Energy Dependence of Product Yields in Copper Spallation by Protons between 3 and 30 GeV*

J. HUDIS, I. DOSTROVSKY, G. FRIEDLANDER, J. R. GROVER, N. T. PORILE, L. P. REMSBERG, R. W. STOENNER, AND S. TANAKA
Chemistry Department, Brookhaven National Laboratory, Upton, New York

(Received 2 August 1962)

Careful comparisons of cross sections for formation of 19 products of copper spallation by 3- and 30-GeV protons indicate only a small energy dependence in this energy range. As the energy is increased from 3 to 30 GeV, products of $A>40$ decrease in yield by 15% or less, except for some highly neutron-deficient species, while low-mass products increase in yield by 10 to 50%. These findings are somewhat at variance with earlier reports from CERN.

RECENT measurements in this laboratory have shown that a wide variety of proton-induced spallation reactions in carbon,[1,2] aluminum,[2] and indium[3] have, within experimental errors, the same cross sections at proton energies of 28 to 30 GeV as at 3 GeV. In view of these findings, it was surprising to learn that Rudstam *et al.*[4] had, in a detailed study of copper spallation by 24-GeV protons at CERN, found the cross sections for formation of almost all of the approximately 50 products investigated ($7 \leqslant A \leqslant 68$) to be substantially lower than the corresponding cross sections reported for 2.2-[5] and 5.7-GeV[6] irradiations of copper. Whereas the CERN workers concluded that the shape of the "yield surface" is almost independent of energy above 1 GeV, they reported that the total spallation cross section of copper at 24 GeV as calculated from their radiochemical data by Rudstam's empirical procedure[7] is about 530 mb, to be compared with a value of 740 ± 20 mb for the nuclear absorption cross section of copper determined by Ashmore *et al.*[8] with counter techniques.

Since Rudstam *et al.*[4] made measurements at a single proton energy and since, therefore, conclusions regarding the energy dependence of copper spallation cross sections can, at present, be based only on measurements performed at different laboratories over a period of ten years, it seemed worthwhile to measure formation cross sections for a number of products of copper spallation with 3- and 30-GeV protons, using the same techniques at the two energies. In such a direct comparison any errors resulting from uncertainties in decay schemes or counting efficiencies should cancel out in the cross section ratios $\sigma_{30\,\mathrm{GeV}}/\sigma_{3\,\mathrm{GeV}}$.

The formation cross sections of 19 spallation products of copper were measured at both 3- and 30-GeV proton bombarding energy. The number of products observed, while not large, is believed to be fairly representative of the total mass-yield surface. The nuclides investigated covered the mass region between 64 and 7 and included both neutron-excess and neutron-deficient products. The results of the present study do not corroborate the conclusions one would draw by comparison of the CERN data[4] with the earlier studies at lower energies,[5,6] but rather show that the cross section ratio $\sigma_{30\,\mathrm{GeV}}/\sigma_{3\,\mathrm{GeV}}$ is between 0.85 and 1.05 for products with $A>40$ (except for highly neutron-deficient species for which the ratio appears to be a little lower) and between 1.1 and 1.5 for low-mass products. Experiments with 6-, 10-, and 20-GeV protons indicate that the formation cross sections of these light products are energy independent above 10 GeV.

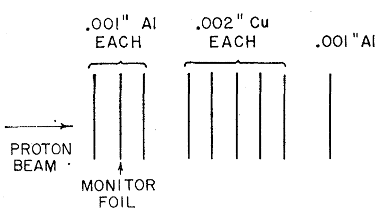

FIG. 1. Target stack.

EXPERIMENTAL PROCEDURE

Targets and Irradiations

Targets consisted of four 0.001-in. aluminum foils plus five 0.002-in. copper foils as shown in Fig. 1. The use of aluminum foils to monitor the proton beam intensity is discussed below. Target stacks were mounted on frames which were then attached to the electromechanical flip mechanism used at the BNL Alternating Gradient Synchrotron. Because of the small aperture of the synchrotron for the 30-GeV proton beam, the most reliable method of targeting was to cause the target to intercept the circulating proton beam before the accelerating voltage was turned off. Thus, the radial position and starting pulse of the flip mechanism were set so that the target intercepted the beam at that time

* Research performed under the auspices of the U. S. Atomic Energy Commission.

[1] J. B. Cumming, G. Friedlander, and S. Katcoff, Phys. Rev. **125**, 2078 (1962).
[2] J. B. Cumming, G. Friedlander, J. Hudis, and A. M. Poskanzer, Phys. Rev. **127**, 950 (1962).
[3] N. T. Porile, Phys. Rev. **128**, 1916 (1962).
[4] G. Rudstam, E. Bruninx, and A. C. Pappas, Phys. Rev. **126**, 1852 (1962). We are grateful to the authors for communicating their results to us prior to publication.
[5] G. Friedlander, J. M. Miller, R. Wolfgang, J. Hudis, and E. Baker, Phys. Rev. **94**, 727 (1954).
[6] D. W. Barr, University of California Radiation Laboratory Report UCRL-3793, 1957 (unpublished).
[7] G. Rudstam, doctoral thesis, NP 6191, University of Uppsala, Uppsala, Sweden, 1956 (unpublished).
[8] A. Ashmore, G. Cocconi, A. N. Diddens, and A. M. Wetherell, Phys. Rev. Letters **5**, 576 (1960).

TABLE I. Properties of radioactive products studied.[a]

Nuclide	Half-life	Radiation detected (Energies in MeV)	Branch abundance	Detector[b]
Cu^{62}	9.7 min	β^+	0.98	511–511 coinc.
Cu^{60}	23.4 min	β^+	0.935	511–511 coinc.
Cu^{61}	3.33 h	β^+	0.62	511–511 coinc.
Cu^{64}	12.8 h	β^+	0.19	511–511 coinc.
Co^{61}	1.65 h	β^- (1.22)	1.00	β prop.
Co^{58}	71 day	γ (0.81)	1.00	well[c]
Co^{57}	270 day	γ (0.122, 0.137)	1.00	well[d]
Co^{56}	77 day	γ (>1.5)		well[c]
Co^{55}	18.0 h	β^+ (1.51, 1.04, 0.79)	0.79	β prop., 511–511 coinc.
Fe^{59}	45 day	β^- (0.46 0.27)	1.00	β prop.[e]
Fe^{52}	8.0 h	β^+ (0.8 2.63)		β prop.[f]
Ti^{45}	3.08 h	γ (0.511 annihil.)	1.70	well
K^{42}	12.5 h	γ (1.52)	0.18	3 in.×3 in. NaI
K^{43}	22.2 h	γ (0.374 0.388 0.394)	1.03	3 in.×3 in. NaI
		γ (0.591 0.619)	0.94	3 in.×3 in. NaI
Ar^{37}	35 day	K and L x rays $+e^-$	1.0	gas-filled prop.
Ar^{41}	1.82 h	β^- (1.20 2.49)	1.0	gas-filled prop.
Na^{22}	2.6 yr	β^+ (0.544)	0.898	β prop.
		γ (0.511 annihil.)	1.80	3 in.×3 in. NaI[g]
Na^{24}	15.0 h	β^- (1.39)	1.0	β prop.
		γ (1.37)	1.0	3 in.×3 in. NaI[h]
Be^7	54 day	γ (0.48)	0.12	well[i]

[a] Unless otherwise indicated, the data on half-lives, energies, and abundances are taken from the National Research Council's Nuclear Data Sheets 1958–1961.

[b] The abbreviations used are "511–511 coinc." for a coincidence apparatus with energy channels for the measurement of coincidences between two annihilation quanta at 180° calibrated by means of a Na^{22} source of known disintegration rate; "β prop." for an end-window gas-flow proportional counter calibrated for various β emitters; "well" for a 2 in.×2 in. NaI(Tl) well crystal; "gas-filled prop." for a proportional counter containing the active gas as a component of the counting-gas mixture; "3 in.×3 in. NaI" for a 3-in.-high, 3-in.-diam NaI(Tl) scintillation detector used in conjunction with a multichannel analyzer and calibrated for photopeak efficiency vs γ energy.

[c] Co^{56} was determined in well counter with discriminator cutoff for γ energies below 1.5 MeV (to eliminate Co^{58} contribution); efficiency for Co^{56} in this arrangement was measured by comparison of samples with a pure, calibrated Co^{56} source at the Institute for Nuclear Studies, University of Tokyo. For Co^{58} determination, the well counter with pulse-height analyzer was used to measure the 0.81-MeV photopeak and its response at this energy was calibrated; the Co^{56} contribution was obtained by comparison with a standard Co^{56} sample and subtracted.

[d] Well counter calibrated for 122- and 137-keV γ rays and used with pulse-height analyzer.

[e] Calibration based on measurement of 1.29-MeV γ (0.44 per disintegration) with standard 3 in.×3 in. NaI crystal.

[f] Calibration based on measurement of 1.43-MeV γ from Mn^{52m} (1.0 per disintegration) with standard 3 in.×3 in. NaI crystal.

[g] Direct comparison with NBS Na^{22} standard.

[h] Relative measurement of 1.37-MeV photopeaks in Al monitor and in sodium fraction isolated from Cu target.

[i] Used in conjunction with multichannel analyzer. Photopeak efficiency of well counter for 0.48-MeV γ determined with the aid of standard 3 in.×3 in. NaI crystal.

during the acceleration cycle at which the proton energy was 30 GeV. Oscilloscope readings of beam intensity vs time indicated that the circulating beam disappeared within 2–3 msec. The identical procedure was used for the lower energy irradiations. Lengths of irradiations were from 10–20 min at 10, 20, and 30 GeV and 20–30 min at 3 and 6 GeV, and the average circulating beam intensity during each run was $\sim 4\times 10^{12}$ protons/min, which resulted in integrated fluxes of $(3\text{–}10)\times 10^{14}$ protons through the target stack disks.

After the irradiations the targets were quickly removed from the synchrotron through an air lock, and a $\frac{1}{2}$-in.-diam stack of foils was punched out of the target about 2–3 mm back from the leading edge. Individual copper foils were processed for different elements with care taken that the center foil protected by 0.004 in. of copper on both sides was always used for sodium and beryllium yields since these products (a) are expected to have the largest recoil ranges, and (b) can be formed from aluminum also.

Chemical Separations and Activity Measurements

From 2 mg (Co and Fe) to 15 mg (Ti) of carrier were added to the dissolving acid (HNO_3 or $HCl+H_2O_2$) prior to dissolution of the copper target foil. Standard chemical procedures[9] were employed for the separation and purification of most elements. The procedure used for the isolation and purification of argon will be described elsewhere.[10] The relevant properties of the radioactive products studied are listed in Table I, along with the measurement techniques used. Wherever possible, gamma-ray spectroscopy and annihilation radiation coincidence measurements were used for the identification and assay of the radiations of interest. Where necessary, decay curves were analyzed into components (with the half-lives listed in Table I) by means of a least-squares computer program.[11] The resulting counting rates at end of irradiation were corrected for detection efficiency, abundance of radiations detected (Table I), chemical yield, and duration of irradiation, to yield a saturation disintegration rate for each nuclide. This, in turn, was combined with the measured proton flux through the target to give the formation cross section.

Beam Intensity Measurements

The total proton flux through the target disk was determined from the monitor reaction $Al^{27}(p,3pn)Na^{24}$. Measurements on the formation cross section of Na^{24} from aluminum at 28 and 2.9 GeV have recently been reported[2,12] and the published values of 8.6 mb at 28 GeV and 9.1 mb at 2.9 GeV were used for the 30- and 3-GeV irradiations, respectively. The monitor cross sections at 6 GeV (8.7 mb), 10 GeV (8.6 mb), and 20 GeV (8.6 mb) were also taken from the work of Cumming *et al.*[12] It is known that low-energy secondary particles can also contribute to the observed yield of Na^{24} from aluminum, and since the targets were relatively thick, ≈ 230 mg/cm^2, the effect of secondaries on the monitor reaction was determined empirically. Since the yield of F^{18} from aluminum is relatively insensitive to low-energy secondaries, the ratio of F^{18} to Na^{24} production in aluminum was obtained in at least one run at each energy, and with the thin-target values[12] of $\sigma_{Al}(F^{18})$, was used to correct the observed Na^{24} yields for the secondary effects. The correction amounted to 3% at 30 GeV and 2% at 3 GeV. As shown in Fig. 1, the aluminum foil used for the beam-intensity measurement was placed so that recoil losses from it were compensated by recoil gains from neighboring foils and so that it was shielded from the copper foils.

[9] *National Academy of Sciences Committee on Nuclear Science, Radiochemistry Monographs*, National Academy of Sciences, National Research Council 3001–3056 (U. S. Government Printing Office, Washington, D. C.).

[10] I. Dostrovsky and R. W. Stoenner (to be published).

[11] J. B. Cumming (unpublished).

[12] J. B. Cumming, J. Hudis, A. M. Poskanzer, and S. Kaufman, Phys. Rev. **128**, 2392 (1962).

TABLE II. Ratio of formation cross sections of Cu spallation products at 30 and 3 GeV and absolute formation cross sections in millibarns at various energies.

Nuclide	σ_{30}/σ_3	σ_{30}	σ_{20}[b]	σ_3	σ_{24}[c]	$\sigma_{5.7}$[d]	$\sigma_{2.2}$[e]
Cu^{64}[a]	1.02 ± 0.10	52.0	53.3	51.0	61	59	55[f]
Cu^{62}	0.96 ± 0.09	31.0	31.2	32.2		26.5	18
Cu^{61}	0.98 ± 0.09	11.1	11.4	11.3	11	12.4	6.9
Cu^{60}	0.87 ± 0.09	1.72	1.78	1.97		2.5	1.9
Co^{61}	0.85 ± 0.14	4.0	3.8	4.7	3.8	4.4	3.8
Co^{58}	0.91 ± 0.08	17.5	18.5	19.3	14	27	
Co^{57}	0.96 ± 0.09	14.0	14.2	14.7	9.2	20	
Co^{56}	1.02 ± 0.16	5.6	5.6	5.5	7.6	4.2	
Co^{55}	0.71 ± 0.13	0.83	0.77	1.17	0.76	1.49	1.6
Fe^{59}	0.92 ± 0.12	1.44	1.44	1.59	1.3	0.64	0.6
Fe^{52}	0.74 ± 0.09	0.11	0.11	0.16	0.10	0.11	0.20
Ti^{45}	0.84 ± 0.09	3.04	2.92	3.63	2.1	2.7	2.6
K^{43}	1.05 ± 0.10	0.98	1.08	0.93	1.0	0.91	1.4
K^{42}	0.98 ± 0.10	2.83	3.0	2.90	2.7	4.5	3.9
Ar^{41}	0.85 ± 0.10	0.73	0.71	0.87		0.72	
Ar^{37}	0.79 ± 0.09	4.80	4.7	6.1		4.9	
Na^{24}	1.18 ± 0.11	3.48	3.91	2.96	1.8	4.0	3.4
Na^{22}	1.49 ± 0.23	2.75	2.56	1.85	1.4	2.4	1.9
Be^7	1.36 ± 0.14	10.1	10.6	7.4	7.0	13.7	10.5

[a] The cross sections for Cu^{64} are for production from Cu^{65} only.
[b] Single determination.
[c] From reference 4.
[d] From reference 6, adjusted for $\sigma_{Al}(Na^{24})=8.7$ mb.
[e] From reference 5, adjusted for $\sigma_{Al}(Na^{24})=9.5$ mb.
[f] From S. S. Markowitz, F. S. Rowland, and G. Friedlander, Phys. Rev. **112**, 1295 (1958), with adjustment of $\sigma_{Al}(Na^{24})$.

RESULTS

The results of the experiments are listed in Table II. The second column contains the ratio of the formation cross section at 30 GeV (σ_{30}) to that at 3 GeV (σ_3) for each nuclide; results of duplicate experiments were averaged at each energy. The ratios are, of course, unaffected by any possible errors in counting efficiencies and assumed branching ratios. The errors quoted in this column were obtained as follows. The error on the cross section at each energy arising from uncertainties in the determination of the Na^{24} disintegration rate in the aluminum foil, and in the disintegration rate and chemical yield of the copper spallation product was estimated and compared with the deviation of the duplicate results from their average. The larger of these two error estimates (in percent) at 30 GeV and at 3 GeV were combined by root mean square addition with the

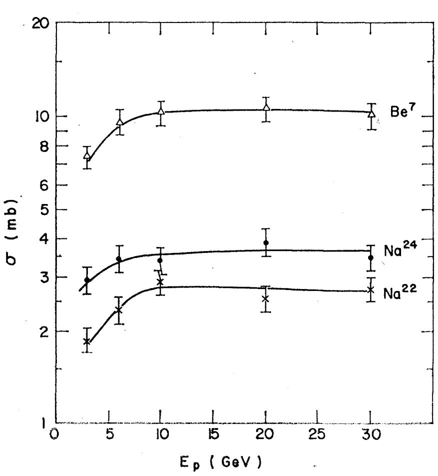

FIG. 2. Excitation functions for the production of Be^7, Na^{24}, and Na^{22} from copper between 3 and 30 GeV.

$\pm7\%$ uncertainty[2,12] in the ratio of the monitor cross section at the two energies to yield the quoted errors. It is seen that the uncertainty in the monitor cross section is dominant in most cases.

Columns 3, 4, and 5 of Table II list the formation cross sections in millibarns at 30, 20, and 3 GeV, respectively, and these results are, of course, subject to a larger error than the ratio measurements since counting efficiencies and decay scheme corrections do not cancel. In the last three columns the published data of Rudstam *et al.*[4] at 24 GeV, of Barr[6] at 5.7 GeV, and of Friedlander *et al.*[5] at 2.2 GeV are shown for comparison.

It is obvious from column 2 that there are no large changes in formation cross sections when the energy of the incident protons is raised from 3 to 30 GeV. In the region of $A>40$, only the highly neutron-deficient products Co^{55} and Fe^{52} (which are produced in small yield) show σ_{30}/σ_3 ratios as low as ~0.7; for all other products investigated the ratio is $\geqslant0.84$, and for most of the high-yield products even closer to unity. The formation cross sections of the three products of lowest mass studied—Na^{24}, Na^{22}, Be^7—definitely increase when the proton energy is raised from 3 to 30 GeV. Values of $\sigma_{Cu}(Na^{24})$, $\sigma_{Cu}(Na^{22})$, and $\sigma_{Cu}(Be^7)$ were measured at 6 and 10 GeV also, and the results are given in the form of excitation functions in Fig. 2. The data indicate that the increase in these cross sections occurs almost entirely below 10 GeV, with no obvious change between 10 and 30 GeV.

DISCUSSION

The most striking result of the present study is that the spallation cross sections studied do not show a large energy dependence between 3 and 30 GeV. More detailed examination indicates a shift of the mass-yield curve towards lighter products and a slight relative decrease of neutron-deficient species within about 20 mass numbers of the target, when the bombarding energy is increased. Although only 19 products were selected for study, they would seem to be sufficiently representative to allow these conclusions to be generalized. The question then arises why Rudstam *et al.*,[4] in their more complete investigation with 24-GeV protons, did not arrive at the same result. Comparison of the present data with the CERN results (columns 3, 4, and 6 of Table II) indicates rather good agreement for about half the nuclides measured in both studies, with the CERN cross sections lower than ours for most of the other nuclides and with the largest discrepancies found for Na^{24} and Na^{22}. In our work, the Na^{24} yields obtained by β detection were checked at least once at each bombarding energy by a direct comparison of the photopeak intensities of the 1.37-MeV γ ray of Na^{24} in the sodium sample separated from the copper target and in the aluminum monitor. Agreement between the two methods was always excellent. The absolute Na^{24} cross sections measured in this study are believed to be reliable to $\pm10\%$ or better. In addition to discrepancies between the present 30- and 20-GeV results and the ones of Rudstam *et al.* at 24 GeV, there are also significant differences between the present 3 GeV and the older 6 GeV[6] and 2.2 GeV[5] cross sections. The older measurements were, in general, less precise and less reliable, and the conclusion of Rudstam *et al.* that the cross sections at 24 GeV are, in general, lower than those at lower energies must in part be ascribed to this fact. Without doubt, the present direct comparison of 3 GeV and 30 GeV cross sections provides a sounder test of energy dependence than the comparison of data from different laboratories obtained with different techniques.

The present results are too sparse to allow construction of a mass yield curve and therefore do not shed any direct light on the apparent discrepancy, reported by Rudstam *et al.*,[4] between the total reaction cross section of copper determined by counter measurements and that inferred from radiochemical data. However, the small energy dependence of spallation cross sections found here indicates that this discrepancy is probably not real at 30 GeV unless it should also exist at about 3 GeV. Possibly some of the erroneous spallation cross sections reported by the CERN group (e.g., of the Na isotopes) as well as the shortcomings of their empirical interpolation formula are responsible for their apparent deficit in total reaction cross section.

Our data also indicate that the shape of the yield surface is not quite as independent of energy as Rudstam *et al.* reported although the changes with energy in the region investigated are not dramatic. It would seem that the yield pattern shifts somewhat to lower mass products as the proton energy is increased from 3 to 20 or 30 GeV. In other words, the slope of the mass yield curve (parameter P in Rudstam's equation[4,7]) decreases further in this energy range. This same trend, but to a lesser extent, was noted in aluminum spallation.[2,12]

The interaction between high-energy incident nucleons and complex nuclei has been described in terms of a cascade-evaporation model.[13] Monte Carlo calculations[14] based on this model account quite well for the gross features of the spallation cross sections up to 2 GeV. The small energy dependence noted in this work between 3 and 30 GeV, which also holds for carbon,[1,2] aluminum,[2,12] and indium[3] targets indicates that as the energy of the incident particle is raised above 3 GeV, little of the additional energy is deposited in the target nuclei; most of it probably appears in the form of kinetic energy of the nucleons and especially of rest and kinetic energy of the mesons ejected during the intranuclear cascades. The emulsion work of Barbaro-Galtieri *et al.*[15] is in accord with this premise. They find that the multiplicity of black prongs which includes all evaporated charged particles and protons with energies up to $\sim$400 MeV remains essentially constant between 2 and 27 GeV whereas the multiplicity of shower particles which consist mainly of pions increases linearly with incident energy, from $\sim$1 per event at 3 GeV to $\sim$7 per event at 27 GeV.

ACKNOWLEDGMENTS

The cooperation and help of the AGS operating staff is gratefully acknowledged. Thanks also go to Dr. K. Rowley and Miss E. Norton for performing many of the chemical yield analyses.

---

[13] R. Serber, Phys. Rev. **72**, 1114 (1947).

[14] N. Metropolis, R. Bivins, M. Storm, J. M. Miller, G. Friedlander, and A. Turkevich, Phys. Rev. **110**, 204 (1958).

[15] A. Barbaro-Galtieri, A. Manfredini, B. Quassiati, C. Castaguoli, A. Garnotti, and I. Ortalli, Nuovo Cimento **21**, 469 (1961).