Radiochemical Evidence for the $Cu^{65}(p,p\pi^+)Ni^{65}$ Reaction*

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(Received March 29, 1954)

A study of the formation of the radioactive nickel isotopes in the irradiation of copper foil with high-energy protons has shown the presence of 2.56-hr Ni^{65}. The energy dependence of the cross section for the formation of this species, and its recoil properties, indicate that it is formed by the $Cu^{65}(p,p\pi^+)Ni^{65}$ reaction.

INTRODUCTION

THERE have been several attempts to demonstrate radiochemically the production of mesons in the irradiation of complex nuclei with high-energy particles.[1] These have usually involved examination for products with charges sufficiently higher than the target nucleus to require the accompanying formation of negative mesons. The results to date have been negative, due either to the low intensities available, or to the masking effect of secondary reactions produced in the target by alpha particles (or particles with higher charge) made in a primary interaction of the high-energy particle with the target. It is, therefore, of some interest to report radiochemical evidence for, and some characteristics of, a nuclear reaction unambiguously involving mesons.

The reaction studied was the production of 2.56-hr Ni^{65} in the irradiation of copper with protons.[2] Since the heaviest stable isotope of copper is Cu^{65}, the reduction in charge without reduction in mass number can only be accomplished with protons by the emission of one nucleon and two positive charges. The simplest interpretation is to assume that the reaction involved is $Cu^{65}(p,p\pi^+)Ni^{65}$. Quite obviously the production of Ni^{65} from impurities in the copper or via other particles than protons had to be excluded. Likewise the energy dependence of this reaction and the recoil properties of the product had to be consistent with the stated interpretation. In the following we give evidence on these points.

EXPERIMENTAL PROCEDURES

All the irradiations were made with the internal beam of the University of Chicago 170-inch synchrocyclotron. The radial position of the probe target was used to select the particle energy. A clamp arrangement held a thin pure copper foil (less than 10^{-2} percent zinc) sandwiched between 0.25-mil aluminum foils. The edges of all the foils were flush with each other so that all were exposed to the same beam intensity.

The target was arranged so that the proton beam first hit an aluminum foil A, then the copper foil, then four aluminum foils in the order B, C, D, and E. The Na^{24} or Na^{22} activity in foil D was used as a monitor of the beam intensity. The cross section for the production of these sodium isotopes from aluminum as a function of energy was taken from Stevenson and Folger,[3] Marquez,[4] and Fung.[5] After an irradiation, nickel was separated radiochemically from the copper foil and from the aluminum foils A, B, C, and E.

The chemistry consisted of repeated precipitations of nickel dimethyl glyoxime, with and without holdback carriers, together with numerous acid scavenging steps using copper, palladium, and antimony sulfides, and ammoniacal scavengings with iron and chromium. On several occasions the adequacy of the chemistry was tested by recycling.

The radioactivity was measured with end-window atmospheric pressure methane proportional counters. 2.6-hr and 36-hr half-lives were the main periods observed in the radiochemically purified nickel, and it was assumed that they were Ni^{65} and Ni^{57}, respectively.

The Ni^{57} and Ni^{65} radioactivities were found predominantly in the copper, and measured the production of these nuclides in proton reactions with copper. The amount of these nuclides in the aluminum foils C and E was very small (a few percent of the amount in foil B), presumably resulting from the activation of impurities in the aluminum. The amount of Ni^{57} and Ni^{65} in the aluminum foils A and B (after correcting for the amounts in C and E) measured the loss backward and forward from the copper due to recoil effects.

RESULTS

Table I gives the cross sections observed for the production of Ni^{57} (column 2) and Ni^{65} (column 3) from copper as a function of proton energy between 100 Mev and 440 Mev (column 1). It is seen that the cross section for producing Ni^{57} stays relatively constant ($\sim$1.5 mb) in this energy range. The value of 1.4 mb at 350 Mev is in satisfactory agreement with the value of 1.8 mb obtained by Batzel, Seaborg, and Miller with 340-Mev protons.[2] On the other hand, the

* This work was supported in part by a grant from the U. S. Atomic Energy Commission.
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[1] See, e.g., Bonner, Friedlander, Pepkowitz, and Perlman, Phys. Rev. **71**, 511 (1947).
[2] Batzel, Miller, and Seaborg, Phys. Rev. **84**, 671 (1951) studied the spallation of copper with 340-Mev protons and found both Ni^{57} and Ni^{65} among the products. They ascribed the formation of the latter to secondary neutrons causing (n,p) reactions (see text).
[3] P. C. Stevenson and R. L. Folger (private communication).
[4] L. Marquez (private communication).
[5] Si-Chang Fung (to be published).

TABLE I. Cross section for producing Ni^{65} and Ni^{57} and their recoil properties in the proton bombardment of copper.

Energy of proton Mev	Cross section for forming Ni^{57} (millibarns)	Cross section for forming Ni^{65} (millibarns)	Cross section for the reaction $Cu^{65}(p,p\pi^+)Ni^{65}$ [a] (millibarns)	Percent of Ni^{57} caught on Al foil[b] For.	Back.	Percent of Ni^{65} caught on Al foil[b] For.	Back.
100	1.3	0.006	0	1.14			
200	1.9			1.11			
200	1.8	0.009	0.003	1.06			
250	1.4	0.021	0.015				
250	1.5			1.07			
300	1.5			1.01			
300	1.3	0.032	0.026	1.05		1.47	
350	1.4	0.056	0.050				
400	1.4	0.078	0.072	0.93		1.11	
400	1.4			1.00			
430	1.4	0.087	0.081	0.91	0.095	1.10	0.06
430	1.3			0.93			
440	1.1	0.084	0.078	0.84		1.12	
440	1.2	0.092	0.086	0.77		1.12	
440	1.3	0.093	0.087	0.82		1.11	
440	1.3	0.088	0.082	0.96		1.17	

[a] The values in this column were obtained from those in the preceding one by subtracting 0.006 mb ascribable to secondary neutrons (see text).
[b] The main copper foil had a thickness of 23.9 mg/cm^2.

cross section for making Ni^{65} rises from 0.006 mb at 100 Mev to around 0.09 mb at 440 Mev, with a sharp rise occurring above 200 Mev.

Table I also indicates the recoil properties of Ni^{57} and Ni^{65} from 23.9 mg/cm^2 copper. It is seen that Ni^{65} recoils forward an amount comparable to, but always a little more than, Ni^{57}. The backward recoil was determined only at 430 Mev. It is very small for both nuclides, but the asymmetry seems definitely larger for Ni^{65}.

Because of the low cross section for producing the Ni^{65}, assorted extraneous mechanisms for making it had to be considered. In the first place, spallation or a fission-type reaction on impurities with charge higher than copper could give rise to Ni^{65}. To test this, the production of Ni^{66} (by milking its Cu^{66} daughter) was investigated. It was found that the ratio Ni^{66}/Ni^{65} was $\sim$0.005 in our experiments, whereas spallation and fission reactions form this pair in ratios much closer to unity. For example, this ratio varies between 0.5 and 1 in the 450-Mev proton spallation of elements between holmium and thorium[6] and is 0.2 in the spallation of arsenic by 190-Mev deuterons.[7]

A second possible extraneous source of Ni^{65} are (n,p) reactions in the copper induced either by neutrons formed in the target or by stray neutrons in the cyclotron. The contribution of these was investigated by studying the production of Ni^{65} relative to Ni^{57} under different experimental conditions. Ni^{57} is made from copper in high yield by the primary proton beam and should be made in low yield, if at all, with lower-energy neutrons. Important contributions from neutrons born in the target can be excluded on the basis of an experiment involving a target made up of five copper foils of standard thickness and somewhat wider. These gave a Ni^{65}/Ni^{57} ratio of 0.0272 in each foil under certain conditions of irradiation, counting, and decay, compared to a ratio of 0.0247 obtained in single-foil irradiations. The average path length of secondary neutrons is considerably larger in this thick-foil experiment than in the thin-foil experiment. The small increase in the Ni^{65}/Ni^{57} ratio observed indicates a possible seven percent contribution form secondary neutrons to the cross section of formation of Ni^{65} at 440 Mev.

A second argument for excluding large contributions from secondary neutrons is the asymmetry of the recoils. Bernardini *et al.*[8] have shown that the ratio of forward to backward black prongs (energy less than 30 Mev) in all stars in photographic plates induced by 375-Mev protons and 300-Mev neutrons is approximately two to one. This same asymmetry would then be expected for the secondary neutrons and for the recoil products from their (n,p) reactions. On the other hand, in the experiments reported here, the Ni^{65} recoil activity found in the forward aluminum foil was about seventeen times as great as in the backward one. We conclude, then, that secondary neutrons born in the target are not the main cause of Ni^{65}.

The general neutron background at the cyclotron is excluded both by the recoil behavior described above and by an experiment in which two parts of a copper

[6] Paul Kruger, Ph.D. thesis, University of Chicago, January, 1954 (unpublished).
[7] H. H. Hopkins, Phys. Rev. **77**, 717 (1950).
[8] Bernardini, Booth, and Lindenbaum, Phys. Rev. **85**, 826 (1952).

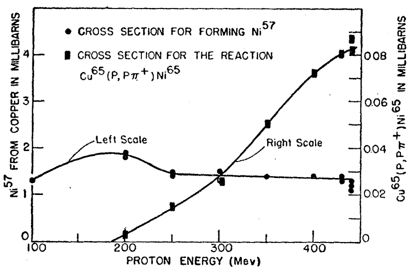

FIG. 1. The cross section for producing Ni^{57} and Ni^{65} in the proton bombardment of copper.

target were analyzed for Ni^{57} and Ni^{65} production. The first part was the leading edge of the target that received the main part of the proton beam. The other was a strip 1.4 cm further out radially. This was irradiated with only one-third the number of protons. The Ni^{65}/Ni^{57} ratio was 0.0247 in the leading edge and 0.0250 in the outer piece. Since it is not likely that any general neutron background is confined to the beam region, the constancy of this ratio precludes any important contribution to the formation of Ni^{65} by this mechanism.

The possibility that the Ni^{65} is caused by secondary particles other than neutrons is made unreasonable by the large cross section that would have to be assigned to reactions of usual character, e.g., $(d,2p)$, $(He^4,3pn)$, etc., and by the directional properties and the effect of thickness discussed above.

From the activation of thick and thin targets at 440 Mev (see above), about 7 percent of the Ni^{65} produced is ascribed to secondary neutrons. This is a 0.006-mb apparent cross section, close to that found at 100 Mev. We ascribe the Ni^{65} made at the low energy to this origin. The cross section at intermediate energies is corrected for the neutron effect by assuming 0.006 mb for this mode of production at all energies. The corrected values of the cross section for the reaction $Cu^{65}(p,p\pi^+)Ni^{65}$ are given in column 4 of Table I. These should be good to about $\pm 5\times 10^{-3}$ mb. These corrected values for forming Ni^{65} and the cross sections for forming Ni^{57} are plotted in Fig. 1.

DISCUSSION

The most reasonable interpretation of the curve in Fig. 1 giving the cross section for forming Ni^{65} is that it represents the reaction $Cu^{65}(p,p\pi^+)Ni^{65}$. The observed apparent threshold at around 200 Mev is consistent with this picture. For example, the production of π^+ at 90° in the proton bombardment of carbon has a very similar energy dependence (see Fig. 2). The recoil behavior of the Ni^{65} formed is likewise consistent in that it is similar to that of Ni^{57} which also requires the absorption of energy of the order of 100 Mev to extract the required particles from Cu^{63}.

The absolute value of the cross section appears to be reasonable by the following argument: The $Cu^{65}(p,p\pi^+)Ni^{65}$ process can be thought of as due to the reaction

$$p+p\rightarrow p+n+\pi^+$$

occurring inside the nucleus. If the final nucleus is to be Ni^{65}, the requirement exists that the resulting neutron acquire less than about 10-Mev kinetic energy and thus remain in the nucleus, and that the proton and meson get out without further interaction. This "meson-production scattering" inside the nucleus might then be compared to "elastic scattering" inside the nucleus. An example of this is the $Ni^{64}(p,n)Cu^{64}$ reaction being studied radiochemically by Koch and Turkevich.[9] This can be regarded as arising from the scattering process

$$p+n\rightarrow p+n$$

occurring inside the nucleus, with again a restriction on the final energy of the proton left in the nucleus. This reaction has a cross section of ~1 mb at 440 Mev.[9]

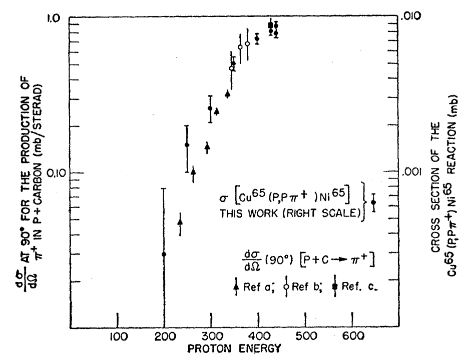

FIG. 2. Comparison of the energy dependence of the cross section of the $Cu^{65}(p,p\pi^+)Ni^{65}$ reaction with the energy dependence of the production of positive mesons in the proton bombardment of carbon at 90°. The references in the figures are as follows: (a) D. A. Hamlin, University of California Radiation Laboratory Report UCRL 2414, November 20, 1953 (unpublished); (b) Passman, Block, and Havens, Phys. Rev. **88**, 1247 (1952); (c) A. Rosenfeld, University of Chicago (private communication). (The numbers on the right-hand side of the diagram should read 0.10 and 0.01.)

The total production of π^+ in the 440-Mev proton bombardment of protons (leading to an unbound neutron and proton) is 1 to 2 mb.[10] The total (n,p) cross section at these energies is 34 mb.[11] Thus the ratio of the cross section of the $p(p,p\pi^+)n$ reaction to the (p,n) scattering cross section is about the ratio of the cross section for the $Cu^{65}(p,p\pi^+)Ni^{65}$ reaction to that of the $Ni^{64}(p,n)Cu^{64}$ process. We conclude that the absolute cross section observed is a reasonable one for the interpretation made.

ACKNOWLEDGMENT

The cooperation in this work of the directing and operating staff of the 170-inch synchrocyclotron is appreciatively acknowledged.

[9] R. C. Koch and A. Turkevich, University of Chicago (private communication).
[10] A. Rosenfeld, University of Chicago (private communication).
[11] V. A. Nedzel, Phys. Rev. **90**, 169 (1953).