The Yield of F^{18} from Medium and Heavy Elements with 420-Mev Protons

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The yield of F^{18} from medium and heavy elements, when bombarded with 420-Mev protons, was determined. Chemical procedures and techniques are described and the cross sections varying from 2.2×10^{-2} barn for F to 4.4×10^{-6} for Au are given. Comparisons are made of these results with those of other heavy fragments obtained under similar conditions.

INTRODUCTION

PREVIOUS work[1,2] has definitely established that Li and Be are produced with 340-Mev protons, and some cosmic-ray evidence indicates that some heavier fragments are produced likewise.[3] The purpose of this work was to study the yield of one of these heavy fragments under controlled laboratory conditions as are available in the synchrocyclotron at the University of Chicago.

F^{18} was chosen because it has a fairly energetic positron of 0.64 Mev which makes its counting simple, and it has a half-life of 112 min which is also adequate for radiochemical measurements.

The experiments were carried out at 420 Mev instead of 450 Mev which is the maximum energy available to get higher cyclotron currents, since it drops fast in this region. It corresponds to a radius of 74 inches.

TARGETS AND CHEMICAL PROCEDURES

The elements bombarded were O, F, Al, Cl, Cu, Ag, Au. Oxygen was bombarded as Li_2CO_3 and as H_3BO_3, fluorine was bombarded as a teflon foil $(C_2F_4)_n$, chlorine was bombarded as NH_4Cl, and all the other elements were bombarded as metal foils. All the targets were of high chemical purity (99.9 percent or better). The metal foils were wrapped in foils of the same metal, and the powder targets were wrapped in foils of neighboring elements, for instance, Li_2CO_3 and H_3BO_3 were wrapped in cellulose acetate. The stack of foils was sandwiched between two aluminum foils of nearly the same size as the target, and they served as monitors to measure the cross sections through the reaction Al^{27}-$(p,3pn)Na^{24}$.

Great care was taken that the aluminum and the inner target foil would not face each other to prevent recoiling F^{18} nuclei from reaching the target. The stack of foils was irradiated with the beam perpendicular or parallel to the direction of its thickness according to the desired yield; the size of the stack was always about 2 cm×1 cm×0.2 cm. The length of irradiation varied also from 15 sec to one hour. The check for the homogeneity of the bombardment was the closeness of the specific activities in the Al foils; they never differed by more than a factor of two in irradiations with the beam perpendicular to the direction of the thickness and a few percent with the bean parallel to the direction of the thickness.

The irradiations of H_3BO_3, teflon, and Al were counted without doing chemistry since all the activities could be accurately resolved.

To the other targets were added 20 to 40 mg of F^- carrier and dissolved in an appropriate solvent which was H_2O for NH_4Cl, HNO_3 for Cu and Ag, and aqua regia for Au; immediately diluted with water (gold was extracted with ethyl acetate) and 10 ml of 1.5M $BaCl_2$ added followed by ammonia. The BaF_2 precipitate was treated differently, according to the amount of contaminating activity remaining. But the basic steps were as follows: the BaF_2 was dissolved in 3N HCl and Ba^{++} removed with an excess of $SO_4^=$; 1 mg of carriers of suspected activities was added and the solution made basic with ammonia, several precipitations of $Fe(OH)_3$ made as scavenger and precipitation of sulfides with H_2S. The solution was made acid with HCl, the $SO_4^=$ removed with a large excess of Ba^{++}, and then when made basic with ammonia, the BaF_2 precipitated. It was dissolved and treated as previously but bubbling H_2S in acid solution this time and then scavenging with $Fe(OH)_3$ and the BaF_2 reprecipitated as described. Finally, the BaF_2 was again dissolved and the Ba removed with $SO_4^=$, diluted to 20 ml, made basic with NH_4OH, and 50 mg of Ca^{++} added to precipitate CaF_2. In this last step all reagents were kept to a minimum. The CaF_2 was centrifuged and thoroughly washed with H_2O and centrifuged twice, suspended in H_2O and placed in a tared Al dish, dried at 115°C and weighed as CaF_2.

From the basic procedure changes were made. For instance, for Cl only one iron scavenging was done before precipitating CaF_2, and for Au the whole purification scheme was done twice before precipitating CaF_2. Accordingly the chemical yield varied from 75 percent to 20 percent.

CROSS SECTIONS

The samples containing F^{18} were counted in an end-window Geiger counter at a distance of 3.4 cm from the window. The identity of F^{18} was not only established by the chemical procedure but also by an accurate determination of its half-life which never differs more

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[2] L. Marquez and I. Perlman, Phys. Rev. **81**, 953 (1951).
[3] D. H. Perkins, Proc. Roy. Soc. (London) **203**, 399 (1950).

TABLE I. Yields of F^{18}, N^{13}, and C^{11} from various nuclei with 420-Mev protons.

Target nucleus	Product nucleus	Yield in barns	Estimated error (%)
O	F^{18}	8.3×10^{-5}	25
O^{18}	F^{18}	4.1×10^{-2}	25
F	F^{18}	2.2×10^{-2}	25
Al	F^{18}	8.4×10^{-3}	15
Cl	F^{18}	2.2×10^{-3}	25
Cu	F^{18}	8.0×10^{-5}	50
Ag	F^{18}	1.6×10^{-5}	50
Au	F^{18}	4.4×10^{-6}	50
O	C^{11}	3.1×10^{-2}	25
O	N^{13}	1.4×10^{-2}	25

than 2 min from the half-life of 112 min. This activity measurement, extrapolated back to the time of bombardment and properly corrected for the length of irradiation and for self-absorption and absorption in the air and in the window, was used to calculate the cross sections.

The Al monitors were counted about 24 hours after the irradiation; then it was only pure 15.1-hr Na^{24}. If it was counted later, however, it was followed through in order to deduct for the Na^{22} activity. The monitors were mounted in Al dishes equal to those where the CaF_2 were mounted; since back-scattering is independent of energy,[4] this eliminated automatically the back-scattering correction. To correct for self-absorption and absorption in the air and window the absorption coefficient of F^{18} and Na^{24} were measured with Al absorbers and found to be very close to those given by the formula $\mu=0.017E^{-1.43}$; as given by Gleason[5] this is of course under the assumption that absorption is exponential at the beginning. All cross sections are corrected for self-absorption and absorption in the air and window. The thickness of the Al foil was about 9 mg/cm², that of the CaF_2 precipitate about 10 mg/cm², and that of the air and window added up to 7 mg/cm². The cross section for the reaction $Al(p,3pn)Na^{24}$ was found to be 10.8 ± 1.1 mb, as it will be described in the next section.

The cross section for the production of N^{13} from O and the cross section for the production of C^{11} in O were found as an incidental part of this work. These, together with the F^{18} cross sections are given in Table I. The value for O is given as the actual cross section, and the cross section was calculated as if all the F^{18} came from O^{18}, which is the most likely case.

The values quoted in estimated error are based upon reproducibility, estimates of the uncertainty of the CaF_2 yield in the heavy elements where the chemistry was more cumbersome, and uncertainty in the absorption correction, etc.

SPALLATION OF Al

It is a known experimental result that many cross sections for production of radio isotopes at high energies are very slowly varying functions of energy. In order to see if there is much change going from 340 Mev to 420 Mev, the spallation of Al at 420 Mev was studied. The results are compared in Table II. The values at 420 Mev are reported in two columns, first uncorrected, to compare with the uncorrected values taken at Berkeley[2] under very similar conditions and then the values corrected for self-absorption and absorption, which we consider better.

It can be seen that the results at 340 Mev and 420 Mev are identical within the experimental error. The values in Table II are estimated to be good within 15 percent, except the value of Na^{24} whose error is 10 percent.

The absolute cross section for the formation of Na^{24} from Al was measured at 450 Mev by irradiating an Al target in the circulating beam, the current was determined by measuring the heat evolved in the target and using the value of dE/dx from the formula from Bethe,[6] and the value of I was taken from the experimentally determined value of Segrè.[7] Besides this we added an estimate of the heat from nuclear events. For this estimate we took for the cross section for 420-Mev protons on Al the value 0.29 barn, which is one-half of the total neutron cross section at 240 Mev on Al,[8] and we guessed from other experiments that in each collision the ejected charged particles and the recoiling nucleus dissipate 30 Mev in the target which goes into heat.

The Al target was 4 Mev thick and was screwed tightly to an Al block, the whole weighing about 70 g. The block was insulated with a piece of plastic from the probe, and the temperature of the block was measured with a thermocouple; a plot of the rise in temperature against time indicated that this arrangement was a good calorimeter from which the heat deposited in the target was measured. The length of irradiation was 20 min. The rise in temperature was corrected for the small leakage of heat, and from the known heat capacity of the calorimeter, whose weight was accurately determined, the heat deposited in the target was measured. We estimate the error of this measurement to be 5 percent. The target was dissolved in HCl, several small aliquots ($\sim$1/20,000) taken, dried, and counted.

TABLE II. Yields of spallation products of Al with 335-Mev protons and 420-Mev protons.

Nucleus	Yields (barns) 335-Mev protons (uncorrected)	Yields (barns) 420-Mev protons (uncorrected)	Yields (barns) 420-Mev protons (corrected)
Na^{24}	1.02×10^{-2}	1.0×10^{-2}	1.08×10^{-2}
Na^{22}	1.2×10^{-2}	1.2×10^{-2}	1.7×10^{-2}
F^{18}	5.5×10^{-3}	6.3×10^{-3}	8.4×10^{-3}
N^{13}	not measured	8.9×10^{-4}	9.7×10^{-4}
C^{11}	1.9×10^{-3}	2.4×10^{-3}	2.8×10^{-3}
Be^{7}	1.4×10^{-3}	not measured	...

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[5] Gleason, Taylor, and Tabern, Nucleonics **8**, No. 5, 12 (1951).
[6] M. S. Livingston and H. A. Bethe, Revs. Modern Phys. **9**, 263 (1937).
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[8] J. DeJuren and B. J. Moyer, Phys. Rev. **81**, 919 (1951).

The backing was rubber hydrochloride of 1 mg/cm² thickness; the dried sample weighed about 1 mg/cm².

Two techniques were used to determine the absolute beta-disintegration rate of the sample. One was to count the sample and a RaD, E, F standard mounted on the same rubber hydrochloride in a technique as described by Novey.[9] The other was to count the sample in an arrangement of well-defined geometry as described by Gleason *et al.*[5] The average of these two methods had a probable error of 7 percent.

The heat evolved in nuclear events is about 8 percent of the heat evolved by ionization, and we estimate that our guess for this heat is right within a factor of two; however, if later experiments would show that this quantity is much different, the value of the cross section could be properly corrected.

The value for the cross section for the formation of Na^{24} from Al with 450-Mev protons thus found is 10.8 ± 1.1 mb. In the error quoted all sources of error that we know were included. It is very close to the value of 10 mb at 350 Mev found by Stevenson.[10]

RESULTS

All the results of this work at 420 Mev, together with previous results at 340 Mev and some recent results on yields obtained by Batzel and Seaborg[11] and by Greenberg and Miller,[12] are shown in Fig. 1. It also includes the value for $C^{12}(p,pn)C^{11}$ from Aamodt *et al.*[13] It will be noticed that the curves look alike in all the cases. At the beginning, from a peak there is a fast decrease of yield with increasing mass number which has the same shape for all of them, and then they turn and decrease more slowly. This is related to whether the nucleus is the product nucleus or the ejected nucleus.

It will be noticed also that the yield of the light fragments always turns sooner and crosses over the heavier ones. In the heavier elements the lightest fragments have larger yields. This seems to be somewhat related to the Coulombic barrier. But these yields, as pointed out previously,[2] are hard to reconcile with an evaporation process and are more likely knock-on products. That is, the high energy proton produces through some mechanism the ejection of the heavy fragment before the excitation energy is evenly distributed throughout the nucleus.

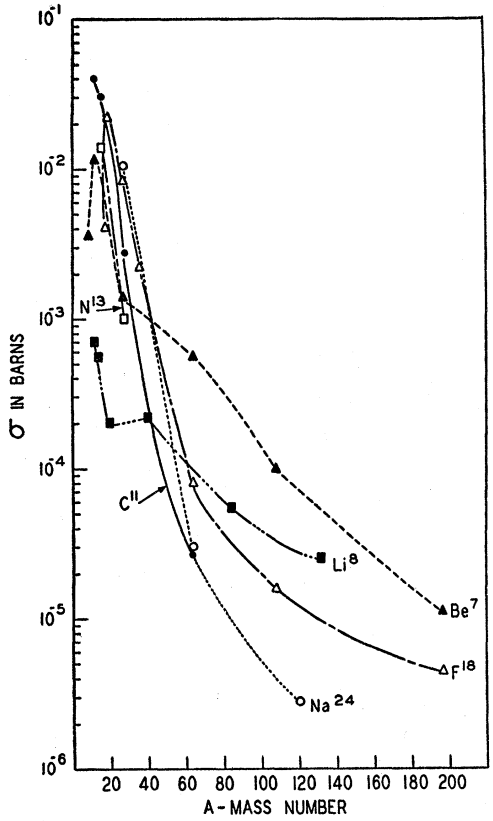

FIG. 1. Yield of heavy fragments in barns as a function of mass number with protons from 335 Mev to 420 Mev.

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[13] Aamodt, Peterson, and Phillips (University of California Radiation Laboratory Unclassified Report No. 526, November, 1949).