Nuclear Reactions of Low-Z Elements with 5.7-Bev Protons*†

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This work describes the results of 5.7-Bev proton bombardments of the target elements Be, C, N, O, F, Na, and Al. Production cross sections were obtained for many radioactive products with half-lives between 1.2 minutes and 2.6 years. The (p,pn) cross sections for the targets C, N, O, F, and Na were found to be 29±3 mb, 7.3±0.7 mb, 33±5 mb, 19±2 mb, and 31±5 mb, respectively. Much of the variation in these values is thought to be due to the difference in the number of neutrons available for (p,pn) reactions in the different target nuclei. The cross sections for other types of reactions studied do not change as much over the above range of target elements as do the (p,pn) cross sections. Comparison of the cross sections measured in this work with those obtained at 0.98 to 3 Bev shows that in the 1- to 5.7-Bev energy range the excitation functions are nearly constant.

I. INTRODUCTION

THE understanding of the structure of the atomic nucleus over the past several years has proved so elusive that many different avenues of approach, both theoretical and experimental, have been developed. The theoretical approaches have led to the development of the statistical model, the optical model, the shell model, the collective model, etc.[1] The experimental approaches have included the study of the scattering of, and nuclear reactions initiated by, various particles incident on nuclei. Much work on nuclear reactions has been divided into the study of either the angular distribution of reaction products, or the total production cross section for different types of reactions. These studies are carried out with the types and energies of particles available on existing accelerators. As soon as an accelerator of higher maximum energy or different particle type is available, many of the experimental studies are repeated, where feasible, to see if a new type of reaction sets in or to investigate the energy dependence of the various processes studied.

This study is an example of the latter type of experiment. It was decided to repeat various measurements of spallation cross sections done at lower incident-proton energies (3 Bev and lower) at the Bevatron using a proton energy of 5.7 Bev. The target nuclei chosen were Be, C, N, O, F, Na, and Al.[2] These elements were chosen because of the relative simplicity of analysis of the bombarded targets (no chemistry was necessary). Also, by measuring the spallation cross sections for several neighboring elements, the dependence of the various reaction cross sections on the atomic weight of the target can be investigated. A simple analysis of the cross sections of the "simple" nuclear reactions, as exemplified by the (p,pn) reaction, will be presented here. In a succeeding paper a much more detailed treatment will be presented.

II. EXPERIMENTAL METHOD AND APPARATUS

A. Mounting of Target Material

The various targets were bombarded as foils or powders in the internal beam of the Bevatron. The foil stacks consisted of target foils, guard foils, and monitor foils all aligned and then taped together. Powders were bombarded in a Lucite powder holder, which is shown in Fig. 1. The hole in the back was used to attach this holder to the Bevatron pneumatic-plunger target holder. The 3-mil aluminum monitor and guard foils were cut to accurately fit the dimensions of the channel in the Lucite block and were taped in place. After the lid was taped on, the remaining space was filled with powder (which had been dried at 110°C and then kept in a desiccator), which was tamped to obtain as uniform a density as possible. The end of the holder was sealed with Scotch tape. Errors from such sources as nonuniform filling of the space with powder, etc., will be treated later.

For each bombardment the target foils and powder were arranged in order of increasing atomic weight, with the proton beam entering the lowest-atomic-weight side of the target packet. This was done so that contamination from spallation products recoiling out of the target foil and secondary neutron and

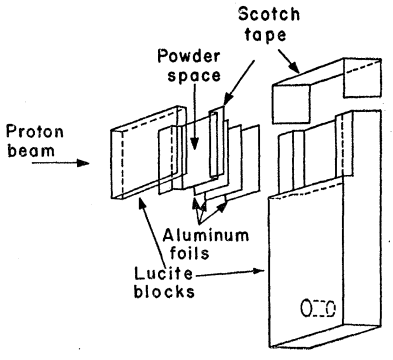

FIG. 1. Powder-target holder.

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[1] R. Peierls, Sci. American **200**, 75 (1959).
[2] The spallation reactions of some of these elements have been extensively studied. See for example references 13, 19, 20, and 32.

proton contributions to the beam could be held to a minimum. The majority of secondary products come out in the forward direction, and the amount of secondary neutrons and protons emitted per nuclear reaction increases with increasing atomic weight of the target nucleus.[3] Guard foils were used in front of and behind each type of experimental foil to keep recoiling spallation products of one target element from contaminating the adjacent target foil. The guard foils also served to protect the target foil from product-recoil loss, because as much product recoiled in from the guard foils as recoiled out of the target foil. Secondary neutrons and protons are not stopped in this manner, however. The best way to minimize formation of spallation products by secondaries is to keep the target thickness as low as possible. It has been shown that the secondary-neutron contribution can be kept to less than 10% if the target thickness is held below 2 to 3 g/cm^2.[4] The maximum target thickness used in these experiments was about 800 mg/cm^2. This included the thickness of the powder as well as the holder and other foils.

B. Sample Mounting

After bombardment, the target and monitor foils or powder were removed from the holder, weighed, mounted on aluminum cards and covered with Scotch tape. The sticky part of the tape over the powder was covered by another piece of tape. It was found that water pickup during weighing of the powders was negligible.

When the activity levels of short-lived isotopes, such as 1.2-min O^{14}, 2.0-min O^{15}, and 10-min N^{13}, were to be measured, the targets were mounted without weighing, and counting was begun immediately. The minimum time delay between the end of bombardment and the first count was 5 to 6 min. This time delay was sufficiently short to see the 1.2- and 2.1-min activities. The resolution of these activities is described in more detail later.

C. Counting Techniques

The sample and monitor foils were gamma counted with a 1- by 1½-inch sodium iodide (thallium-activated) crystal and phototube connected to a 50-channel pulse-height analyzer. Since most of the activities measured are positron emitters, the samples were covered on both sides with a sufficient thickness of steel to annihilate the positrons at the sample. Both the geometry correction (due to the point of origin of the 0.51-Mev gamma rays in the steel instead of the sample), and gamma-ray absorption in the steel were found to be negligible.

Decay curves were taken on the 0.51-Mev photopeak with a sufficient point density and over a sufficient length of time to resolve the desired activities in the sample being counted. The minimum time interval between the midpoint of successive count-rate determinations was 1.8 minutes. Even though this is longer than the O^{14} half-life, it was possible to resolve O^{14} activity from the decay curves in oxygen targets. The details of background subtraction, peak integration, dead-time correction and activity decay during counting are described elsewhere.[5]

D. Resolution of Product Activities

1. Aluminum

The target element with the most products which was bombarded in this work was aluminum. The decay curve of the 0.51-Mev peak contained contributions from Na^{22}, Be^7, Na^{24} (pair production by the high-energy gammas external to the crystal and capture of one of the annihilation gammas in the crystal) F^{18}, C^{11}, N^{13}, and (if the sample was counted soon enough after bombardment) O^{15} and Ne^{24}. Samples which were bombarded for only a few minutes in order to see O^{15}, N^{13}, or C^{11} did not have detectable amounts of Na^{22} or Be^7, so that the only "longlived" component was Na^{24}. The 0.51-Mev decay curve was resolved by first subtracting out the Na^{24} and F^{18} contributions. For the separation of 20.4-min C^{11} and 10-min N^{13},[6] the portion of the remaining decay curve taken more than 20 minutes after the end of bombardment was analyzed by a method given by Biller.[7] This method depends on the fact that at any time t after the end of bombardment, the total measured activity C is related to the two initial-component activities A_0 and B_0 by

$$Ce^{\lambda_1 t}=A_0+B_0e^{(\lambda_1-\lambda_2)t}. \quad (1)$$

Knowing C and t, we can plot $Ce^{\lambda_1 t}$ vs $e^{(\lambda_1-\lambda_2)t}$, which gives a straight line of slope B_0 and intercept A_0. By this method we obtain the initial N^{13} and C^{11} activities. Decay curves of these two activities were constructed and subtracted from the portion of the curve taken less than 20 minutes after bombardment. The remaining short half-life component ($t_{\frac{1}{2}}=2$ to 3 min) was taken to be a sum of 2.1-min O^{15} and 3-min Ne^{24}. The same method described above was used again to separate the two isotopes. The values obtained for O^{15} and Ne^{24} were not very accurate, because of all the previous subtractions that had been made. Also, these isotopes had decayed considerably before the counting was begun. Consequently, the count rates obtained have a larger error associated with them than do the values for the other isotopes.

The Na^{24} counting rate was determined by measuring

[3] G. Bernardini, E. T. Booth, and S. J. Lindenbaum, Phys. Rev. **85**, 826 (1952).

[4] L. A. Currie, W. F. Libby, and R. L. Wolfgang, Phys. Rev. **101**, 1557 (1956).

[5] Paul A. Benioff, thesis, University of California Radiation Laboratory Report, UCRL-8780, July, 1959 (unpublished).

[6] D. Strominger, J. M. Hollander, and G. T. Seaborg, Revs. Modern Phys. **30**, 585 (1958).

[7] William Biller, thesis, University of California Radiation Laboratory Report, UCRL-2067, January, 1953 (unpublished).

the decay of the 1.38-Mev photopeak and subtracting any 1.28-Mev contribution from Na^{22} decay. The Na^{22} and Be^{7} counting rates were found by measuring the activity levels in the 0.51-Mev and 1.28-Mev photopeaks. The contribution to the 0.51-Mev peak from Na^{22} was removed by use of the spectrum of a pure Na^{22} standard. The remaining activity decayed with the correct half-life for Be^{7}.

Several runs were examined for Mg^{27} ($t_{\frac{1}{2}}=9.5$ min)[6] by taking decay points of the activity level in a few channels centered about 0.85 Mev. The decay curve had a small component of approximately the right half-life. Because of the low count rate in this energy range, the values obtained for Mg^{27} are uncertain and have a larger error associated with them than do the values for the other isotopes.

2. Sodium

Sodium was bombarded as anhydrous Na_2CO_3 in the powder holder. The decay curve was analyzed in the same way as that for aluminum except that Na^{24} and Mg^{27} were absent.

3. Fluorine

A 30-mil Teflon foil was used as a fluorine target. The Teflon was analyzed spectrographically and found to contain less than 1 ppm of Na, Mg, A, Si, P, Ca, Fe, and Cu. It was assumed that these elements would be the major contaminants. The decay curve was analyzed in the same way as that for aluminum except that Mg^{27}, Na^{24}, Ne^{24}, and Na^{22} were absent.

4. Oxygen

Anhydrous oxalic acid was used as an oxygen target. A spectrographic analysis for the same elements as those looked for in Teflon yielded the same results; each element was not detectable or present at less than 1 ppm. For those runs in which the cross sections for short-lived activities were desired, the powder was placed on an aluminum card, covered with cellophane tape to prevent water pickup and sample loss, and immediately counted. The weighing of the powder was deferred until all the decay points needed were taken. The 1.2-min O^{14} and 2.0-min O^{15} activities were resolved from the 0.51-Mev gamma-decay curve by first subtracting out the C^{11} and N^{13} activities (the contributions from longer-lived components were quite negligible). The remaining four to seven points were resolved by the method already described for resolution of C^{11} and N^{13} activities.

5. Nitrogen

Anhydrous 5-amino tetrazole was used for a nitrogen target. A spectrographic analysis for the same elements as were determined for fluorine yielded the same limits of detection. The compound was obtained as the monohydrate, dried in an oven at 110°C, and then kept in a desiccator. The compound appeared to lose its water of hydration quite easily, because the crystals quickly changed to powder as heat was applied. This compound has one possible hazard; some of the tetrazole derivatives are explosives. It melts with decomposition at 203°C.[8] During and after bombardment in the Bevatron it seemed to behave perfectly and did not discolor. The compound has a high ratio of nitrogen to carbon and hydrogen. Its formula is CN_5H_3. Bombardment of this compound even for more than 1 hour produced no visible change. The decay-curve resolution was done in the same manner as for oxalic acid targets.

6. Carbon

Carbon was bombarded as foils of polyethylene. Thick foils (50 mg/cm^2) were used for Be^{7} cross-section determinations and thin foils (6 mg/cm^2) were used for C^{11} cross-section determinations. Again a spectrographic analysis of both foils for the elements Na, Mg, Al, Si, P, Ca, Fe, and Cu gave less than 1 ppm for any one of them (i.e., they were not detected). The decay-curve analysis was simple, because only Be^{7}, C^{11} and a minute amount of F^{18} were present.

7. Beryllium

A bombardment of a thick beryllium foil yielded two products, Be^{7} and C^{11}. Analysis of the decay curves was quite simple because only two components were present.

E. Counting Efficiencies

The values of the efficiency and geometry of the gamma counter as a function of sample-to-crystal distance were taken from Kalkstein and Hollander.[9] Values of the geometry for different sample-to-crystal distances were redetermined by Barr.[10] His results agreed to within 2% of the values of the previous work.[9]

F. Cross-Section Ratios

The disintegrations per minute of the various product activities were used to obtain ratios of the product cross sections to the cross section of the monitor reaction. For several of the activities, the experimentally determined ratio was a weighted average of the contributions from each element present in the target. Thus, in Na_2CO_3, C^{11} is produced from the sodium, carbon, and oxygen atoms present. The contribution from the carbon was corrected here by the use of the ratio determined from polyethylene targets. Similarly, the contribution from the oxygen was removed by the use of the ratio determined from oxalic acid targets.

[8] Frederic Benson, Chem. Revs. 41, 1 (1947).

[9] M. Kalkstein and J. Hollander, University of California Radiation Laboratory Report, UCRL-2764, October, 1954 (unpublished).

[10] Donald Barr, thesis, University of California Radiation Laboratory Report, UCRL-3793, May, 1957 (unpublished), and private communication.

The polyethylene ratio was used similarly to correct for the carbon content of oxalic acid.

III. RESULTS

In the manner described in the previous paragraphs, a series of ratios of production cross sections to the monitor cross section were obtained for each bombardment. Almost all of the ratios were determined more than once, and a few were determined many times.

A. Data Rejection

On examining the results, it was found that in some cases the group of determinations of a given cross section would have a reasonable spread except for one datum which was quite different. If this particular determination could not be rejected because of some known experimental error, a statistical rejection criterion was applied to the group to see if the outlying datum could be rejected.[11] The confidence level of the rejection is 90%. Out of all the cross-section ratios determined, only five had an outlying datum that could be rejected by this test.

B. Monitor Reaction Cross Section

For most spallation work, the cross section for the reaction $Al^{27}(p,3pn)Na^{24}$ has been used as a standard, and its excitation function is well-known. This reaction is chosen because of the convenient half-life (15-hr) and beta-decay characteristics. However, for this work, many of the irradiations were too short to use Na^{24}. Also, it was desired to use a source of annihilation radiation for direct comparison with the many positron emitters of interest in this study. For these reasons, F^{18} formed in the reaction $Al^{27}(p,X)F^{18}$ was chosen as a monitor. Use of this reaction as a monitor removes the counting-efficiency correction for many determinations, because F^{18} is a positron emitter (97% of the F^{18} decays are by positron and 3% by electron capture).[6] The counting-efficiency correction, however, must be made to determine the F^{18}/Na^{24} production-cross-section ratio, because of the high energy (1.38 Mev) of the Na^{24} gamma counted. The inclusion of this correction gave a value for the F^{18}/Na^{24} production-cross-section ratio of 0.732 ± 0.063 (this value is the average of fifteen measurements with the standard deviation associated with an individual measurement). The production cross section for the reaction $Al^{27}(p,3pn)Na^{24}$ for 5.7-Bev protons was taken to be 10.5 millibarns (mb).[10] Recent accurate work gives the cross sections for the above reaction as 10.4 ± 0.6 mb at $E_p=2.0$ Bev and 10.0 ± 0.6 mb at $E_p=3.0$ Bev.[12] These results combined with lower-energy work are stated to be consistent with a constant cross section of 10.7 ± 0.6 mb for a proton energy range of 0.3 to 3 Bev. A cross section of 29.8 ± 1.6 mb for the reaction $C^{12}(p,pn)C^{11}$ at 6.0 Bev has recently been determined.[13] In this work the ratio of the cross sections for the reactions, $C^{12}(p,pn)C^{11}$ and $Al^{27}(p,X)F^{18}$, $\sigma_{C^{11}}/\sigma_{F^{18}}$ was found to be 3.83. Combining this with the $\sigma_{F^{18}}/\sigma_{Na^{24}}$ ratio given above and the 10.5-mb production cross section for Na^{24} from aluminum gives the cross section for the reaction $C^{12}(p,pn)C^{11}$ equal to 29.4 ± 3.3 mb. This value agrees very well with the value of 29.8 ± 1.6 mb. The error on the value of 10.5 mb will be taken to be 0.6 mb in agreement with the results at lower energies. The standard deviation, taken here to be one-half the error limit, is then $\frac{1}{2}(0.6\text{ mb})=0.3$ mb.[14] The cross section for the monitor reaction $Al^{27}(p,X)F^{18}$ then turns out to be 7.68 ± 0.17 mb. The value 0.17 mb is the standard error of the mean[14] and does not include the 0.3 mb associated with the cross section for the reaction $Al^{27}(p,3pn)Na^{24}$. The error limits given below include this value of 0.3 mb and an estimate of systematic errors in this work.

C. Production Cross Sections

Table I contains the results of the 5.7-Bev proton bombardments. The two tritium determinations by Currie are included for the sake of completeness.[15] The first column lists the target elements studied. The reaction type for each product is given in column 2. The "X" appearing in some entries refers to any group of emitted particles that conserves charge and nucleon number. Cross sections were determined for all the radioactive products given in column 3. The fourth column gives the number of bombardments made to determine the cross-section ratio for the listed product. The superscript "a" denotes sets from which an outlying datum was rejected. In these cases, n excludes the rejected datum.

The fifth column lists the production cross sections for the reactions given. The cross sections are obtained from the experimental ratio and a cross section of 7.68 mb for the monitor reaction $Al^{27}(p,X)F^{18}$. The values marked with a superscript "d" are based directly on the Na^{24} production cross section.

The standard errors of the mean cross sections are given in column 6. For the results based on F^{18} as a monitor we have[14]

$$S_1=\langle\sigma\rangle\left[\left(\frac{S}{\langle\sigma/\sigma_0\rangle}\right)^2\frac{1}{n}+\left(\frac{0.17}{7.68}\right)^2\right]^{\frac{1}{2}}, \tag{2}$$

and for the few results based on Na^{24},

$$S_1=(10.5/\sqrt{n})S. \tag{3}$$

[11] W. J. Blaedel, V. W. Meloche, and J. A. Ramsey, J. Chem. Educ. **28**, 643 (1951); R. Dean and W. Dixon, Anal. Chem. **23**, 636 (1951).

[12] J. B. Cumming, G. Friedlander, and C. E. Swartz, Phys. Rev. **111**, 1386 (1958).

[13] N. Horwitz and J. Murray, University of California Radiation Laboratory Report, UCRL-8881, September, 1959 (unpublished).

[14] Robley Evans, *The Atomic Nucleus* (McGraw-Hill Book Company, Inc., New York, 1955), pp. 757–771.

[15] Lloyd A. Currie, Phys. Rev. **114**, 878 (1959).

TABLE I. Summary of experimental results.

Target	Reaction type	Product	Number of determinations, n	Cross section, (mb)	Standard error, S_1	Error limit, E_σ
Be	(impurities)	C^{11}	1	0.044	...	...
	$(p,p2n)$	Be^7	1	15	...	...
C	(secondary?)	F^{18}	2	0.01	0.0014	0.003
	(p,pn)	C^{11}	13	29	1	3
	$(p,pn\alpha)$	Be^7	5	11	0.6	1.5
	(p,X)	H^3	...	18[c]	...	...
N	(secondary?)	F^{18}	2	0.0040	0.0007	0.0014
	(p,pn)	N^{13}	3[a]	7.3	0.2	0.7
	$(p,2p2n)$	C^{11}	6	13	1.8	4
	(p,X)	Be^7	3[a]	14	0.8	2
O	(secondary)	F^{18}	2	0.06	0.014	0.03
	(p,pn)	O^{15}	3	33	2.2	5
	$(p,p2n)$	O^{14}	3	11	1.5	3
	$(p,2p2n)$	N^{13}	5	6	1.1	2
	$(p,pn\alpha)$	C^{11}	7	12	1.2	3
	(p,X)	Be^7	2	10	2.6	5
F	(p,pn)	F^{18}	10	19	0.6	2
	$(p,p2n\alpha)$	N^{13}	3	1.5	0.2	0.5
	(p,X)	C^{11}	3[a]	5.4	0.3	0.7
	(p,X)	Be^7	6	9.2	0.7	1.6
Na	(p,pn)	Na^{22}	2	31	2.0	5
	$(p,pn\alpha)$	F^{18}	2	10	0.4	1
	(p,X)	Be^7	2	13	1.1	2
Al	$(p,p\pi^+)$	Mg^{27}	2	0.1	0.07	0.1
	$(p,4p)$	Ne^{24}	2	1.6	0.6	1.2
	$(p,pn\alpha)$	Na^{22}	6[b]	17[d]	1.3	3
	(p,X)	F^{18}	15	7.68[d]	0.17	0.57
	(p,X)	O^{15}	2	4.5	1.8	3.6
	(p,X)	N^{13}	5[a]	1.7	0.2	0.3
	(p,X)	C^{11}	8	6.0	0.4	0.9
	(p,X)	Be^7	4[a]	8.3[d]	0.3	0.9
	(p,X)	H^3	...	50[c]	...	...

[a] An outlying datum has been rejected from this set.
[b] One of these determinations was done by M. Kalkstein.
[c] These values are from the work of Currie (reference 15).
[d] These values were obtained from cross section ratios σ/σ_0 in which σ_0 is the cross section for the reaction $Al^{27}(p,3pn)Na^{24}$, rather than the reaction $Al^{27}(p,X)F^{18}$.

The average experimental cross-section ratio is given by $\langle\sigma/\sigma_0\rangle$, and S is the standard deviation of individual measurements of σ/σ_0.[11,14] The number S_1 gives the contribution of all the random errors in this work. This includes such sources as errors in aligning target foils, nonuniform packing of the powder in the powder holder, random errors in resolving decay curves, etc. The calculation of standard errors for the cross section for a reaction product which can come from more than one source, e.g., the cross section for the reaction $O^{16}(p,X)Be^7$ in oxalic acid targets, is slightly more involved. Details of this calculation are given elsewhere.[5]

Column 7 gives the error limit, E_σ, associated with the given cross section. Besides the random errors, S_1, this includes the error in the $Al^{27}(p,3pn)Na^{24}$ monitor-reaction cross section and an estimate of systematic errors due to the possible experimental bias. This latter category includes such items as the small amount of water pickup in powder samples during weighing, the slight geometry effect of positron annihilation in the steel disks on top of the sample, systematic errors in the method used to obtain the integrated count rate under a gamma peak on the pulse-height analyzer, etc. Individually, each of these errors is less than 1% and they may work in opposite directions. Somewhat arbitrarily a fractional error limit of 6% has been taken to represent the contribution of these sources. Since the standard deviation is taken one half the error limit we get[14]

$$E_\sigma=2\sigma[(0.03)^2+(0.3/10.5)^2+(S_1/\sigma)^2]^{\frac{1}{2}}, \qquad (4)$$

as the error limit to be associated with a given cross section. For those cases which exclude an outlying datum, a 90% confidence limit is associated with S_1 and E_σ.

IV. DISCUSSION

A. Loss of Activity from Powder

In order to check if gaseous activities such as N^{13} were lost from the powder targets, a solid compressed organic target was bombarded. The target was made by subjecting a sample of semicarbazide (CN_3OH_5) to a pressure of 1100 kg/cm^2. Two disks were cut from the resulting hard cylinder, mounted on an aluminum foil, and bombarded with 5.7-Bev protons. The average N^{13} production cross section per 0.75 N-0.25 O atom was found to be 5.60 mb, and the average C^{11} production cross section per 0.2C-0.6N-0.2O atom was 15.2 mb. Use of the N^{13} and C^{11} production cross sections obtained from powder samples (Table I) gives values of 7.0 and 16 mb, respectively, for the N^{13} and C^{11} production cross sections per composite atom as given above. It is not known why the N^{13} cross section from the compressed target was lower than that obtained from the powder targets. A loss of gaseous activity from the powder would result in a higher cross section obtained from the compressed target. At least one can say that within experimental errors there appears to be no loss of gaseous N^{13} (and C^{11}) from powder targets. The same will be taken to hold for oxygen and fluorine activities.

B. Secondary Reactions

The cross section for the production of C^{11} from Be^9 contains contributions from the secondary reaction $Be^9(\alpha,2n)C^{11}$ and the primary reactions, $O^{16}(p,X)C^{11}$ and $C^{12}(p,pn)C^{11}$. Published values for the oxygen and carbon content in beryllium metal (hot-pressed from beryllium powder) are 0.65% and 0.06%, respectively.[16] Applying these values to the beryllium target used in this work and using the appropriate entries in Table I allows one to estimate a C^{11} production cross section of 0.093 mb from carbon and oxygen. This is more than the observed cross section of 0.044 mb, so one may conclude that the observed cross section probably consists mainly of contributions from impurities.

The cross sections of 10 ± 3 and 4 ± 1.4 microbarns (μb) for F^{18} production from 5.7-Bev-proton bombardment of polyethylene and 5-amino tetrazole, respectively, cannot be explained as being due only to

[16] D. White, Jr., and M. Burk, *The Metal Beryllium* (American Society for Metals, Cleveland, Ohio, 1955), p. 658.

impurities. If one assumes a 10-mb cross section for F^{18} production from the impurities, 500 ppm total impurities would be required. This is much more than was found. Crude estimates of cross sections for F^{18} production from carbon and nitrogen by the most probable secondary reactions are 0.04 μb and 0.1 μb, respectively. These numbers are also too small to account for the observed cross sections.

The foil stacks from which these cross sections were determined contained thick Teflon foils (170 mg/cm^2) which were separated from the polyethylene by a 5 mg/cm^2 polyethylene guard foil and from the 5-amino tetrazole by a 5 mg/cm^2 $(CH_2)_n$ guard foil and one layer of cellophane tape. It is possible that some of the F^{18} recoiled or migrated into the carbon or nitrogen target. The lower cross section for the nitrogen target, which had thicker guard foils, supports this possibility.

The oxygen target shows the first real evidence of F^{18} production by nuclear reactions, because the contributions from F^{18} recoils and migration should be the same as for the carbon and nitrogen targets, whereas the observed cross section is 60±30 μb. The principal reactions leading to F^{18} production from oxygen targets would be $O^{16}(\alpha,d)F^{18}$; $O^{16}(H^3,n)F^{18}$; $O^{16}(He^3,p)F^{18}$, and the primary reaction, $O^{18}(p,n)F^{18}$. The contribution from the $O^{18}(p,n)F^{18}$ reaction can be shown to be small. At 400 Mev, the cross section for the reaction $B^{11}(p,n)C^{11}$ is 1.5 mb,[17] and at a proton bombarding energy of 420 Mev, the F^{18}-production cross section from oxygen targets is 0.083 mb.[18] If the reaction $O^{18}(p,n)F^{18}$ is taken to be the main source of F^{18}, then the observed cross section at 420 Mev corrected for the abundance of O^{18} in oxygen,[6] becomes 40 mb. The difference between 40 mb and the $B^{11}(p,n)C^{11}$ reaction cross section at 400 to 420 Mev seems too great to ascribe to differences in the target element. Consequently, the bulk of the 60 μb probably comes from secondary reactions on O^{16} and, as a very rough guess, the $O^{18}(p,n)F^{18}$-reaction cross section will be taken to be 1.5 mb at 420 Mev. The same cross section will be assumed at 5.7-Bev bombarding energy. The value of 1.5 mb gives a contribution of 3 μb from the $O^{18}(p,n)F^{18}$ reaction in natural oxygen.

Rough upper-limit estimates of the contribution to the F^{18} production cross sections from the secondary reactions $O^{16}(t,n)F^{18}$; $O^{16}(He^3,p)F^{18}$; and $O^{16}(\alpha,d)F^{18}$ give values of 4 μb, 7 μb, and 3 μb, respectively.[5] Because the oxalic acid was bombarded under the same conditions in the stack (with respect to guard foils and position) as the 5-amino tetrazole, the maximum F^{18} contribution from possible recoil and migration will be taken to be 5 μb. The addition of all these contributions, which are thought to be upper limits, gives a total estimate of 22 μb. It is difficult to say whether this value is significantly lower than 60±30 μb as these values, including the error limit, almost overlap.

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C. (p,pn) Reactions

The most noticeable feature of the (p,pn)-reaction cross sections is their large variation for the different target elements. Bombardments by Burcham, Symonds, Warren, and Young with 980-Mev protons also show the same variation.[19] In this 980-Mev work, it is suggested that the variation may be correlated with the level structure of the product nuclide and that the deposition energy in the product nuclide must be less than the excitation energy of the first particle-emitting level. The same fluctuation of the (p,pn) cross section for different target elements is shown by recent work with 0.3- to 3-Bev protons.[20] This work also suggests a correlation of the (p,pn) cross section with the separation energies of the most loosely bound particles in the product nuclei and with nuclear shell structure. These suggestions can be used to explain much of the variation of the (p,pn)-reaction cross sections. At Bev bombarding energies most of the (p,pn) cross section comes from reactions in which the incident proton hits a neutron and the products of the (p-n) collision leave without interacting further. For the independent particle model, the excitation energy of the nucleus is equal to the hole-state excitation energy. One can then use the separation energy of the least-bound particle in the product to determine which of the uppermost neutron shells in the nucleus contribute to the (p,pn) reaction. Only those shells would be allowed that left the residual nucleus with insufficient energy to emit another nucleon. Consequently, it might be expected that, for a given energy and over a restricted atomic-weight range of targets, the total (p,pn) cross section divided by the number of "available" nucleons, where known, might be constant.[21] For some of these light elements, the excitation energy of nucleon holes in the "buried" shells can be determined fairly unambiguously from data in the literature.

A careful energy analysis of (p,$2p$) reactions on several low-Z elements with 185-Mev protons has demonstrated the scattering of protons from protons in the buried shells.[22] For C^{12} the $1p_{\frac{3}{2}}-1s_{\frac{1}{2}}$ proton-shell spacing was shown to be 16 Mev. This is more than the separation energy (7.5 Mev) of the least-bound particle (alpha) in C^{11}.[23] The Coulomb energy is small in these low-Z elements, so the neutron-shell spacings should be similar to those of the proton shells. This means that the $1s_{\frac{1}{2}}$ neutrons are unavailable, and only the four $1p_{\frac{3}{2}}$ neutrons are available for the (p,pn) reaction.

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TABLE II. (p,pn) cross sections per available neutron for low-Z elements.

Target	Product	$\sigma_{p,pn}$ (mb)	Available neutrons	(p,pn) cross section per available neutron, σ_i (mb)
C	C^{11}	29±3	4	7.2±0.8
N	N^{13}	7.3±0.7	1	7.3±0.7
O	O^{15}	33±5	6	5.5±0.8
F	F^{18}	19±2	(2)	10±1
Na	Na^{22}	31±5	(4)	8±1.0

The case of N^{14} is unambiguous because only the ground state of N^{13} is bound[23]; all the other levels have much larger particle-emission widths than gamma-emission widths. This means that only the $1p_{\frac{1}{2}}$ neutron of N^{14} can contribute. For N^{15}, the $1s_{\frac{1}{2}}$ and $1p_{\frac{3}{2}}$ proton-hole states are more than 15 Mev and 6 Mev above the ground state. Because N^{15} and O^{15} are mirror nuclei and the Coulomb energies are small, these excitation energies should be similar to those for the corresponding neutron-hole states in O^{15}. This means that in O^{16} the $1s_{\frac{1}{2}}$ neutrons are unavailable, but the $1p_{\frac{3}{2}}$ (and $1p_{\frac{1}{2}}$) neutrons are available because the binding energy of the least-bound particle (proton) in O^{15} is 7.3 Mev. The availability of the $1p_{\frac{3}{2}}$ level is supported by other studies of the O^{15} level structure.[24]

Determinations of the available neutron levels in F^{19} from the excited-state configurations of F^{18} and the binding energy of the least-bound particle (either an α particle, 4.41 Mev or a proton, 5.6 Mev)[23] is not as easy as in the previous cases because less is known about F^{18} level structure. However, isotopic spin, parity, and spin requirements can be used (if we neglect collective effects) to help decide if the known excited states can be "buried-shell" hole states. For example, the parent $1p_{\frac{1}{2}}$ neutron-hole F^{18} states of the ground state of F^{19} can have values of the isotopic spin, T, equal to 1 or 0, negative parity, and values of the spin, I, equal to 1 or 0. The probability of producing any one of these states with specific values of T and I is given by the appropriate fractional percentage coefficients. All the F^{18} excited states below and including the 5.60-Mev state have $T=0$ except two, which have $T=1$. However, these two, at 1.08 and 3.07 Mev, have positive parity.[23,25] Neither the isotopic spin nor the parity of the adjacent 5.67-Mev level are known. If the isotopic-spin selection rules hold, it probably has $T=0$ because it is formed by He^4 bombardment of N^{14}. The higher levels are known to be particle emitting.[23] The above data on the F^{18} excited states indicate that all the negative-parity parent $1p_{\frac{1}{2}}$ neutron-hole F^{18} states with $T=1$ are particle-emitting states. This means that the $1p_{\frac{1}{2}}$ neutrons are unavailable whenever a $T=1$ F^{18} parent state is produced. It is not possible to decide at present from the F^{18} level scheme whether the parent states with $T=0$ are particle-emitting or not. Consequently, the two $1p_{\frac{1}{2}}$ neutrons in F^{19} appear to be at least partially unavailable and may be completely unavailable. In order to have a number to work with, it will be assumed here that $1p_{\frac{1}{2}}$ neutrons are completely unavailable. The $1p_{\frac{3}{2}}$ and $1s_{\frac{1}{2}}$ neutrons should also be unavailable, because the $1p_{\frac{3}{2}}$ and $1s_{\frac{1}{2}}$ hole states would have even higher excitation energies than the $1p_{\frac{1}{2}}$ hole state.

Much less is known about Na^{22} levels. All the excited states below 3.5 Mev should have $T=0$ by the isotopic spin selection rules, because they are directly populated by the $Mg^{24}(d,\alpha)Na^{22}$ reaction.[26] No states have yet been found between 3.5 and 7.5 Mev (the proton binding energy in Na^{22} is 6.74 Mev).[26] The spin, parity, and emission width of the 7.5-Mev state are not given. In the absence of further information it will be assumed here that the $1p_{\frac{1}{2}}$ neutrons are unavailable.

The discussion in the previous paragraphs has shown that the number of available neutrons for (p,pn) reactions can be taken as four $1p_{\frac{3}{2}}$ neutrons for carbon, one $1p_{\frac{1}{2}}$ neutron for nitrogen, four $1p_{\frac{3}{2}}$ and two $1p_{\frac{1}{2}}$ neutrons for oxygen, two ($1d_{\frac{5}{2}}$?) neutrons for F^{19}, and four $1d_{\frac{5}{2}}$ neutrons for Na^{23}. Table II has been prepared from these numbers of available neutrons. Columns 1 and 2 list the target element and the product nuclide. The third and fourth columns list the (p,pn) reaction cross section in millibarns and the number of available neutrons. The tentative nature of the value given for the number of available neutrons for F^{19} and sodium is indicated by the parentheses. The fifth column gives the (p,pn)-reaction cross section per available neutron σ_i. It is immediately seen that σ_i is more nearly constant than is $\sigma_{p,pn}$, indicating that there is indeed some correlation between the number of available neutrons and $\sigma_{p,pn}$. The variation remaining outside of the error limits indicates that, as would be expected, σ_i varies with the atomic weight of the target and with the shell quantum numbers of the available neutrons. This comparison between σ_i and $\sigma_{p,pn}$ supports the idea that the (p,pn) reaction proceeds mainly by a p-n knock-on mechanism.

D. $(p,p2n)$ Reactions

Cross sections for only two examples of this type of reaction where determined. For the reactions $Be^9(p,p2n)$-Be^7 and $O^{16}(p,p2n)O^{14}$ the cross sections are 15 mb and 11±3 mb, respectively. This type of reaction is not so easy to interpret as the (p,pn) reaction, as one would expect two main contributing mechanisms. One of these would consist of the incident proton knocking out a neutron from a "buried shell," which leaves the residual

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nucleus in a sufficiently excited state to emit another neutron (the 1s shell in C^{12} is a possible example). The other mechanism consists of a proton knocking out a neutron available for the (p,pn) reaction, and then one of the collision products knocking out another neutron from shell. The residual nucleus must be left with insufficient energy to emit another particle. It is difficult to estimate the relative contribution of these mechanisms to observed cross sections.

E. $(p,2p2n)$ Reactions

Cross sections for two examples of this type of reaction were determined. The cross sections for the reactions $N^{14}(p,2p2n)C^{11}$ and $O^{16}(p,2p2n)N^{13}$ are 13 ± 4 mb and 6 ± 2 mb, respectively. The contributing mechanisms for this reaction are numerous. Besides one, two, or three successive collisions followed by emission of two, one, or no particles, respectively, from the excited nuclear state, deuterons can be emitted. The initial collision can be either with a neutron or a proton, etc. It appears from the above two cross sections that the fact that N^{13} has only one bound state, whereas C^{11} has several, is influencing the cross section. This can also be seen from a general study of the data in Table I, i.e., from any given target element the N^{13}-production cross sections are always less than those for C^{11} or O^{15}.

F. $(p,pn\alpha)$ Reactions

Cross sections for four examples of this type of reaction were determined. For the reactions $Al^{27}(p,pn\alpha)Na^{22}$, $Na^{23}(p,pn\alpha)F^{18}$, $O^{16}(p,pn\alpha)C^{11}$, and $C^{12}(p,pn\alpha)Be^{7}$, cross sections of 17 ± 3 mb, 10 ± 1 mb, 12 ± 3 mb, and 11 ± 1.5 mb, respectively, were found. Unlike those for the (p,pn) reactions, these values are all fairly uniform. There does not appear to be any correlation with the number of bound levels in the products, i.e., Be^{7} has two, C^{11} has seven or eight, F^{18} has ten or eleven, and Na^{22} has more than eleven.[23,25,26] Division of the total cross sections by the same number of available neutrons as was taken for the (p,pn) reactions gives cross section contributions per available neutron of 2.8 ± 0.5, 2.5 ± 0.3, 2.0 ± 0.5, and 2.8 ± 0.4 mb/neutron for aluminum, sodium, oxygen, and carbon targets, respectively. The six neutrons beyond the major shell closure in Al^{27} were taken to be available.[27] It is seen that these values are almost constant within the error limits. The constancy of these numbers is of doubtful use, however, because there are even more mechanisms leading to the final product than in the previous case. The writing of this reaction as a $(p,pn\alpha)$ reaction is not meant to imply that, after a proton-neutron collision, an alpha particle is emitted for all the reactions leading to the $(p,pn\alpha)$ product. The alpha may be emitted as single nucleons by knock-on collisions or deexcitation, or deuterons, tritons, or He^{3} may be emitted. Because of the large binding energy of the alpha-particle it is possible that the proton-neutron collision followed by excitation by the collision products of an alpha-emitting mode of the nucleus with or without any further nucleon-nucleon collisions is a likely mechanism.

There are several other types of reactions, each represented in Table I by one or two examples. Again the large number of pathways from target to product as well as the small number of examples for each reaction type precludes any definite conclusion about the likelihood of the possible mechanisms.

G. Comparison with Lower-Energy Data

Comparison of the 5.7-Bev results obtained here with cross sections measured from 980-Mev to 3-Bev bombarding energies reveal no striking changes.[12,13,19,20,28–32] The (p,pn) excitation functions all show a very slight or no decrease over a 1- to 5-Bev energy range. There may be a very slight minimum in the excitation functions at 2 to 3 Bev; however, the minimum is within the error limits on the various measured cross sections. The excitation functions for most other types of reactions such as $(p,2p2n)$; $(p,pn\alpha)$; etc., decrease with increasing energy in the 1- to 5.7-Bev energy range. However, the excitation functions for the reactions $Al^{27}(p,X)N^{13}$ and $Al^{27}(p,X)C^{11}$ increase slightly with increasing bombarding energy. N^{13} and C^{11} may be a sufficient number of mass units away from the target atomic mass that high-energy incident particles would be needed to cause the emission of the requisite number of nucleons either singly or in chunks.[32] Possibly the increase in the average number of mesons produced per nucleon-nucleon collision in the nucleus is important.

V. SUMMARY

In general the spallation cross sections for 5.7-Bev protons incident on low-Z elements are about equal to or slightly lower than those obtained on the same elements between 980 Mev and 3 Bev. A couple of the (p,pn) excitation functions may have a slight minimum in the 2- to 3-Bev energy range. Excitation functions for which the product is at least 14 atomic mass units away from the target show a rise with increasing energy in the Bev region.

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Most reactions yield cross sections whose values are fairly independent of the mass of the target. However, the values of the (p,pn)-reaction cross sections vary quite a bit as the target mass is changed. Much of this variation appears to be due to differences in the number of available neutrons in the various target nuclei (Table II). In another paper an attempt will be made to describe the magnitude and variation of the (p,pn)-reaction cross sections.

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Nuclear Structure and Simple Nuclear Reactions*†

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Recently it has become increasingly evident that some assumptions in the nuclear model used for the Monte Carlo calculations yield cross section values which are not in accord with experiment. In particular, calculations of (p,pn)-reaction cross sections in the Bev energy range give values which are low by factors of two to nine when compared to experimental values. The calculated cross sections also show a smooth variation with the target atomic weight whereas the experimental values show quite an erratic variation. Reasons which have been advanced to account for this lack of agreement are the lack of a nuclear surface and failure to account for shell effects in the nuclear model used.

In this work a theory is developed to take account of surface and shell effects and thereby describe the observed magnitude and variation of the cross sections for simple nuclear reactions as exemplified by the (p,pn) reaction. At multi-Bev energies to which this treatment is restricted, the main contribution to the (p,pn)-reaction cross section comes from inelastic collisions between the incident protons and target neutrons, with all the p-n collision products escaping without further interaction. Approximations and assumptions used include the impulse approximation, 0° lab scattering angle for the inelastic p-n collision products, classical trajectories for the incident and scattered particles, and a quantum-mechanical treatment for the target nucleons. The multi-Bev, n-p, cloud-chamber data was used to determine the average total exit cross section for the inelastically scattered particles. The only neutron shells in the target nucleus contributing to the (p,pn) reaction are those for which the instantaneous knocking out of a neutron creates a product-neutron hole state stable to particle emission. The combination of these assumptions gives integral expressions which, when evaluated on the IBM-701 computer for the independent particle harmonic-oscillator shell model, give the (p,pn) reaction cross sections as a function of the nuclear density distribution and the number of available shells.

For the low Z nuclei where the available shells can be unambiguously determined, the results give a half-central-density radius parameter, r_0, ($r_0=R_{\frac{1}{2}}/A^{\frac{1}{3}}$), of about 1.2 fermis compared to 1.03 fermis for the charge half radius from the electron-scattering work. Use of reasonable limits on the value of r_0 allows one to set the minimum number of shells available for some targets. For example, the Zn^{64}, Cu^{65}, and Cu^{63} (p,pn) cross sections require that a large part or all the $1f_{7/2}$ neutrons be available, or, equivalently, that a $1f_{7/2}$ neutron hole state (across a major shell) in the product nucleus have less than 8- to 9-Mev excitation energy. The results also show that the energy associated with nuclear rearrangement to particle-stable product states must be less than 8 to 9 Mev. In several cases, the upper limit can be lowered considerably (to 1.5 Mev and 0 Mev in the cases of O^{16} and N^{14}, respectively).

I. INTRODUCTION

IN recent years a large number of cross sections for various types of spallation reactions has accumulated.[1–7] The Monte Carlo method[8,9] coupled with a Fermi gas model of the nucleus has been used to interpret the experimental results. At medium and

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