}(\text{NO}_3)_2$ and $\text{Sr}(\text{NO}_3)_2$ reprecipitated with 15 ml fuming HNO_3 , centrifuged.

(3) Precipitates again dissolved in 5-10 ml H_2O .

(4) Five mg Fe^{+++} carrier was added, and $\text{Fe}(\text{OH})_3$ precipitated by addition of 2 ml NH_4OH . Precipitate was centrifuged and discarded.

C. Supernatant solution from B.

(1) Neutralized with 6 N HNO_3 .

(2) One ml 6 M HAc and 2 ml 6 M NH_4Ac were added, and the solution heated nearly to boiling.

(3) One ml 1.5 M Na_2CrO_4 added dropwise with stirring, and solution allowed to stand for several minutes.

(4) Precipitate was centrifuged, and supernate (Sr^{++}) set aside.

D. BaCrO_4 precipitate from C.

(1) Precipitate was washed with 10 ml hot H_2O , then dissolved in 1-2 ml 6 M HCl .

(2) Fifteen ml HCl - ether reagent was added, and the mixture stirred for several minutes.

(3) Precipitate of BaCl_2 centrifuged, and the supernate discarded.

(4) BaCl_2 was dissolved in 1 ml H_2O , and reprecipitated as in (2).

(5) The precipitate was transferred to a weighed aluminum disk with 4% HCl in alcohol.

(6) Precipitate was washed thrice with ether, and dried in a vacuum desiccator.

(7) Precipitate was weighed as $\text{BaCl}_2 \cdot \text{H}_2\text{O}$.

E. Supernate (Sr^{++}) from C.

(1) Two ml concentrated NH_4OH added, and solution heated nearly to boiling.

(2) Five ml saturated $(\text{NH}_4)_2\text{C}_2\text{O}_4$ added slowly with stirring.

(3) Mixture stirred for several minutes, then centrifuged.

(4) Precipitate was washed thrice with 5 ml dilute hot NH_4OH , likewise with 5 ml 95% alcohol, and 5 ml ether.

(5) Precipitate transferred to weighed aluminum dish, and dried in a vacuum desiccator.

(6) Precipitate was weighed as $\text{SrC}_2\text{O}_4 \cdot \text{H}_2\text{O}$.

F. Supernate (Ag^+) from A.

(1) Solution was boiled, then diluted to 5-6 N HNO_3 .

(2) AgCl was precipitated by the addition of 1 ml 0.5 N HCl .

The precipitate was coagulated by boiling with a microburner.

(3) Supernate was discarded.

(4) The AgCl was washed twice with 10 ml 1 N HNO_3 containing a drop of 2 N HCl .

- (5) AgCl was dissolved in 2 ml 6 N NH_4OH , and the solution diluted to 10 ml.
- (6) Approximately 2 mg Fe^{+++} was added, and the $\text{Fe}(\text{OH})_3$ precipitate was centrifuged and discarded.
- (7) The $\text{Fe}(\text{OH})_3$ scavenge was repeated as in (6).
- (8) Supernate from (7) was saturated with H_2S in the cold.
- (9) Precipitate of Ag_2S was washed, then dissolved in 2 ml concentrated HNO_3 .
- (10) Solution boiled, then diluted to approximately 4 N.
- (11) AgCl precipitated by the addition of 1 ml 0.5 N HCl , and washed as in (4).
- (12) AgCl dissolved as in (5) and solution diluted to 10 ml.
- (13) Scavenge twice with Fe^{+++} as in (6) and (7).
- (14) Ag_2S precipitated again as in (8).
- (15) Precipitate dissolved in 1 ml concentrated HNO_3 , and the solution boiled.
- (16) Solution diluted to 6 ml.
- (17) One mg Fe^{+++} added, and solution made basic with NH_4OH , and the $\text{Fe}(\text{OH})_3$ discarded.
- (18) Solution acidified to approximately 2 N with HNO_3 , and 2 drops 6 N HCl added.
- (19) Mixture was boiled to coagulate the AgCl.
- (20) AgCl centrifuged, washed thrice with 5 ml 0.5 N HNO_3 , same with 5 ml alcohol, transferred to weighed aluminum disk.
- (21) Precipitate was dried for 10 minutes at 110°C , and weighed as AgCl.

The following activities were found:

Fraction	Nuclide	Half-life	Yield (dis/min)	Yield (atoms)
Silver	Ag ¹¹² + Ag ¹¹³	~4 hours	5 x 10 ⁴	~10 ⁷
	Ag ¹¹¹	7 days	160	2.2 x 10 ⁶
Strontium	Sr ⁹¹	9.7 hours	900	7.6 x 10 ⁵
	Sr ^{89?}	long	<20	--
Barium	Ba ¹⁴⁰	12.8 days	42	1.1 x 10 ⁶

The approximately 4 hour decay period observed in the silver fraction undoubtedly represents an unresolved mixture of 3.2 hour Ag¹¹² and 5.3 hour Ag¹¹³, so we shall assume that approximately equal amounts of each were formed. Figure 9 shows the observed yields.

The data accumulated in this experiment are neither numerous nor reliable enough to allow one to draw any definite conclusions as to the shape of the fission product distribution. However, it is of interest to consider how the various possibilities would correlate with existing pictures of the fission process.

On the basis of the liquid drop model of Bohr and Wheeler, and Frenkel, it has been possible to estimate the activation energies necessary to reach the "critical shape" for fission.¹⁰ This activation energy is given in terms of the function Z^2/A . From this "fissionability parameter" it is found that for nuclides such as U²³⁵ and Pu²³⁹, where $Z^2/A \approx 36$, the activation energy is less than the binding energy of a neutron; thus these nuclei will fission with

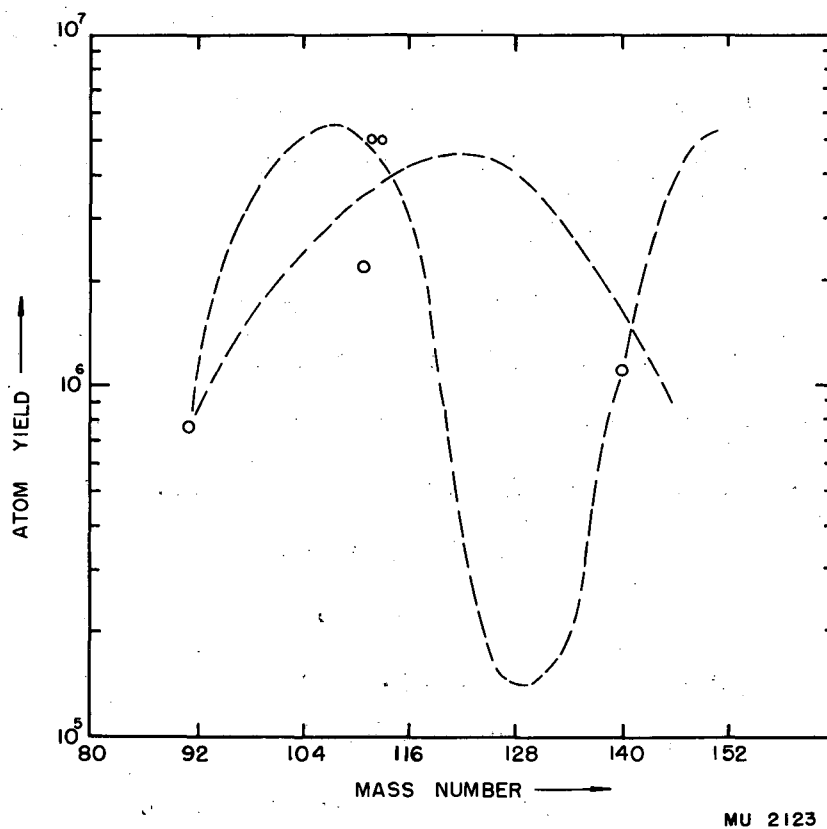


Fig. 9

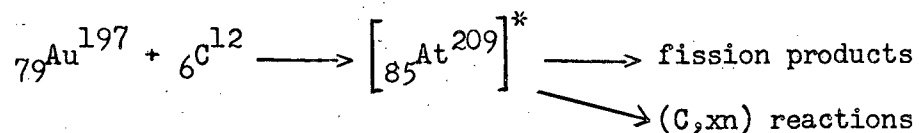
Fission Yield from Uranium Bombarded with C^{12}

slow neutrons. Associated with slow neutron fission, but as yet unjustified theoretically, is the familiar double-humped fission product distribution curve with its valley centered at about half the mass of the parent nucleus.

Nuclei whose activation barriers are higher than the binding energy of a neutron can still be caused to fission, if the requisite energy is supplied as kinetic energy of the bombarding particle. However, the process of fission will now have to compete with emission of heavy particles, as well as with gamma emission. In the case of such "fast" fission, distribution curves have been observed from the extreme of a single humped (symmetrical) curve in the case of bismuth fission¹¹ to a shallow double-humped curve for uranium.¹²

In their study of the fission of bismuth with high energy deuterons, Goeckermann and Perlman¹¹ point out that the fission seems to occur most probably from the parent nucleus Po^{199} or Po^{200} , only after about 10 neutrons have evaporated from the original compound nucleus. At this point $Z^2/A \approx 36$, and the activation energy is approximately equal to 6 Mev. Thus, it seems that the activation energy necessary for fission must be less than or equal to the neutron binding energy before fission will compete with neutron emission.

If this reasoning may be extended to another case, one would predict that the fission of gold with 80 Mev carbon ions will occupy a very small portion of the total cross section; for in the reaction

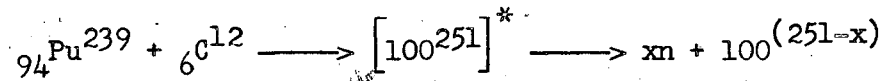


at least seven neutrons would have to be boiled off before the point is reached where fission would compete with neutron emission; and we saw above that at 80 Mev, a (C,8n) reaction should not be too likely.

Perhaps entropy considerations may help one understand why fission does not compete with neutron emission until the barrier energy is of the order of the neutron binding energy. If the excitation energy for fission is higher, then the fission fragments must take off a greater amount of kinetic energy than a neutron, which will on the average escape with only kT of kinetic energy. Thus, the residual nucleus from fission will be left in a state of lower energy; that is, a state of lower level density and lower entropy. Here, neutron emission will predominate. But, when the fission barrier is of the order of the neutron binding energy, then the entropy effects are more or less the same for both processes, and fission competes with neutron emission. This occurs when $Z^2/A \longrightarrow 36$.

In the bombardment of uranium with carbon ions, the compound nucleus is ${}_{98}\text{Cf}^{250}$, for which $Z^2/A \approx 38$. The fission activation energy for nuclei of this Z^2/A is given by Frankel and Metropolis¹⁰ as ≈ 3 Mev, considerably less than the neutron binding energy. One is then led to the expectation that in this instance, fission will be the predominant mode of energy dissipation. Neutron emission will be unlikely from the compound nucleus; of course, the fission fragments will be formed in a state of high excitation, perhaps high enough to evaporate quite a few nucleons. This predominance of fission may make it difficult to prepare sufficient quantities of transcalifornium elements by carbon ion bombardments, because

of the neutron deficiency of the product nuclei, which leads to high values of Z^2/A . For instance, in the reaction



the value of Z^2/A for the compound nucleus is 39.8, indicating a fission barrier of only 2 Mev.

Even though the hypothetical fission of Cf^{249} with slow neutrons would undoubtedly show a double humped fission product distribution curve, it would be much more likely in the case of the splitting up of Cf^{250} formed with carbon ions on uranium to find a symmetric distribution, or at most only a small valley; the high potential barrier (~60 Mev) prevents the formation of the compound nucleus at low excitation, so presumably those nuclides undergoing fission at high excitation will follow the mechanism usually postulated for fast fission, resulting in a symmetric distribution, and an unchanged Z/A ratio in the fission fragments.

A thorough study of the characteristics of carbon ion induced fission would be very desirable, as soon as the experimental limitations are reduced sufficiently so that quantitative work becomes feasible.

Calculation of Fission Cross Section

An estimate of the fission cross section of uranium for carbon ions ($60 < E < 90$ Mev) may be estimated. Making the rough guess of a 3 percent fission yield for the nuclide Ag^{112} , one has:

$$\sigma = \frac{N}{Int}$$

where $N \approx 3 \times 10^8$ atoms

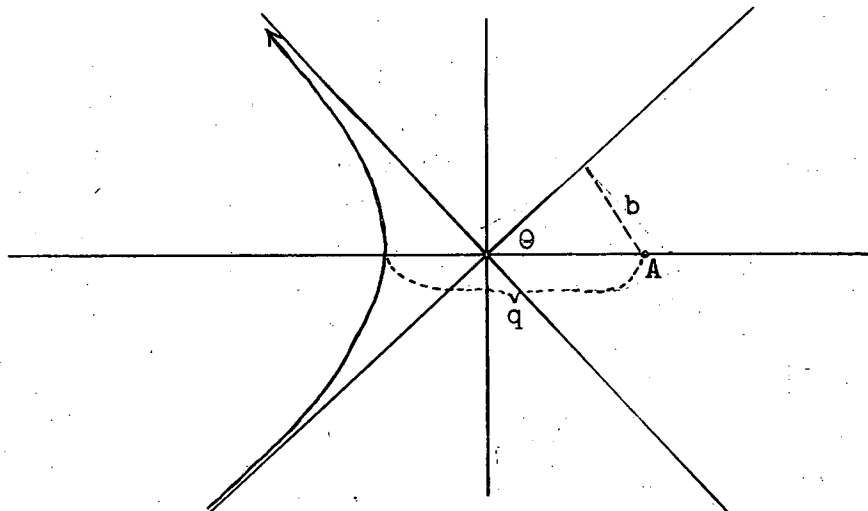
$I \approx 10^{-8}$ amperes $\approx 10^{10}$ ions/second

$n = 6 \times 10^{19}$ atoms/cm²

$t = 2$ hours = 7200 seconds

$\therefore \sigma_f \approx 0.1$ barn .

This figure seems to be rather low, in view of the conclusion reached above that fission should be the primary reaction. However, one must take note of the fact that coulomb scattering will play a significant role in reducing the effective total cross section, when the interaction is between a heavy element and so highly charged a projectile as a carbon nucleus. This can be shown by the following approximate calculation based on classical, Rutherford scattering formulae.



A = target nucleus

q = distance of closest approach

b = impact parameter

The Rutherford scattering formulae are:

$$\begin{aligned} Q &= b \cot \theta/2 \\ b &= K \tan \theta \quad \text{where } K = \frac{Z_1 Z_2 e^2}{2E} \\ &= \frac{R}{2E} V_0 \end{aligned}$$

In order for reaction to occur, the particles must approach each other within the range of their forces. We will approximate this condition by setting $q = R_f = R_u + r_c$, the sum of the nuclear radii of target and projectile. Eliminating θ from these two equations, one has:

$$\begin{aligned} b^2 &= q^2 - 2Kq \\ &= R_f^2 - 2KR_f \\ &= R_f^2 \left(1 - \frac{2K}{R_f} \right) \\ &= R_f^2 \left(1 - \frac{V_0}{E} \right) \quad \text{where } V_0 < E \end{aligned}$$

The effective cross section will be πb^2 .

$$\begin{aligned} \text{Thus } \sigma_{\text{eff}} &= \pi R_f^2 \left(1 - \frac{V_0}{E} \right) \\ &= \sigma_{\text{geom}} \left(1 - \frac{V_0}{E} \right) \end{aligned}$$

For an average energy of ~80 Mev

$$\sigma_{\text{eff}} = \frac{\sigma_{\text{geom}}}{4}$$

≈ 1 barn

This figure is still high. However, if the potential barrier, V_0 , should be higher than that estimated, the energy of the beam might fall below V_0 in the latter part of the foil. This would reduce n (effective) below 6×10^{19} atoms/cm², and would further raise the calculated value for the observed cross section.

It is interesting to note that the yield of Cf^{244} obtained by Thompson, Street, and Ghiorso in this bombardment corresponds to a cross section for the $(\text{C}, 6n)$ reaction which is smaller than the fission cross section by at least a factor of 10^4 . This ratio is in agreement with the general argument presented in this section.

IV. PRODUCTION OF BROMINE ISOTOPES FROM COPPER

One of the more practical applications of high energy carbon ion beams is their utilization in the production of neutron deficient radionuclides. In the center portion of the periodic system, one must add an average of 1.7 neutrons for every proton to follow the line of stability; whereas in a typical carbon ion reaction (e.g., $C,4n$) only 0.3 neutron is added per proton. In short, this is one of the best methods of adding protons to a nucleus.

The study of bromine isotopes produced from bombardment of copper was advantageous, since the chemistry involved is simple and the target material is readily available in high purity and in foils of any desired thickness. Bombardments of thin copper foils with the carbon beam have resulted in the production of two light isotopes of bromine, one of which had previously been reported.¹³

A chart of the isotope section under consideration is shown in Figure 10.

The chemical procedure which has been used to isolate the bromine fraction is as follows:

1. The copper foil was placed into a distilling flask, and 10 mg each of arsenic, selenium, and bromine carriers added.
2. Several ml concentrated HCl was added, and the flask closed.
3. 30% H_2O_2 was added dropwise to the flask (via a built-in funnel stopcock arrangement) while heating.
4. The Br_2 was swept over into a $CCl_4 - H_2O$ trap by a slow air stream.

^{35}Br	Br^{72}	Br^{73}	Br^{74} 35 min. β^+ , K	Br^{75} 1.5 hours β^+ 1.8Mev K	Br^{76} 16.5 hours β^+ 3.5Mev γ	Br^{77} 57 hours β^+ 0.4Mev γ , 0.8, 0.3 e^- 0.3	Br^{78} 6.4 min. β^+ 2.3Mev γ , e^-	Br^{79} 50.6%
^{34}Se	Se^{71} 44 min. β^+	Se^{72} 9.5 days K	Se^{73} 6.7 hours β^+ 1.3Mev	Se^{74} 0.87%	Se^{75} 127 days K, γ , e^-	Se^{76} 9.0%		
^{33}As						As^{75} 100%		
	37	38	39	40	41	42	43	44

NEUTRON NUMBER

MU 2131

Fig. 10

Isotopic Chart of Bromine Region

5. The H_2O and CCl_4 layers were shaken, and then separated.
6. The Br_2 was reduced out of the CCl_4 layer with NaHSO_3 into a few ml H_2O .
7. A few drops of Ag^+ carrier were added, to precipitate the Br^- .

A typical decay curve of the bromine fraction is shown in Figure 11. This curve represents the results from a 10 minute bombardment. All experiments gave very similar decay curves, except for small variations in the ratios of the two components, due to differences in length of bombardment. The half-lives of the two activities, clearly resolved, have been determined as 35 ± 5 minutes and 95 ± 5 minutes.

The 95 minute activity is in all likelihood the same isotope of bromine which was reported by Pool et al.¹³ as 1.7 hour Br^{75} , produced from the (p,γ) reaction on selenium enriched in Se^{74} . From Pool's data, it is fairly certain that this isotope is either Br^{74} or Br^{75} , but his assignment of the activity to the latter does not seem to be definitive. He states, "In the decay curve of activity from $\text{Se}^{74} + p$ bombardment, a 125 ± 5 day half-life appeared which was presumed to be that of Se^{75} . The ratio of cross sections for production of the 1.7 hour Br activity compared to the 127 day Se^{75} activity by proton bombardment was determined to be approximately one. This indicates that the 1.7 hour Br activity decays into the 127 day Se^{75} period." However, in the absence of knowledge of the decay schemes of these nuclides, one cannot come to a conclusion such as the above without making some assumption regarding their counting efficiencies; experience with K-capturing isotopes with small amounts of positron branching has shown that such assumptions can be quite misleading.

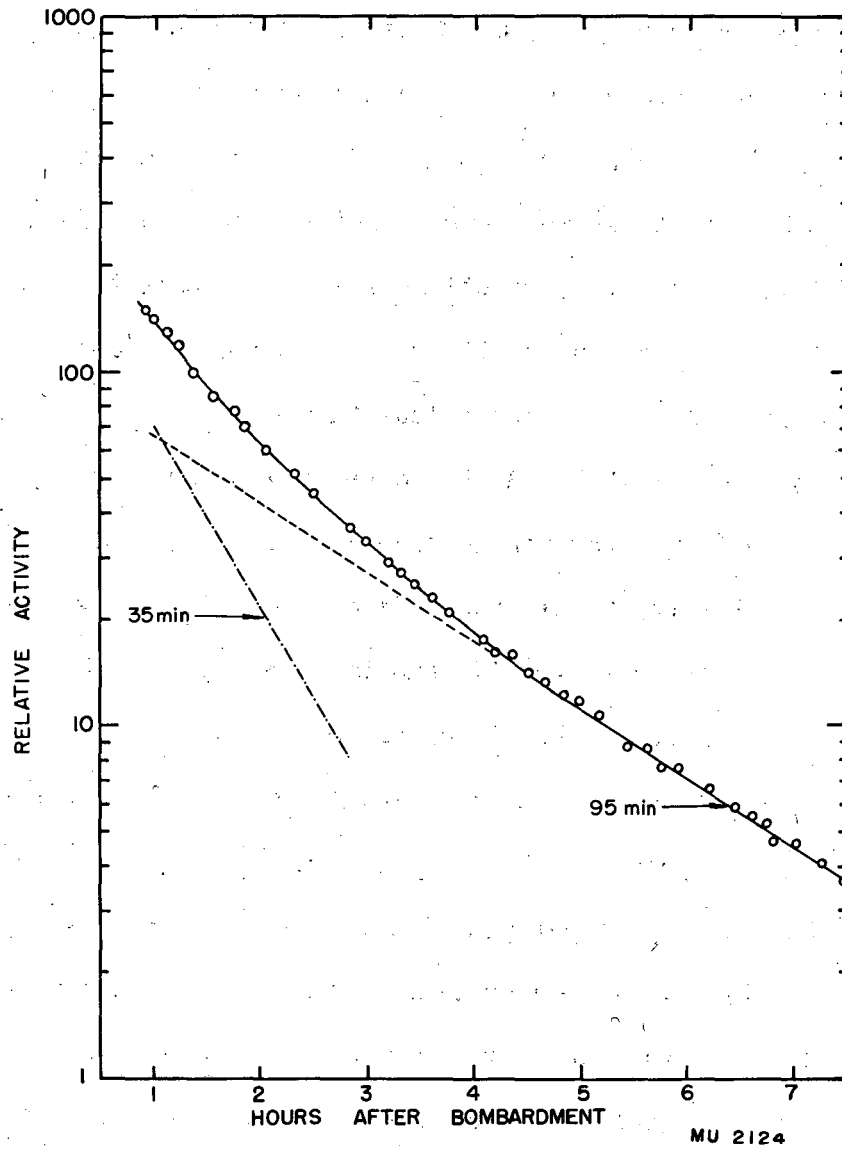


Fig. 11

Decay Curve of Bromine Fraction
from $\text{Cu} + \text{C}^{12}$

Though one acknowledges that the parent of the 127 day activity was, indeed, Br^{75} , whether or not it was the 1.7 hour isotope is still in doubt.

There are some considerations to favor the argument of Pool: that the (p,γ) cross section should be about as large as the (p,n) cross section in his bombardments of selenium, in view of the fact that protons of 5 Mev were used. Quite high threshold energies are often required by (p,n) reactions where the transition is from an even-even to an odd-odd nucleus, as it is in the case of ${}_{34}\text{Se}^{74}(p,n){}_{35}\text{Br}^{74}$. A rough calculation using the mass equation indicates the threshold in this case to be approximately 7 Mev, whereas the (p,γ) reaction to give Br^{75} is exothermic. Thus it is quite possible that these bombardments were done at an energy very close to the (p,n) threshold, and that the (p,γ) reaction constituted a sizable fraction of the total cross section.

Beryllium absorption measurements on our 90 minute activity showed an end point of the beta spectrum of approximately 1.8 Mev, in qualitative agreement with the value of 1.6 Mev as reported by Pool.

The difficulty of ascertaining the mass of this nuclide is increased by the fact that the several possible daughters are either stable or very long-lived, so that chemical isolation of the daughter would be of no value. Such a situation might best be met by carefully controlled excitation experiments with helium ions on arsenic (which contains only one stable isotope, ${}_{33}\text{As}^{75}$). By varying the bombarding energy between 40 and 60 Mev, one should be able to bring in successively the $(\alpha,4n)$ to $(\alpha,6n)$ reactions; the mass number is

assigned by noting where the 90 minute activity first appears.

In the bombardments of copper with carbon ions, the decay curve of the bromine fractions showed only the two activities, and in all experiments decayed to background level without tailing out into a longer-lived component. This would indicate that the new 35 minute activity is not the parent of 7.0 hour Se^{73} , which should have appeared as a tail in the decay curve. Br^{72} is not ruled out, because of the "long" half-life of its daughter, 9.5 day Se^{72} . Therefore, the present choice of mass assignment for the 35 minute bromine rests between Br^{72} and Br^{74} ; half-life considerations would favor the latter.

To rule out the possibility that the 35 minute activity might be Se^{71} (44 minute β^+) impurity, the following were done:

(a) In one bombardment, the chemical procedure was altered to the following:

1. The copper target was dissolved in HNO_3 , and 10 mg Br^- carrier added.
2. Solution was made 1 N in HNO_3 .
3. Equal volume of CCl_4 added, and 0.1 M KMnO_4 added dropwise until Br_2 color persisted. Br_2 was extracted into CCl_4 several times.
4. To CCl_4 layer, 10 ml H_2O and 0.1 N NaHSO_3 were added until the CCl_4 layer became colorless upon shaking.
5. H_2O and CCl_4 layers separated.
6. Steps (3) and (4) repeated twice.
7. Br^- precipitated from solution by addition of Ag^+ .

The activities found were the same as in all other experiments.

(b) A bombardment of copper with carbon ions was made, in which a selenium fraction was separated. Periods of ~40 minutes, 7.0 hours, and 9 days were observed, corresponding to Se^{71} , Se^{72} , and Se^{73} from the $(\text{C},\text{p}5\text{n})$, $(\text{C},\text{p}4\text{n})$, and $(\text{C},\text{p}3\text{n})$ reactions on Cu^{65} and the $(\text{C},\text{p}3\text{n})$, $(\text{C},\text{p}2\text{n})$, and (C,pn) reactions on Cu^{63} .

The repeated absence of the last two periods in bromine fraction decay curves rules out the presence of selenium impurity, in view of the fact that the 35 minute period always occurred in high abundance (comparable to that of the 1.5 hour period).

To rule out the possibility that the 35 minute activity might be 38 minute Zn^{63} made in $\text{Cu}^{63}(\text{d},2\text{n})$ reactions from deuteron contamination in the carbon beam, the following experiment was done: Copper was bombarded in the regular deuteron beam of the 60-inch cyclotron for 10 minutes, and then subjected to the bromine distillation chemistry. Very little activity was observed in the "bromine" fraction; what activity there was decayed with periods either <20 minutes or >6 hours.

Assignment of the 35 minute bromine activity to Br^{74} seems the most reasonable in light of present data. The reaction would be $\text{Cu}^{65}(\text{C},3\text{n})\text{Br}^{74}$.

Mass Assignments of Br^{76} and Br^{77}

In the course of the bromine work, it was considered advisable to make certain the isotopic assignments of Br^{77} and Br^{76} . These nuclides had been characterized in the Table of Isotopes¹⁴ as follows:

Br⁷⁷ -- 57 hour β^+ , K Element certain, isotope probable.

Br⁷⁶ -- 16 hour β^+ Element certain, mass assignment uncertain.

Since arsenic has but one stable isotope, $^{75}_{33}\text{As}$, the method of critical excitation was selected for this problem, utilizing the following reactions:

$\text{As}^{75}(\alpha, n)\text{Br}^{78}$, threshold estimated at ~6 Mev.

$\text{As}^{75}(\alpha, 2n)\text{Br}^{77}$, threshold estimated at ~14 Mev.

$\text{As}^{75}(\alpha, 3n)\text{Br}^{76}$, threshold estimated at ~26 Mev.

Three bombardments of As_2O_3 powder wrapped in platinum envelopes were made in the 60-inch cyclotron helium ion beam. The thickness of the platinum was changed in each bombardment, to provide the variation in beam energy.

(a) 2 mil platinum, $E_\alpha \approx 33$ Mev.

(b) 3 mil platinum, $E_\alpha \approx 24$ Mev.

(c) 5 mil platinum, $E_\alpha \approx 11$ Mev.

Separation of the active bromine from the As_2O_3 target material was effected in the following way:

(1) The As_2O_3 powder was dissolved in hot dilute KOH, and several mg of Br^- carrier added.

(2) Solution was placed in distilling flask, with slow air stream passing through.

(3) Solution slowly acidified by addition of HNO_3 with heating, and the Br_2 swept over into a CCl_4 trap.

(4) The CCl_4 (containing the Br_2) was shaken with a few ml 0.1 N HNO_3 to which had been added a trace of KMnO_4 .

(5) The Br_2 was reduced out of the CCl_4 with NaHSO_3 and then precipitated from the aqueous phase by addition of Ag^+ .

The decay curves from the three bombardments are shown in Figures 12, 13, and 14. The activities produced are given below:

Run	Platinum Thickness (mil)	Beam Energy Reaching Target (Mev)	Activities Observed	Reaction
(a)	2	~33	16.5 ± 0.5 hour	($\alpha, 3n$)
			57 ± 1 hour	($\alpha, 2n$)
			6 ± 1 minute	(α, n)
(b)	3	~24	57 ± 1 hour	($\alpha, 2n$)
			6.5 ± 1 minute	(α, n)
(c)	5	~11	6.5 ± 0.5 minute	(α, n)

From these results, it may be inferred fairly unambiguously that the isotopic assignments as given in the Table of Isotopes are correct.

The positron spectrum of the 16.5 hour Br^{76} was examined, using the crude beta spectrometer, or "bender." An end point of the positron distribution was observed which corresponded to an energy of 3.5 Mev.

An electron conversion line of 350 ± 50 kev was observed; this line decayed with a half-life of 53 ± 5 hours, indicating that it is associated with a gamma ray from Br^{77} . This was verified by bombardment (b), in which also the upper limit of the positron spectrum of Br^{77} was found to be 400 ± 50 kev.

The electromagnetic radiation from Br^{77} was cursorily examined in a gamma ray pulse analyzer which is still in the developmental stage. A very complex spectrum with many lines was seen; however, in addition to the intense annihilation gamma line at 0.5 Mev, there seemed to be an indication of strong lines at ~0.8 and ~0.3 Mev. These figures are probably quite rough.

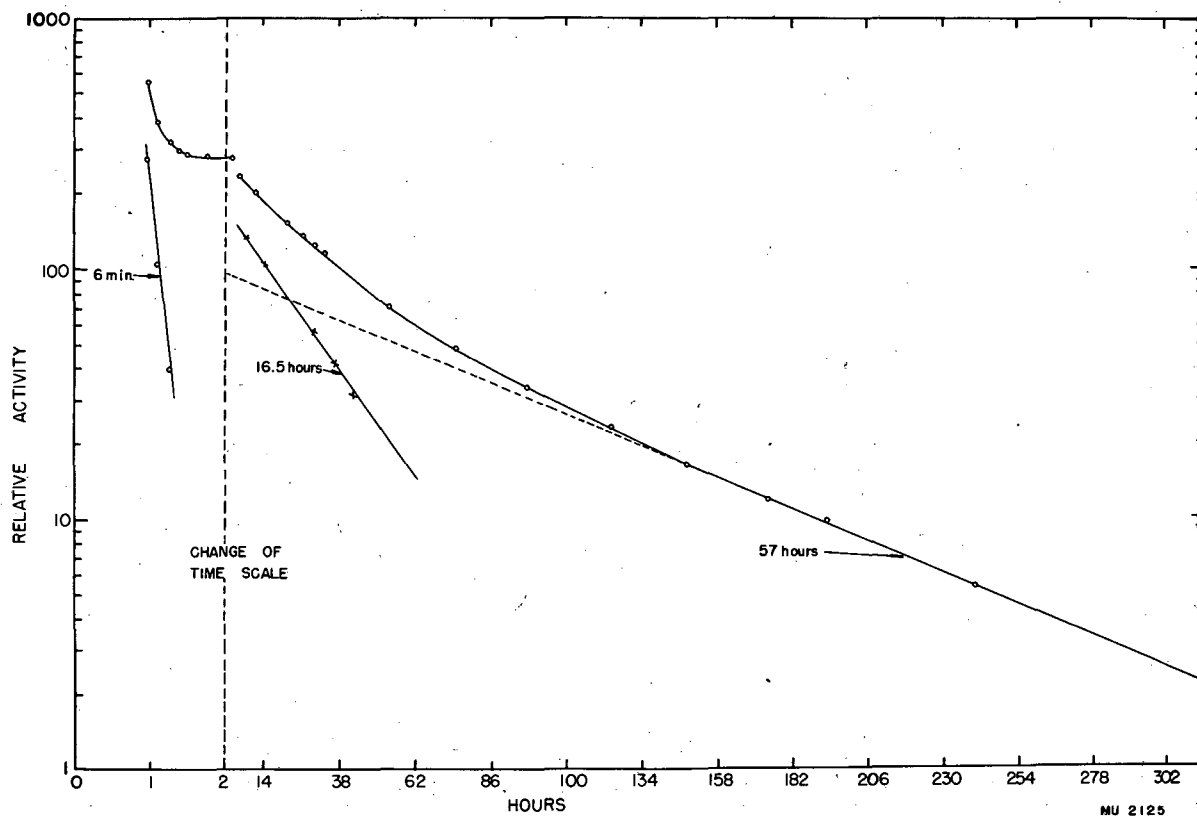


Fig. 12

Decay Curve of Bromine Fraction
from $\text{As}_2\text{O}_3 + 33 \text{ Mev He}^4$

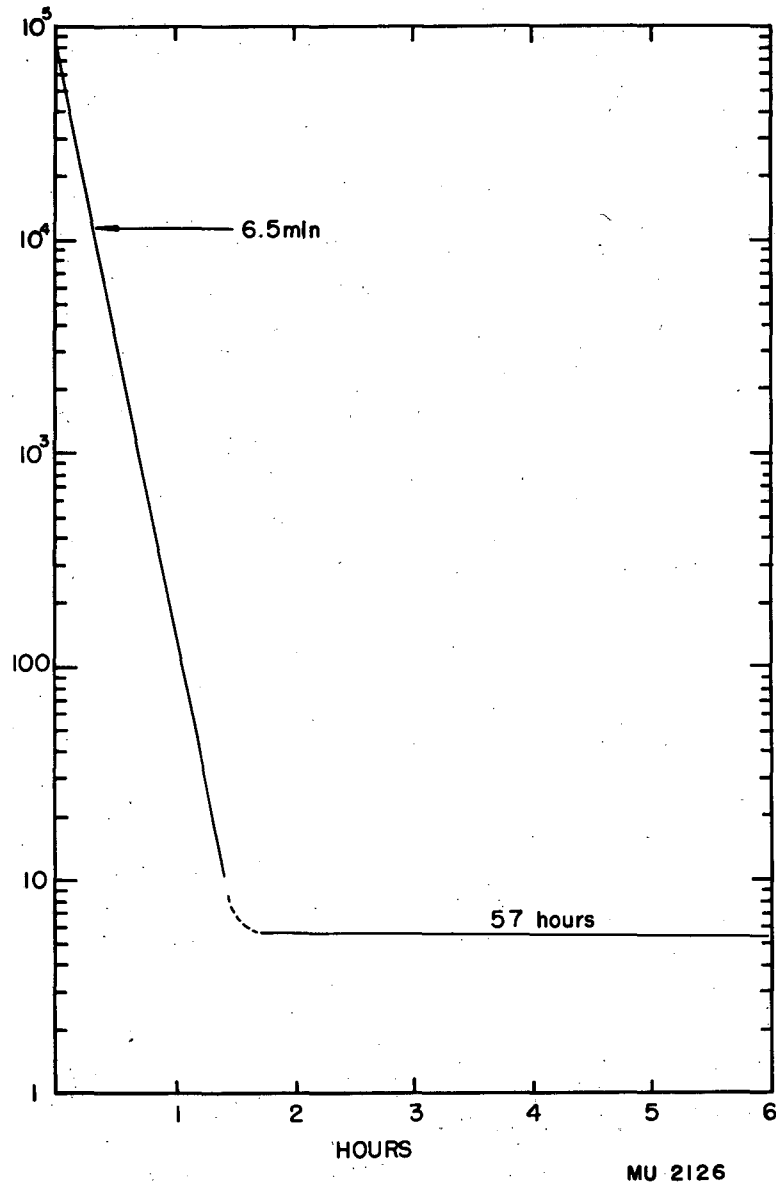
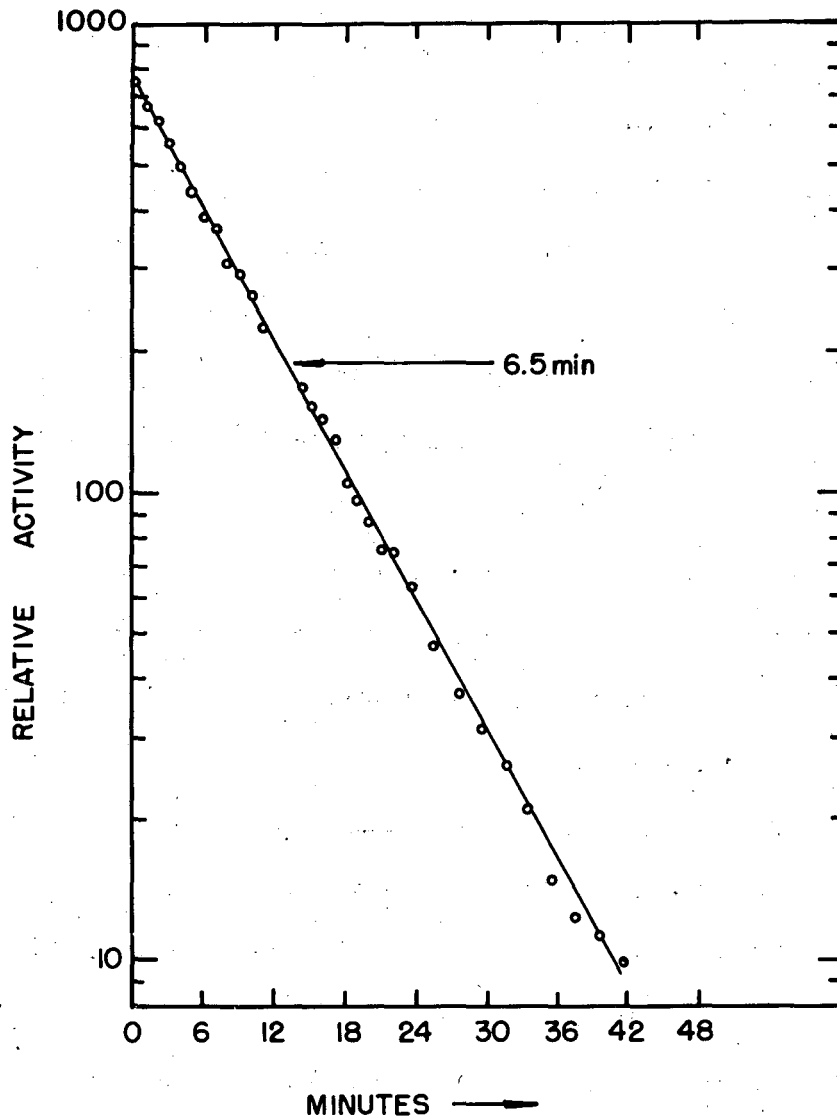


Fig. 13

Decay Curve of Bromine Fraction
from $\text{As}_2\text{O}_3 + 24 \text{ Mev He}^4$



MU 2127

Fig. 14
Decay Curve of Bromine Fraction
from $\text{As}_2\text{O}_3 + 11 \text{ Mev He}^4$

V. PRESENT THEORY OF CARBON BEAM FORMATION

Recent experiments by A. Ghiorso and B. Rossi have established that 90 Mev $C^{12}(+6)$ ions are found at a smaller dee radius than would be possible if these particles had originated at the ion source, in the geometric center of the dee. This information plus the fact that the intensity of the $C(+2)$ beam and the $C(+6)$ beam seem to be related have led to the following theory of the $C(+6)$ beam formation:

The ion source, which does not produce electrons of sufficiently high energy to strip the carbon atom completely, produces an intense beam of $C(+2)$ ions riding on the $1/3$ harmonic, as discussed earlier. Some of the $C(+2)$ ions lose electrons by collisions with air molecules (or fast electrons) near the ion source, and a small fraction of these which reach $C(+6)$ get in resonance and are accelerated to the full radius. This mechanism is able to explain the following phenomena:

- (a) The unusually large ratio of external to internal carbon beam intensity. Because of the manifold centers of formation of $C(+6)$ ions, the resulting beam will have components with many radii of curvature; only a small fraction of these will be passed by the exit channel for the external beam.
- (b) Wide energy distribution of the beam. Those ions which reach the $C(+6)$ resonance far from the ion source will travel a much shorter distance than those ionized near the center in reaching the periphery, and will attain a much lower energy.
- (c) Of boron, carbon, nitrogen, oxygen, and fluorine, only carbon ions have thus far been successfully accelerated in

the 60-inch cyclotron. Since the voltage of the ion source is not high enough to completely ionize any of these atoms, it should be necessary to initiate the beam in a sub-harmonic. The $1/3$ harmonic is possible only in the case of C^{12} ; the successful utilization of lower harmonics such as $1/5$, etc., is probably much less probable.

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REFERENCES

1. C. A. Tobias, Ph.D. Thesis, University of California, 1940.
2. G. W. Barton, A. Ghiorso, and I. Perlman, Phys. Rev. 82, 13 (1951).
3. A. Ghiorso, A. H. Jaffey, H. P. Robinson, B. B. Weissbourd,
National Nuclear Energy Series, Plutonium Project Record, Vol. 14B,
"The Transuranium Elements: Research Papers," Paper No. 16.8
(McGraw-Hill Book Co., Inc., New York, 1949).
4. J. F. Miller, J. G. Hamilton, T. M. Putnam, H. R. Haymond, and
G. B. Rossi, Phys. Rev. 80, 486 (1950).
5. W. A. Aron, B. G. Hoffman, and F. C. Williams, "Range-Energy
Curves," University of California Radiation Laboratory Report
UCRL-121 (1949).
6. G. T. Seaborg and I. Perlman, Phys. Rev. 77, 26 (1950).
7. Los Alamos Lecture Series, Chapter V (Lecture by V. F. Weisskopf).
8. E. Fermi, "Nuclear Physics," The University of Chicago Press,
Chicago, Illinois, 1947, p. 35.
9. A. Ghiorso, S. G. Thompson, K. Street, and G. T. Seaborg,
Phys. Rev. 81, 154 (1951).
10. S. Frankel and N. Metropolis, Phys. Rev. 72, 914 (1947).
11. R. H. Goeckermann and I. Perlman, Phys. Rev. 76, 628 (1949).
12. P. R. O'Connor and G. T. Seaborg, Phys. Rev. 74, 1189 (1948).
13. E. L. Woodward, D. A. McCown, and M. L. Pool, Phys. Rev. 74,
870 (1948).