

the interference terms between neighboring resonances at an energy somewhere *between* the two resonant energies. These interference terms would produce odd powers of x in the angular distribution if the two resonances have different parities, but only even powers if the parities are the same. Thus a measurement of the differential cross section at two angles symmetric about 90° could give some information. This scheme, however, is not very promising either, since if the two resonant-

state amplitudes differ in phase by an amount of the order of 90° , as it is conjectured, then these interference terms are likely to be very small and could be easily masked by contributions of the small phases.

It appears, therefore, that polarization measurements, although more difficult to carry out, provide much better information about the assignment of the resonances than do the simple differential cross-section experiments.

Some Cross Sections for the Production of Radio-Nuclides in the Bombardment of C, N, O, and Fe by Medium Energy Protons

MASATAKE HONDA AND DEVENDRA LAL*

Scripps Institution of Oceanography, and School of Science and Engineering, University of California, La Jolla, California

(Received November 9, 1959)

A number of nuclide formation cross sections have been measured, using the Berkeley 184-in. cyclotron, to assist in the interpretation of the data on cosmic-ray-produced nuclides in the atmosphere and in iron meteorites.

Cross sections of H^3 and Be^7 have been measured in bombardments of organic targets containing nitrogen and oxygen by protons of energy 225–730 Mev. Semicarbazide (CH_5N_3O) targets were used to obtain cross sections in air nuclei. The targets were prepared by mixing with a few percent of aluminum dust to permit reliable monitoring of the beam. Polyethylene, aluminum lactate, and Plexiglas targets provided elementary cross sections in carbon and oxygen.

The cross sections for the production of the long lived isotopes Cl^{36} (3×10^5 yr) and Al^{26} (8×10^5 yr) at 730 Mev, and of a number of short lived radionuclides at 500 and 730 Mev, in iron bombardment by protons have been measured. These data and those of earlier workers suggest some modifications in empirical relations used for predicting spallation cross sections in the case of nuclides close to stability.

I. INTRODUCTION

THIS paper describes the results of a selective study of cross sections for the production of some isotopes in the bombardment of carbon, nitrogen, oxygen, and iron by protons of energy between 225 and 730 Mev. The targets and the isotopes have been chosen with the aim of evaluating the data on cosmic-ray-produced isotopes in the atmosphere and in iron meteorites.

The natural rate of production of tritium in the atmosphere by cosmic rays have been calculated by Currie et al.¹ and Fireman and Rowland² using the formation cross sections of tritium in nitrogen and oxygen by protons of energy, $E \geq 450$ Mev. However, substantial isotope production in the atmosphere occurs due to nucleons of lower energy (Lal et al.³) and it becomes essential to know the cross sections at lower energies. Similarly, in the case of the isotope Be^7 , the only measurements in nitrogen and oxygen are those

by Benioff⁴ at a proton energy of 5.7 Bev. We have studied the cross sections for the formation of H^3 and Be^7 in bombardments of organic targets containing nitrogen and oxygen by protons of energy between 225 and 730 Mev. The targets were chosen to permit measurements of the average H^3 and Be^7 cross sections for a mixture of nitrogen and oxygen corresponding to that of the atmosphere. Elementary cross sections in carbon, and in some cases in nitrogen and oxygen, have been given.

In iron bombardments we have studied the cross sections for the production of long-lived isotopes, Al^{26} and Cl^{36} , and short-lived isotopes, Be^7 , Na^{22} , $Cl^{34m,38,39}$, $K^{42,43}$, and $Mn^{52,54}$ at proton energies 500 and 730 Mev. These measurements were made in order to interpret the results of Shedlovsky et al.⁵ and of Honda⁶ in this laboratory, who have studied cosmic-ray-produced radio-nuclides Be^{10} , Al^{26} , Cl^{36} , K^{40} , and Mn^{53} in some iron meteorites.

The energy range studied was limited by practical considerations, but it provides for the present a rea-

* On leave from the Tata Institute of Fundamental Research, Bombay.

¹ L. A. Currie, W. F. Libby, and R. L. Wolfgang, *Phys. Rev.* **101**, 1558 (1956).

² E. L. Fireman and F. S. Rowland, *Phys. Rev.* **97**, 780 (1955).

³ D. Lal, P. K. Malhotra, and B. Peters, *J. Atmospheric and Terrest Phys.* **12**, 306 (1958).

⁴ P. A. Benioff, *Phys. Rev.* **104**, 1122 (1956); Thesis, University of California, Radiation Laboratory Report (unpublished).

⁵ J. Shedlovsky, M. Honda, and J. R. Arnold (to be published).

⁶ M. Honda, *Geochim. et Cosmochim. Acta.* **17**, 148 (1959).

sonable basis for extrapolation to energy regions of importance in such studies. A detailed discussion of the production rates of the isotopes produced by cosmic rays in the atmosphere and in meteorites will be presented elsewhere.

II. TARGET MATERIALS AND IRRADIATIONS

a. Carbon, Nitrogen, and Oxygen Targets

In view of the difficulties in bombarding nitrogen and oxygen gases, we have chosen organic targets containing nitrogen and oxygen. For determining the contributions to isotope production in carbon, we have used polyethylene $(\text{CH}_2)_n$. Other targets chosen were semicarbazide $(\text{CH}_5\text{N}_3\text{O})$, Plexiglas $(\text{C}_5\text{O}_2\text{H}_8)_n$ ⁷ and aluminum lactate, $\text{Al}(\text{C}_3\text{H}_5\text{O}_3)_3$. Semicarbazide was considered to be a very suitable target since it contains nitrogen and oxygen in about the same proportions as in the atmosphere. For determining the cross section separately in oxygen, aluminum lactate formed the best choice in view of its relatively high content of oxygen.

The size of targets used was $1.2 \text{ cm} \times 0.7 \text{ cm} \times 0.7 \text{ cm}$ (within 15%). The targets were bombarded in the internal beam of the 184-in. Berkeley Cyclotron at different radial distances to correspond to energies 225, 300, 400, and 730 Mev. The beam intercepted the target perpendicular to the face of linear dimensions, $1.2 \text{ cm} \times 0.7 \text{ cm}$. Absolute cross sections were based on the monitor reaction $\text{Al}^{27}(p,3pn)\text{Na}^{24}$. In the case of polyethylene and Plexiglas, the targets were prepared by stacking thin sheets. Three aluminum foils (thickness 6.52 mg cm^{-2}) were included in the stack for monitoring the beam. The Na^{24} activity was measured in the central foil.⁸ The edges were then carefully machined so that the target and the monitor foils projected equal areas to the beam. Alignment of the monitor could not be done with confidence in the case of the semicarbazide target. In order to avoid this difficulty, which one always faces in the case of powder targets, we mixed the powder homogeneously with pure aluminum dust (3.47% by weight). Cakes of semicarbazide-aluminum mixture and aluminum lactate were then prepared in a hydraulic press. The cakes were properly shaped for the purpose of bombardments by filing on fine sandpaper.

In some of the targets, we placed a 1-mm thick target slice, facing the leading edge of the beam which could be easily removed at the end of bombardment. In the remaining targets a 1-mm slice was removed by scraping. This was considered necessary to prevent recoil losses, which are serious only in the case of

tritium. A semiquantitative analysis of the beam intensity as a function of distance from the leading edge, using the reaction $\text{Al}^{27}(p,3pn)\text{Na}^{24}$, showed that the intensity decreased exponentially with a half thickness of ca 1 mm.

The various targets were bombarded for time periods ranging from 10 to 40 min. The beam currents varied between approximately 0.01 and 0.2 microamperes. With the higher beam currents, the targets showed some signs of heating. Polyethylene and Plexiglas targets showed light yellowish-brown coloration at the edges. The tritium cross sections, however, were found to be the same as obtained with lower beam currents within the experimental errors of measurement. That is, we detected no loss of tritium with the beam intensities used, when the leading edge (1 mm) of the targets was removed.

b. Iron Targets

The iron targets were bombarded at 500- and 730-Mev proton energies in the internal beam of the cyclotron. The arrangement of the target was different in the case of short- and long-lived isotopes.

The targets for the measurement of short-lived activities were prepared by stacking thin sheets of iron foils and aluminum foils of thicknesses 19.8 and 6.52 mg cm^{-2} , respectively. The number of iron foils was three in two target assemblies and ten in one. The size of the stacks was $5 \text{ cm} \times 1 \text{ cm}$. The beam currents were of the order of 1 microampere and the duration of bombardment was 5–30 minutes.

For the measurement of cross sections of long lived activities, Al^{26} and Cl^{36} , we bombarded a sheet of cold rolled steel of dimensions $5 \text{ cm} \times 5 \text{ cm} \times 0.15 \text{ cm}$ in such a way that the beam intercepted the target approximately normal to the $5 \text{ cm} \times 0.15 \text{ cm}$ face. The beam current was ca 1 microampere and the exposure lasted for two hours. In order to estimate the path lengths of protons in the target, we measured the radioactivity of the target through a lead slit at intervals of 0.5 cm using a survey meter. It was found that the target was not aligned parallel to the beam trajectory and the average proton path in the target was ca 1.5 cm. Absolute cross sections were based on the cross section for production of K^{43} which was determined by us in shorter bombardments of iron foils using an Al monitor.

III. TARGET PROCESSING AND COUNTING

a. Organic Targets

The F^{18} and C^{11} activities were first allowed to decay. Usually the time of waiting was 24 to 36 hours. The Na^{24} activity in the aluminum monitor (central foil) and other organic targets containing aluminum was determined by a beta-gamma coincidence technique or by counting the 1.39-Mev gamma radiation in a 100-channel gamma spectrometer employing a $\text{NaI}(\text{Tl})$

⁷ Various trade names (Plexiglas, Crystalite, and Lucite) are used in industry for this type of material. The composition of the material used by us conformed to this formula. The analysis was carried out by Dr. E. Jeung and Dr. D. Sisson of the chemistry department of the University of California, Berkeley, to whom we are indebted.

⁸ G. Friedlander, J. Hudis, and R. L. Wolfgang, Phys. Rev. **99**, 263 (1955).

TABLE I. Measured Be⁷ and H³ cross sections. (Average cross section per atom in various organic targets in mb.) Numbers in parentheses below the cross section values give the number of independent measurements made. Mean cross sections are given.

Energy (Mev)	Polyethylene (C)		Semicarbazide (CN ₃ O) _{av}		Plexiglas (C ₅ O ₂) _{av}		Aluminum Lactate (CO) _{av}	
	Be ⁷	H ³	Be ⁷	H ³	Be ⁷	H ³	Be ⁷	H ³
225	8.2±0.6 (2)	7.0±1.1 (1)	8.8±0.3 (10)	13.7±0.9 (6)	9.0±0.5 (4)	8.7±1.0 (2)	8.2±0.4 (4)	...
300	10.3±0.7 (2)	7.0±0.8 (2)	11.9±1.2 (1)	10.5±1.6 (1)
400	9.6±0.7 (2)	8.6±1.0 (2)	10.7±0.8 (2)	11.2±0.8 (2)	22.2±3.5 (1)
730	11.6±1.2 (1)	7.6±1.2 (1)	10.9±0.8 (1)	33.5±5.3 (1)	10.5±0.7 (2)	...

crystal. The aluminum monitor activity was also checked every time on a methane flow proportional counter. The calibration of these instruments is described in Appendix A. The Be⁷ activity was then determined in the various targets by counting the 479-keV gamma radiation in the photopeak. The targets were then completely burned in a combustion furnace (quartz) and the resulting water was collected in a dry ice trap.

In some samples, from runs at low beam intensity, the tritium activity was counted by a liquid scintillation method⁹ (Packard Tri-carb.) Water was added to a phosphor solution of toluene-ethanol-dioxane-naphthalene, following Kinard¹⁰ except for the use of POPOP instead of α-NPO. Solution volumes were about 10 ml. The counting efficiency was determined in each run using a tritium standard supplied by the National Bureau of Standards (stated accuracy ±1.5%). In most runs the efficiency was 12–14% in a 10–80 v channel.

In the remaining samples where the tritium activity was high, its activity was measured following the technique of Currie et al.¹ The water collected after com-

bustion was vacuum distilled over zinc maintained at 415°C. The evolved hydrogen was then passed through a red hot palladium thimble and put in a gas counter having a volume of 190 ml. The counter was then filled to atmospheric pressure with methane and operated in the proportional region. The partial pressure of hydrogen in the counters was kept between 3 and 5 cm Hg. In this range, the plateau and the background counting rates were found to be reproducible. Since sufficient quantity of hydrogen was available from the targets, we could check on the reliability of the counting data by filling additional counters. Results of duplicate fillings in three cases agreed within the statistical errors of counting.

The calibration of the counters used in this work has been described by González-Vidal.¹¹

b. Iron Targets

The chemical procedures used for the extraction of various activities are described in Appendix B. In the case of short-lived activities the samples were deposited on a 2.3 cm² circular filter disk by the filter chimney method. The disks were then mounted on standard aluminum supports and covered with a thin Mylar film, 1 mg cm⁻². The weak activities of Al²⁶ and Cl³⁶ were counted on a low level beta-counter¹² (background 0.2 counts/min).

TABLE II. Measured cross sections in iron (mb).

Product nuclide	Half-life	730 Mev ^a	500 Mev	340 Mev Rudstam ^b
Mn ⁵⁴	291 days	33	28	12
Mn ⁵²	5.6 days	13	16	12.9
K ⁴³	22.4 hr	0.88 (1.0)	0.59	0.4
K ⁴²	12.52 hr	3.2 (2.5)	2.2	0.7
Cl ³⁹	55.5 min	0.21 (0.32)	0.12	0.045
Cl ³⁸	37.3 min	0.83	0.47	0.17
Cl ³⁶	3.1×10 ⁵ yr	6.8
Cl ^{34m}	32.4 min	0.59	0.38	0.11
Cl ^{34m} +Cl ³⁸	...	1.42 (1.6)	0.86	...
Al ²⁶	8×10 ⁵ yr	0.47
Na ²²	2.58 yr	0.36	...	0.02
Be ⁷	53 days	2.9

^a Data in parentheses were obtained from an iron target (44 mg cm⁻²) placed behind one of the thick targets, and monitored by an aluminum foil
^b G. Rudstam, P. C. Stevenson, R. L. Folger, Phys. Rev. **87**, 358 (1952)

Note added in proof.—A careful analysis of the decay scheme of Al²⁶ by Rightmire et al., Z. Naturforsch. **13a**, 847 (1958) yields for its half-life a value of 7.38×10⁵ yr (84.6% β⁺). The corresponding cross section of Al²⁶ in iron at 730 Mev is 0.43 mb.

⁹ See, e.g., J. R. Arnold, Science **119**, 155 (1954).

¹⁰ F. E. Kinard, Rev. Sci. Instr. **28**, 293 (1957).

IV. RESULTS AND DISCUSSION

The measured cross sections for the formation of Be⁷ and H³ in organic targets and for the various radio-nuclides in iron targets, are summarized in Tables I and II, respectively. These data are based on values of cross section for the monitor reaction, Al²⁷(p,3pn)Na²⁴, of 9.3 mb at 225 Mev, and 11 mb at 300–730 Mev (for discussion, see Benioff⁴). Corrections for the production of Be⁷ and H³ in aluminum present in the targets were found to be negligible. Approximate Be⁷ and H³ cross sections in aluminum at our bombard-

¹¹ J. González-Vidal, thesis, University of California Radiation Laboratory Report UCRL-8330, Berkeley, 1958 (unpublished). We are indebted to Dr. González-Vidal for allowing us to use the tritium proportional counters.

¹² T. T. Sugihara, R. L. Wolfgang, and W. F. Libby, Rev. Sci. Instr. **24**, 511 (1953).

ment energies were inferred from the data of Baker et al.¹³ and Currie.¹⁴ The target thicknesses used were less than 1 g cm^{-2} in all experiments except in the case of iron targets used for the measurement of cross sections for Na^{22} , Al^{26} , and Cl^{36} . The average proton track length in these experiments was ca 1.5 cm (ca 12 g cm^{-2}). Isotope production due to low-energy secondary neutrons and protons produced in the targets can be estimated from the data on the number of secondary particles produced in nuclear emulsions by particles of different energies (Camerini et al.¹⁵). We find that secondary corrections amount to less than 2% in most targets and are of the order of 10% in the thick iron targets. However, in this case we used K^{43} as the internal monitor and the cross sections of Na^{22} , Al^{26} , and Cl^{36} are not expected to be modified to any appreciable extent. (Table II.)

Uncertainties in cross sections due to loss of radio-nuclides escaping the targets because of their high velocities of emission are unimportant except in the case of tritium. We have experimentally checked this in the case of Be^7 , where we have measured its cross section in the 1 mm outer disks of organic targets. (These agreed with those obtained for the rest of the target within the experimental errors of measurements.) We have attempted to evaluate the upper limit of the fraction of tritons escaping the targets by using the energy spectrum of tritons of 2–30 Mev as measured by Bailey¹⁶ who has studied tritons produced in the bombardment of carbon by 190-Mev protons. For our targets ($0.7\text{--}1.0 \text{ g cm}^{-2}$) we estimate that the loss of tritons of energy less than 30 Mev is less than 5%. (The losses would have been much greater if we had not removed the edge 1 mm thickness from the targets.) Currie et al.¹ have measured tritium cross section in various targets as a function of depth. They find that most tritons produced in the bombardment of targets between carbon and lead by protons of 450 and 2200 Mev have energies less than 15 Mev (range $<120 \text{ mg cm}^{-2}$) and corrections in targets of 1 g cm^{-2} thickness are negligible. Loss of tritons is probably slightly over-compensated in our experiments due to a greater number of tritons recoiled in from the leading edge than those lost from the target, as the beam intensity decreased exponentially inwards with a half thickness of ca 1 mm.

There exists a possibility of underestimating tritium cross sections due to escape of tritons which have energies in excess of 30 Mev, for which the efficiency of retention by the target is poor. If we make the assumption that the proportion of tritons among protons

does not vary much with energy, an analysis of Bailey's data in 190-Mev bombardments of carbon shows that tritons of energy greater than 30 Mev constitute less than 18% of those below 30 Mev. In the results presented in Table I, this correction has, however, not been made in view of the uncertainty in this calculation.

a. Be^7

As seen from Table I, Be^7 cross sections are based on at least two determinations in most cases. The statistical errors can be evaluated from the ten determinations in semicarbazide at 225 Mev. The standard deviation of a single determination is found to be $\pm 10\%$. Individual cross section values in multiple determinations at a given energy agreed within 20%, thus supporting the estimated errors in cross section values. The estimated statistical errors are given in Table I.

The only measurement reported in the range of our measurement is by Marquez and Perlman,¹⁷ who have studied its cross section in the bombardment of carbon by 335-Mev protons. Their value of 12 mb has been multiplied by 36/41 for later revision of their monitor cross section,⁴ $\text{C}^{12}(p, pn)\text{C}^{11}$, and further reduced by 1.2 mb since they used a value of 10% for the branching ratio of 479-kev gamma ray. We have used the value of 11% which has come to be more commonly used in the literature. The corrected value of 9.5 mb is in excellent agreement with our results (Table I). The excitation function of Be^7 in carbon based on the data of Dickson and Randle¹⁸ up to 156 Mev, of Baker et al.¹³ at 1.0, 1.8, 2.2, and 3 Bev, and of Benioff⁴ at 5.7 Bev and present measurements, is given in Fig. 1. The values of Dickson and Randle¹⁸ have been raised by 11.2% since they have used a branching ratio of 12.3%.

The elementary cross sections in nitrogen and oxygen can be calculated from the data in Table I. They are found to be equal to that for carbon over the entire energy range studied within 10%. The Be^7 cross section in all cases seem to increase with energy between 225 and 730 Mev. This is probably due to an increasing contribution to Be^7 formation by direct knock-on processes.

b. H^3

The errors in tritium cross sections are found to be larger than in the case of Be^7 . The standard deviation of a single determination, based on the six independent cross-section values obtained for semicarbazide target at 225 Mev, is found to be $\pm 16\%$. The estimated statistical errors are given in Table I.

The excitation function of tritium in carbon between 225 Mev and 6 Bev is given in Fig. 2. Our value of $8.6 \pm 1 \text{ mb}$ at 400 Mev is in good agreement with 7.3 mb obtained by Currie et al.¹ at 450 Mev.

¹⁷ L. Marquez and I. Perlman, Phys. Rev. **81**, 953 (1951).

¹⁸ J. M. Dickson and I. C. Randle, Proc. Phys. Soc. (London) **A64**, 902 (1951).

¹³ E. Baker, G. Friedlander, and J. Hudis, Phys. Rev. **112**, 1319 (1958).

¹⁴ L. A. Currie, Phys. Rev. **114**, 878 (1959).

¹⁵ U. Camerini, W. O. Lock, and D. H. Perkins, *Progress in Cosmic-Ray Physics*, edited by J. G. Wilson (North Holland Publishing Company, Amsterdam, 1957), Vol. 1.

¹⁶ L. E. Bailey, thesis, University of California Radiation Laboratory Report UCRL-3334, 1956 (unpublished).

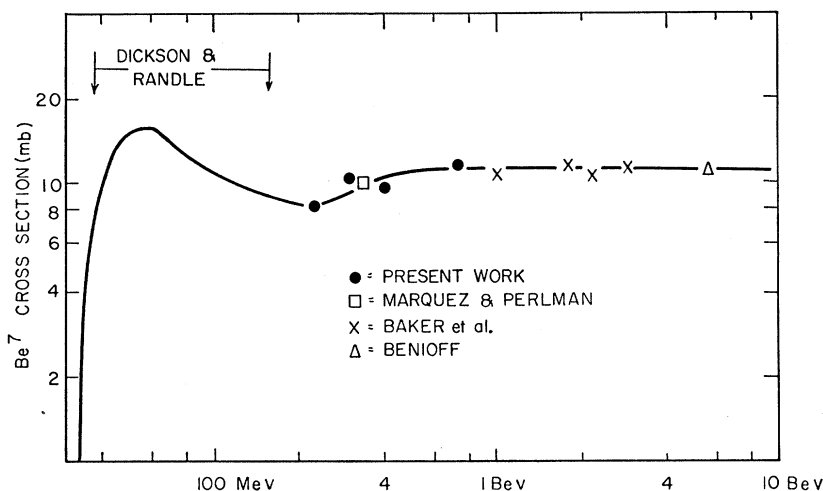


FIG. 1. Excitation function for Be^7 production in carbon.

We have calculated the average H^3 cross section per atom for a mixture of 3 nitrogen and 1 oxygen atoms using the measured cross sections in carbon and semi-carbazide. This gives us the tritium cross section for an average atom in the atmosphere. The H^3 excitation function for air is shown in Fig. 2. The calculated points at 0.45 and 2.05 Bev (Currie et al.¹), 2.2 Bev (Fireman and Rowland²) and 6 Bev (Currie¹⁴) have also been included.

The elementary cross sections in nitrogen and oxygen can be obtained at a few energies from Table I. The cross section in nitrogen is 16 ± 2 mb at 225 Mev. The cross sections in oxygen at 225 Mev, 300 Mev, and 400 Mev are found to be 13 ± 4.5 , 19 ± 6 and 36 ± 7 mb, respectively. The value at 400 Mev is in fair agreement with the value of 28 mb reported by Currie et al.¹ at 450 Mev. Unfortunately our data do not yield elementary H^3 cross sections in nitrogen and oxygen at 730 Mev and therefore we have not drawn the excitation functions separately for these elements. However, it seems that the excitation functions in nitrogen and oxygen agree with that for air within 15%, for energies ≥ 225 Mev.

c. Cross Sections in Iron Bombardments

The various measured cross sections in iron bombardments at 500 and 730 Mev are summarized in Table II.

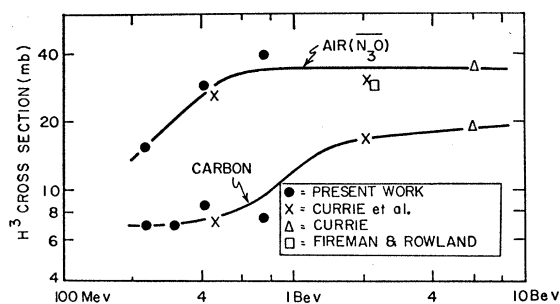


FIG. 2. Excitation functions for H^3 production in carbon and air.

The statistical errors in cross sections are estimated to be about 20% for the nuclides, Cl^{34m} and Al^{26} . In the remaining cases, the errors are smaller and expected to lie between 10 and 15%. In Table II, we have also listed the cross sections in iron at 340 Mev as measured by Rudstam.¹⁹ The production cross sections for nuclides of mass number less than 44 are seen to be increasing rapidly with energy, the slope being steeper for lighter mass numbers in this energy region. However, the cross section ratios for isotopes, e.g., $\text{Cl}^{38}/\text{Cl}^{39}$, $\text{K}^{42}/\text{K}^{43}$, are nearly constant at different energies. These facts can be used to estimate the cross section of the long-lived isotopes, Cl^{36} and Al^{26} , at other energies where their cross sections are not measured. No cross-section measurements of radio-nuclides of chlorine, potassium and sodium exist in iron in the Bev region and estimates of cross section in this region could be based on measurements in copper.

An empirical relation has been developed by Rudstam²⁰ for the prediction of formation cross sections, $\sigma(A, Z)$, of nuclides in a particular spallation group for targets of mass lying between 50 and 75:

$$\sigma(A, Z) = \exp[PA - Q - R(Z - SA)^2]. \quad (1)$$

Reference is made to Rudstam²⁰ for discussion of parameters, P , Q , R , and S . This relation has been shown to predict cross sections of various nuclides within a factor of two in general. A study of the available cross sections in iron bombardments showed that if we confine ourselves to product nuclides with mass numbers between 20 and 45 with values of $|Z - SA| \leq 1$, a relation of type 1 can be fitted to agree within ca 30% with the measured values. Most of the cosmic-ray-produced nuclides studied in iron meteorites fall in this criterion.

The parameters can be chosen in several different ways since the data are few: we find that the simplest

¹⁹ G. Rudstam, P. C. Stevenson, and R. L. Folger, Phys. Rev. 87, 358 (1952).

²⁰ G. Rudstam, thesis, Uppsala, 1956 (unpublished).

way is to use the values of P ²¹ and S as given by Rudstam²² and determine the values of R and Q . The value of R which fits the measured values best is found to be 1.9 in the case of iron bombardments studied so far (340–730 Mev). This can be seen from the yield distribution curve, Fig. 3, where we have plotted $\ln\sigma - PA + Q$ vs $(Z-SA)$. The measurements of Schaeffer and Zähringer²³ of cross sections²⁴ of argon isotopes, $A^{36,37,38,39}$, in iron at 430 Mev have been included. The values of Q based on $R=1.9$ are 9.39, 6.41, 6.14, and 4.06 at 340, 430, 500, and 730 Mev, respectively.

The yield distribution curve based on the value of $R=1.37$, which was found by Rudstam²⁰ to give the best fit for all product nuclides in iron spallations at 340 Mev, is shown by the dotted line in Fig. 3. Whereas this is indeed the best value for representing cross sections for nuclides over a wide range, it leads to a serious underestimation of cross sections of nuclides of mass 20–45 with $|Z-SA| < 1$. A similar situation is observed in the spallation of V, Mn, Co, and Cu at various energies^{19–21,25}; the cross sections of nuclides

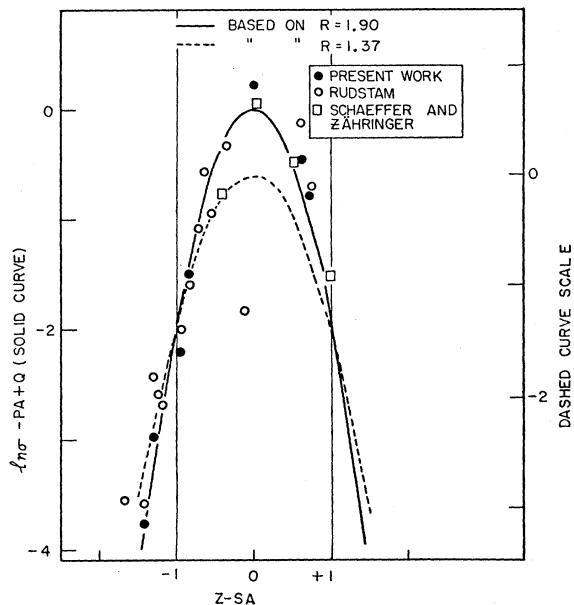


FIG. 3. The yield distribution of nuclides produced in iron spallations.

²¹ D. W. Barr, thesis, University of California Radiation Laboratory Report UCRL-3793, 1957 (unpublished).

²² The value of S used is 0.472, P is found to be independent of target mass and only energy dependent. It can be expressed by the relation, $P=10.2 E^{-0.63}$ for $100 \text{ Mev} < E < 5.7 \text{ Bev}$. E is expressed in Mev. The value of P at 5.7 Bev was measured by Barr (see reference 21).

²³ O. A. Schaeffer and J. Zähringer, Z. Naturforsch. **13a**, 346 (1958); Phys. Rev. **11**, 674 (1959).

²⁴ A^{38} and A^{39} cross sections have been reduced by 0.6 and 0.1 mb to take into account the contribution from decay of Cl^{38} , K^{38} , and Cl^{39} , respectively.

²⁵ G. Friedlander, J. M. Miller, R. Wolfgang, J. Hudis, and E. Baker, Phys. Rev. **94**, 727 (1954).

close to stability, e.g., P^{32} , Na^{24} , Na^{22} , Ti^{45} , are underestimated by 50–100% when the value of R used is that obtained from a best fit for all nuclides.

Summarizing, the formation cross sections of nuclides in iron spallations for nuclides which fall in the range, mass number 20–45, $|Z-SA| \leq 1$, can be described fairly accurately ($\epsilon=0.3$)²⁶ by relation 1, with $S=0.472$, $R=1.9$, and $P=10.2 E^{-0.63}$. The values of Q have been given for 340–730 Mev. At other energies it can be calculated by equating the total isobaric cross sections to the inelastic cross section for the target, σ_t .

$$e^{-Q} \cong P\sigma_t(R/\pi)^{\frac{1}{2}} [\exp(PA_t) - \exp(PA_t/2)]^{-1}. \quad (2)$$

A_t is mass of the target.

ACKNOWLEDGMENTS

This research was supported by Air Force Geophysical Research Directorate. We are grateful to Dr. I. Perlman for the excellent working facilities at the Radiation Laboratory, Berkeley. We are indebted to Dr. L. Winsberg, Dr. P. Benioff, Dr. S. Freed, Dr. J. Alexander, and Dr. H. Craig for helpful discussions, and to Dr. J. R. Arnold for discussions and assistance with the liquid scintillation counting of tritium. We are also indebted to Mr. J. Vale for arranging the cyclotron irradiations.

APPENDIX A. CALIBRATION OF COUNTING INSTRUMENTS

The calibration of the gamma spectrometers and the methane proportional beta counter was done by two independent methods: (1) Two I^{131} sources were prepared on thin zapon film and calibrated²⁷ on a 4π beta counter. The sources were mounted on the standard aluminum supports used and counted for their gross beta activity, and the 364 and 284 keV gamma radiations in the photopeak, under the conditions employed for the sample counting. This provided an absolute calibration for the mixed beta radiation of I^{131} (87.2%, $E_{max} 0.61$ Mev), and the photopeak efficiency for ~ 360 -keV gamma radiation. Absolute photopeak efficiencies for other gamma energies were then calculated using the relative intrinsic photopeak efficiencies measured by Lazar et al.²⁸ for the two NaI(Tl) crystals used (beveled 3 in. \times 3 in. and cylindrical 1 in. \times 1 in.).

(2) The absolute activity of an artificial Na^{24} source prepared by bombarding an aluminum foil (6.52 mg cm^{-2}) was determined by the beta-gamma coincidence technique. Its beta activity and the counts in the 1.39-Mev photopeak was then measured under conditions similar to that employed for sample counting. This provided an absolute calibration for 1.4 Mev and the

²⁶ ϵ is the root mean square error of the logarithm of the predicted and observed cross sections.

²⁷ We are grateful to Mr. M. Blaine for doing most of the calibration work.

²⁸ N. H. Lazar, R. C. Davis, and P. R. Bell, Nucleonics **14**, 52 (1956).

photopeak efficiency for the 1.39-Mev gamma radiation.

The counting efficiencies obtained by the two methods agreed within 4%.

APPENDIX B. CHEMICAL SEPARATION AND COUNTING OF RADIO-NUCLIDES FROM IRON TARGETS

1. Short Lived Activities

Chlorine

The iron foil was brought into solution by the addition of dilute sulphuric acid (1:3) (or in 1M CuSO₄ solution), and treated with 3M HNO₃; 17 mg chlorine carrier was added. Silver chloride was precipitated with an excess of AgNO₃, filtered, dissolved in NH₄OH, and reprecipitated using excess nitric acid. The precipitates were dissolved in NH₄OH and the solution passed through 5 ml of iron-hydroxide-dispersed cation exchange resin column (Dowex 50) in the ammonium form (Merrill et al.²⁹). Silver chloride was precipitated from the effluent, deposited on a filter disk, washed with alcohol, dried, and counted. The chemical yields were greater than 90%.

The samples were counted on a beta counter and a gamma spectrometer over a period of about 6 hr. Cl³⁹ and Cl^{34m}+Cl³⁸ activities were separately determined from the decay curve of gross β activity. Cl³⁹ activity was also determined from the counts in the photopeak at 246 keV (44.7%). The gross decay curve of the counts in the photopeak around 1.6 MeV was analyzed using the Biller plot³⁰ to resolve the counting due to the 1.6-Mev gamma of Cl³⁸ (31%) and 1.52-Mev gamma of Cl³⁹ (41.3%). Thus Cl³⁹ activity could be determined in three ways and Cl³⁸ in one way. The Cl^{34m} activity was determined by subtracting Cl³⁸ from the total Cl^{34m}+Cl³⁸ activity measured by beta counting.

Potassium

Four mg potassium carrier was added. The filtrate from the first precipitation of AgCl was treated with HCl to remove excess Ag⁺. Iron (III) was extracted with isopropyl ether from the 8M HCl solution of the sample, and excess acid was evaporated from the aqueous layer. A small excess of NaOH was added and precipitates were removed. The potassium salt was then precipitated by the addition of sodium tetra-phenylboron (Flaschka³¹) in the presence of formalin and Mg-EDTA. The precipitates were collected on a glass filter and passed through the filter by dissolving in acetone. The filtrate was warmed and acetone was evaporated by an air stream. The white crystals were deposited on a filter disk and dried for counting. The

²⁹ J. Merrill, M. Honda, and J. R. Arnold, Anal. Chem. (in press)

³⁰ W. Biller, thesis, University of California Radiation Laboratory Report UCRL-2067, 1953 (unpublished); A. C. Wahl, Phys. Rev. **99**, 730 (1955).

³¹ H. Flaschka, Z. anal. Chem. **136**, 99 (1952).

chemical yields (70–90%) were determined gravimetrically. The activity of the potassium sample was followed on a beta counter for 10 days and the contributions of K⁴² and K⁴³ were determined by the Biller's plot method.

Beryllium

The chemical procedure was similar to the one used by Baker et al.¹³ After an anion exchange and an acetylacetone extraction, the beryllium oxide was obtained, and counted in the gamma spectrometer for the 479-keV gamma ray in the photopeak. The chemical yield was 81% as determined gravimetrically.

Sodium

One mole of NaOH was added as carrier. After an iron extraction step by isopropyl ether the solution was made to 10M HCl and passed through an anion exchange column. Fe(OH)₃ scavenging was performed on the effluent. NaCl was then precipitated from the solution by bubbling HCl gas in the water-acetone (1:1) solution (Barr²¹). The chemical yield was determined to be 70% by the Mohr's volumetric method. The Na²² activity was determined by counting on a gamma spectrometer in the 1.28-Mev peak and also by β counting.

Manganese

Six mg of manganese carrier was added along with holdback carriers of Co and Cr. Mn(OH)₄ was repeatedly precipitated in acidic (HCl) and alkaline (NH₄OH) solution by the addition of NaClO₃ (Barr²¹) and H₂O₂, respectively. The chemical yields were 60 and 90% as measured by the EDTA titration method.

The Mn⁵² activity was determined by counting the 0.73-Mev gamma ray in the photopeak. The sample was found to have V⁴⁸ impurity. The Mn⁵⁴ activity was counted after a period of about 3 months in the photopeak of 0.85 MeV.

2. Long Lived Activities, Al²⁶ and Cl³⁶

Cl³⁶

The thick iron target was dissolved in dilute H₂SO₄ in a flask with a reflux condenser after adding 35 mg of chloride carrier and a crystal of copper sulphate. AgCl was repeatedly precipitated as described earlier. The ammoniacal solution of AgCl was passed through a 10 ml of a cation exchange column in NH₄R form to remove Ag(NH₃)₂⁺. The filtrate was concentrated and passed through 7 ml of HR form cation exchange column. The filtrate (HCl) was evaporated after the addition of NH₄OH (excess) to obtain NH₄Cl. Pure NH₄Cl was obtained by sublimation. The sample was still contaminated to a small extent. The chloride salt was dissolved in water, passed through a cation exchange column and 34 mg of NH₄Cl was recovered by

the sublimation process. Its beta activity was measured on a low-level counter (this sample is designated Cl-1).

A second purification of the sample was carried out by passing through a 15 ml anion exchange column (Dowex 1) in nitrate form. The column was eluted by 0.5M NaNO₃ solution acidified to pH=3 (Rieman and Lindenbaum²²). The elution curve of Cl⁻ was obtained by argentometric titration by using a fraction collector. The AgCl precipitates in different fractions were collected on filter disks after acidifying to dissolve chromate which was used as an indicator. The specific activity of Cl in the three major fractions obtained agreed within the experimental errors of counting (ca 10%) (Cl-2).

A third purification of the sample was carried out by repeating the cation exchange and sublimation steps (Cl-3); the measured disintegration rates of the samples were 14±0.6 (Cl-1), 16±1.3 (Cl-2), and 17±1.0 (Cl-3) disintegrations/min after correcting for the chemical yields.

Al²⁶

The targets were dissolved in HCl by passing HCl gas. Iron was oxidized to Fe³⁺ by adding H₂O₂. 53 mg equivalent Al₂O₃ of aluminum carrier was added in each sample along with Be, NH₄⁺ and Cs carriers. Iron was removed by the isopropyl ether extraction from 8M HCl sample solution. The aqueous layer was evaporated to dryness. The residue was extracted in sodium acetate solution. Sodium tetraphenyl-boron was added with a few mg of NH₄⁺ to precipitate potassium fraction. The filtrate was treated with NH₄OH to precipitate the hydroxides of Be, Al, and others. Acetone was used to remove excess ammonium tetraphenyl-borate in the hydroxides. Aluminum was separated by 1M NaOH in solution and the hydroxides

were precipitated from nearly neutral solution. A 9M HCl solution of the sample was passed through an anion exchange column in the RCl form. The ice cold ether-HCl solution of the sample was saturated with HCl gas and AlCl₃·6H₂O was separated in the form of white crystals (Gooch's method).

The chloride was dissolved in water and a mixture of holdback carriers, Zn, Cu, Ni, Co, Fe, Mn, Cr, and V (a few mg each) was added.

(1) Hydroxides were precipitated by adding ammonia in the presence of H₂O₂ and dissolved in a hot HCl solution. An excess of NaOH was added and aluminum was separated. (2) The chloride was precipitated by Gooch's method. (3) The aqueous solution was passed through a 30 ml cation exchange column in the HR form. 0.4M oxalic acid was used to elute aluminum (Merrill et al.²⁹). 1-3 column volumes of the effluents were treated with sulphuric acid and the oxalic acid was decomposed by heating. (4) Copper sulphide scavenging was performed in the 0.3M HCl solution. (5) About 10 mg of Sr carrier was added and SrSO₄ removed from the acidic solution. (6) The solution was made to 10M HCl and passed through an anion exchange column.

Purification steps 1-6 were repeated once and steps 1, 2, and 3 were repeated several times. The chemical yields in the two targets analyzed were 7 and 21%. The latter sample was counted for its beta activity on a low-level counter (Al-1). Two samples were combined (Al-2) and purified by steps 1, 4, 5, and 2, after the addition of hold back carriers. The sample was counted and purified again by steps 3, 4, 1 and 2 (Al-3).

The measured disintegration rates of the three samples Al-1, Al-2, and Al-3, were 2.9±0.6, 1.7±0.2, and 1.7±0.2 dis/min per 10⁹ dis/min K⁴⁸, respectively, after correcting for chemical yields (the aluminum content of the iron target was found to be ca 0.01%). The results of Al-2 and Al-3 were used for the cross-section calculations.

²² W. Rieman, III, and S. Lindenbaum, anal. Chem. 24, 1199 (1952).