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Spallation of Cu with High Energy Neutrons

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The yields of spallation products of copper with high energy neutrons have been measured and are given. The results are similar to those obtained with high energy protons. This similarity and some apparent differences are pointed out.

I. INTRODUCTION

SO far, there have not been any experimental results of spallation with high energy neutrons, although there are some results of star production with high energy neutrons[1,2] and there are results of spallation in Sb with protons, deuterons, and alpha-particles,[3] in As with deuterons,[4] in Cu with protons, deuterons, and alpha-particles[5] and some others. Of all these spallation works, it seems that the work on Cu by Batzel, Miller, and Seaborg is the most careful and accurate. So it seemed that the study of the spallation of Cu with neutrons and its comparison with the results with protons and deuterons might shed some light on the question of the nuclear forces and more specifically on the question of charge independence.

II. IRRADIATION PROCEDURE

The irradiations were done in the synchrocyclotron at the University of Chicago. The 450-Mev proton beam at a radius of about 76 inches was allowed to strike a Be target of 2-inch thickness. This produced a beam of neutrons mostly in the forward direction, the energy distribution of this beam of neutron has a peak at 370 Mev; it decreases smoothly towards higher energies until it vanishes at about 440 Mev; toward lower energies it is smaller by a factor of five from the peak at 300 Mev and keeps going down smoothly at smaller energies.[6] This neutron distribution is similar to those obtained by other investigators.[7] Throughout this paper, the expression high energy neutrons means this energy distribution just described.

Near the synchrocyclotron tank there is a shield of 12-foot thickness made of concrete and iron in which there is a hole located in such a way as to let the neutron beam go through. Its central axis coincides with the tangent to the orbit at the point where the Be is located inside the tank.

This thick shield was also suitable to study the influence of secondary radiation in our experiments by irradiating samples in the hole near the tank and far from the tank. Secondary protons, mesons, etc., probably do not travel parallel to the neutrons because the magnetic field will deviate them. Only the gamma-rays and slow neutrons would follow exactly the same trajectory as the high energy neutrons. The gamma-rays have a small nuclear cross section and the slow neutrons produce nuclear reactions whose products are, in most cases, different from the spallation products, except Cu^{64} and perhaps Ni^{65} in the irradiation of Cu and Mg^{27} in the irradiation of Al.

The targets consisted of copper foils wrapped in a cylindrical shape and the irradiations were monitored by a foil of Al on one side of the cylinder. In the Al the well-known activity of Na^{24} was produced, and it served to monitor the irradiations. Irradiations lasted from 10 min to 3 hr. The fast neutron flux was of the order 10^5 neutrons/cm^2 sec.

The Cu and Al targets were placed in the hole in the shield in two positions; one close to the tank and the other far from it at the other side of the shield.

It was found that the yields were independent of whether the Cu and Al targets were close to the tank or far from it, except for Cu^{64}. This seems to prove that what was causing the nuclear reactions was coming from the Be target inside the tank in a straight line. The value for Cu^{64} might be somewhat high due to thermal neutron background.

III. CHEMICAL AND COUNTING PROCEDURES

The chemical procedures used were essentially those described by BMS,[5] or some described in the compilation of chemical procedures used in Berkeley,[8] except in the first step of the chemistry in which we usually had a heavy target of 20 to 50 g, from which about 20 mg of carrier had to be separated. This first step was accomplished in a different way for each of the different elements.

Cobalt and Nickel

The target was dissolved with the carriers added in a mixture of concentrated HCl and 30 percent H_2O_2 and boiled. The Cu^{++} was reduced to Cu^{+} with Na_2SO_3

[1] Bernardini, Booth, and Lindenbaum, Phys. Rev. **85**, 826 (1952).
[2] H. Fishman and A. M. Perry, Phys. Rev. **86**, 167 (1952).
[3] M. Lindner and I. Perlman, Phys. Rev. **78**, 499 (1950).
[4] H. H. Hopkins, Jr., Phys. Rev. **77**, 717 (1950).
[5] Batzel, Miller, and Seaborg, Phys. Rev. **84**, 671 (1951), and unpublished University of California Radiation Laboratory Report No. 1077 (1951). Hereafter this paper will be referred to as the BMS paper.
[6] J. Marshall and A. Nedzel (private communication).
[7] Fox, Leith, Wouters, and MacKenzie, Phys. Rev. **80**, 23 (1950). Also J. DeJuren and B. J. Moyer, Phys. Rev. **81**, 919 (1951).
[8] W. W. Meinke, unpublished University of California Radiation Laboratory Report No. 432 (1949).

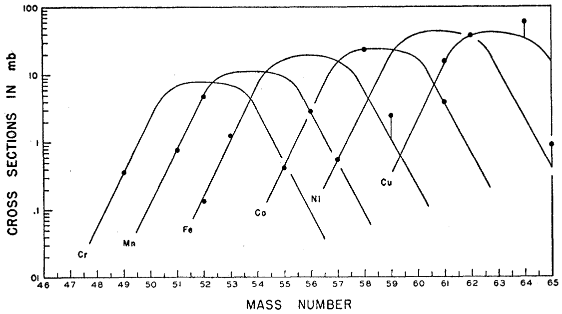

FIG. 1. Yield in millibarns of spallation products of Cu with high energy neutrons, as a function of mass number; each curve represents one element.

and CuSCN precipitated with NH_4SCN. From the filtrate Ni was precipitated with dimethyl glioxime by making the solutions slightly ammoniacal. From the filtrate also the Co was extracted, adding first an excess of NH_4SCN and then extracting with a mixture of amyl alcohol and ether.

Iron

The target with Fe^{+++} carrier was dissolved in a mixture of $12N$ HCl and 30 percent H_2O_2 and boiled. $12N$ HCl was added to make the solution of $8N$ in HCl, and from it the iron was extracted into isopropyl ether.

Chromium and Manganese

The target with Cr and Mn carriers was dissolved in concentrated HNO_3 by heating. From the boiling solution MnO_2 was precipitated by adding $KClO_3$ which also oxidized the Cr. The MnO_2 was separated by filtering through a sintered glass filter. The filtrate was diluted with H_2O and $6N$ NaOH added while heating until the solution was basic and the copper precipitated as CuO. The CuO was filtered and a concentrated solution of $BaCl_2 \cdot 2H_2$ added to the filtrate which gave a precipitate of $BaCrO_4$.

Titanium

The target with titanium carrier added was dissolved in HNO_3. The solution was diluted with water and made basic with ammonia. The copper remained dissolved in the ammonia complex form and the titanium hydroxide was centrifuged.

The samples for counting were spread over a circular area of 1.25-cm radius to have the same area as the aluminum disk used as monitor which also had a circular shape with 1.25-cm radius. All samples and monitors were counted on end-window Geiger counters at a distance of 0.4 cm from the window, and corrections were applied for coincidence, background, absorption, self-absorption, self-scattering, and backscattering. The four last corrections were made using the results of Gleason *et al.*[9] and Nervik and Stevenson.[10]

[9] Gleason, Taylor, and Tabern, Nucleonics **8**, No. 5, 12 (1951).
[10] W. E. Nervik and P. C. Stevenson, Nucleonics **10**, No. 3, 18 (1952).

IV. RESULTS

All cross sections measured in the spallation of copper relative to the cross section for the formation of Na^{24} from Al with high energy neutrons are shown in the third column of Table I. Since there are only one, two, or three points measured for each element, we tried to fit these data with a curve consistent with all of them by interpolating and extrapolating values. After that was done, it was possible to express the results as cross sections by adding the area under all curves, plus an estimate of the area under the lighter elements not measured, and equating this to one-half the total cross section for copper with high energy neutrons,[11] that is, 570 mb. In this way it was found that the cross section for the formation of Na^{24} from Al had to be 24.4 mb and the yield of spallation products of Cu in mb are given in the fourth column of Table I and shown in Fig. 1. We estimate that the error introduced by this method of measuring total cross section is about 50 percent which, however, applies to all results in the same direction. The experimental error of each cross section due to corrections, chemical yield, etc., is between 20 percent and 50 percent.

In the calculation of the cross sections, we used the counting efficiency given by BMS in order to make our results comparable; in the case of 9.2-hr Co^{58} which they did not report, we used 10 percent. The activity of 70-day Co^{58} was probably a mixture of Co^{56} and Co^{58}, since their half-lives are almost the same. In order to get the yield of 70-day Co^{58} we assumed that the yield of Co^{56} was that given by the interpolated yield curve of Co.

In Table II we compare the results of neutron spallation with the results of protons and deuterons. The second column has the yield in mb of the spallation product of copper with 340-Mev protons taken from

TABLE I. Yields of spallation products of copper with high energy neutrons.

Isotope	Observed half-life	Yield relative to yield of Na^{24} from Al	Yield in mb
Ti^{45}	3.5 hr	0.00032	0.0078
Cr^{49}	44 min	0.0142	0.35
Mn^{51}	47 min	0.031	0.76
Mn^{52}	5.9 day	0.192	4.68
Mn^{56}	2.58 hr	0.115	2.81
Fe^{52}	7.4 hr	0.0055	0.134
Fe^{53}	8.9 min	0.051	1.24
Fe^{59}	45 day	0.10	2.44
Co^{55}	18 hr	0.017	0.415
Co^{58}	9.2 hr	$\sim$2.0	$\sim$49
Co^{58}	70 day	0.95	23.2
Co^{61}	1.62 hr	0.155	3.78
Ni^{57}	36 hr	0.022	0.54
Ni^{65}	2.57 hr	0.036	0.88
Cu^{61}	2.9 hr	0.63	15.4
Cu^{62}	10.4 min	1.52	37.1
Cu^{64}	12.4 hr	2.40	58.6

[11] J. Marshall and A. Nedzel (private communication).

BMS's paper, and whose yield we determined also with high energy neutrons. We used the value of 17 mb for Cu^{64} as given in their University of California Radiation Laboratory report. The third column has the yield with 190-Mev deuterons, but since their value of 60 mb for Cu^{64} was a rough estimate, we used instead 30 mb, which brings the total cross section closer to the value 570 mb that it seems it should be. The fourth column has the ratio of the yield with protons divided by the yield with neutrons, and the fifth column has the ratio of the yields with deuterons divided by the yields with neutrons.

The spallation of Al with high energy neutrons was studied too, and the results are shown in Table III. In the case of neutrons, the high yield of 9.6-min Mg^{27} made it difficult to have an accurate value for the 10.1-min N^{13} and made it impossible to resolve the 20.4-min C^{11} activity. The yield of Mg^{27} was determined radiochemically and the others were measured directly from the irradiated Al foil. The yield of N^{13} is the difference between the total 10-min yield and the yield of Mg^{27}. These results are shown in Table III and compared with the protons results. The results with neutrons have of course the 50 percent error of the value 24.4 mb for Na^{24}, which was found as described previously, and this error also applies to all the neutron yields.

TABLE II. Yields of spallation products of copper with 340-Mev protons and 190-Mev protons according to BMS and ratios of these to the yields with high energy neutrons.

Isotope	Yield in mb with 340-Mev protons	Yield in mb with 190-Mev deuterons	Yield with protons/yield with neutrons	Yield with deuterons/yield with neutrons
Ti^{45}	0.75	0.06	96	7.7
Cr^{49}	0.70	0.30	2.0	0.86
Mn^{51}	1.21	0.87	1.59	1.14
Mn^{52}	5.27	2.61	1.13	0.56
Mn^{56}	1.87	3.00	0.67	1.07
Fe^{52}	0.136	0.090	1.01	0.67
Fe^{53}	1.24	1.41	1.00	1.14
Fe^{59}	0.58	1.35	0.24	0.55
Co^{55}	1.70	0.66	4.1	1.59
Co^{58}	42.5	...	1.83	...
Co^{61}	3.57	2.82	0.94	0.75
Ni^{57}	1.34	1.62	2.48	3.00
Ni^{65}	...	0.81	...	0.92
Cu^{61}	17.0	30	1.10	1.95
Cu^{62}	35.7	45	0.96	1.21
Cu^{64}	16.7	24	0.28	0.41

TABLE III. Yields of spallation products of Al with 420-Mev protons, high energy neutrons and their ratio.

Isotope	Yield in mb with 420-Mev protons	Yield in mb with high energy neutrons	Yield with protons/yield with neutrons
C^{11}	2.8	<3.4	>0.82
N^{13}	0.97	3.2±2.4	0.31±0.26
F^{18}	8.4	6.8±2.4	1.24±0.47
Na^{24}	10.8	24.4	0.44
Mg^{27}	...	5.1±2.0	...

V. CONCLUSION

It can be seen from the cross sections on Table I and II and from the ratios in columns four and five of that Table II that, while the cross sections for different nuclides with a given bombarding particle ranges from about 50 mb to a few tenths of a mb, the ratio of the yields of the same nuclide with different bombarding particles remains very closely the same. In fact, ten out of fifteen of the proton to neutron ratios in column four of the Table II lie between 0.67 and 2.00; and twelve out of fifteen of the deuteron to proton ratios in column five of Table II lie between 0.55 and 1.95.

However, a closer examination of these ratios reveals what seems to us a definite trend, which is very apparent in the proton to neutron ratios and somewhat less apparent in the deuteron case. The ratio is larger than unity for the lightest isotopes of a given element, which are isotopes formed in small yield. It is close to unity for isotopes near stability, which are isotopes formed in large yield. It is less than unity for isotopes on the heavy side, which again are isotopes formed in small yield. This same trend, less pronounced, seems to exist in the deuteron to neutron ratio and perhaps is somewhat blurred, because according to BMS, their results with deuterons are less accurate than their results with protons.

Perhaps we should say why we compared results with particles of quite different bombarding energies. The reason is that the yields in most spallation reactions studied so far in the region of 100 Mev to 450 Mev are slow varying functions of energy. Our results in an indirect way seem to corroborate it.

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