Spallation of Copper with 24-Gev Protons

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(Received December 11, 1961)

The yields of more than 50 spallation products obtained from the irradiation of copper with 24-Gev protons have been measured. A comparison with results at lower energies shows that the cross-section distribution of the spallation products hardly changes above 1 Gev.

I. INTRODUCTION

THE successful start of the CERN proton synchrotron in November, 1959, opened up the possibility of extending spallation studies to the ultra-high-energy region. Using this experimental facility, the cross sections of the spallation products of copper irradiated with 24-Gev protons have been determined.

Copper was chosen as target material in this experiment for two reasons. In the first place, the spallation of copper has been extensively investigated at lower energies and with different kinds of bombarding particles (see below) so that a vast amount of experimental data is available for comparison. In the second place, copper foils are obtainable in a very high purity, and can easily be mounted as flip targets of the type used in the proton synchrotron. A disadvantage is that natural copper contains two stable isotopes, Cu^{63}(69.0%) and Cu^{65}(31.0%). Except for a few cases, it is therefore impossible to state the extent to which each of these nuclides contributes to the measured cross sections of the spallation products.

Earlier spallation studies of copper have been carried out with protons of energy 50 Mev,[1] 90 and 190 Mev,[2] 340 Mev,[3] 480 and 680 Mev,[4] 660 Mev,[5] 980 Mev,[6] and 2.2 Gev[7] and 5.7 Gev.[8] Moreover, 190- and 280-Mev deuterons,[3,4] 190 and 380-Mev α particles,[3] and 90- and 400-Mev neutrons[2,9] have been used as bombarding particles.

II. EXPERIMENTAL

1. Irradiations

Copper foils of 99.999% purity were irradiated with 24-Gev protons in the internal beam. The thickness of the targets was always less than 4 mm. The energy degradation in such a target is of the order of a few hundred Mev, which is negligible in comparison with the maximum proton energy used. Other sources which tend to broaden the energy spectrum are also unimportant. The energy spread of the protons in the beam is therefore expected to be small.[10] The maximum energy varied somewhat from irradiation to irradiation however, and the proton energy for the present investigation can be given as 24 ± 1 Gev.

The nuclide Ni^{57} was chosen as an internal monitor and all cross sections were determined relative to the formation cross section of this nuclide. In some irradiations the target was a small stack consisting of three thin aluminium foils (each of thickness 0.01 mm) on each side of a 0.05-mm copper foil. Using this target arrangement the cross section of Ni^{57} formed from copper was determined relative to the cross section of the reaction $Al^{27}(p,3pn)Na^{24}$. It is necessary to make such a determination with a thin target as the effect of secondary neutrons in producing Na^{24} via the reaction $Al^{27}(n,\alpha)Na^{24}$ might become disturbingly large in a thick-target arrangement. Similar secondary reactions might also contribute to the measured cross sections of simple reactions induced in the copper target. Therefore, the targets normally used were thin (0.05 mm) except for a few early runs with 3.5-mm targets. In the experiments with thick targets no simple reactions were studied, however, and no secondary effect was detected.

Owing to the rather high intensity of the proton beam (about 2×10^{11} protons per pulse and one pulse every third second), short irradiations were usually sufficient. Occasionally long irradiations were used when long-lived products were measured.

2. Chemical Separations

The irradiated target was dissolved in nitric acid containing carriers of the elements to be studied. The determination of β activities is facilitated if the samples for activity measurements are thin. For this reason the amounts of carriers used were only 0.2 or 0.4 mg, except for the elements fluorine, phosphorus, sulfur, and chlorine, where the chemical separation methods require a larger amount of carrier, 5–10 mg. The chemical methods given by one of the authors[11] were used, except for

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sodium, magnesium, potassium, and calcium. These four elements were isolated as a group and then separated from each other by cation exchange. Beryllium was purified from most contaminants by precipitation with ammonia, followed by dissolution into sodium hydroxide. A subsequent purification step was the extraction of the acetyl acetonate into benzene from an acetic acid solution containing ethylenediaminetetraacetic acid (EDTA). Fluorine was purified by distillation of hydrogen fluoride, and precipitation of calcium fluoride and lead chlorofluoride.

The chemical yield of the elements studied was determined by spectrophotometry (Be, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu), flame photometry (Na, K), microtitration with EDTA using a spectrophotometer to determine the end point (Mg, Ca, Zn), or gravimetric methods (F, P, S, Cl).

3. Preparation of Samples for Counting

The disintegration rates of the different nuclides in question must be known in order to make possible the determination of the respective cross sections. The disintegration rates were, in most cases, obtained from the β-counting rates in a low-geometry arrangement using a flow proportional counting tube. It is well known that the conversion of β-counting rates to disintegration rates offers considerable difficulties because of complicated scattering and absorption effects. In the early irradiations the samples for counting were prepared simply by the evaporation of a measured amount of a solution on a thin aluminum foil, after the addition of some insulin. Later on, the electro-spraying method[12] was used. This method is more satisfactory because the self-scattering and self-absorption effect is small and can be accurately corrected for. When 5–10 mg of carrier were used, the samples for counting were prepared by the filtering technique.

The correction factors for conversion of counting rates to disintegration rates were taken from the works of Rudstam[11] and of Pappas.[13]

The complex β-decay curves of chlorine, scandium, manganese, and cobalt are difficult to resolve. For the isotopes of these elements, a γ spectrometer was used to determine the disintegration rates. In addition, certain electron-capturing nuclides such as Be^7 and Ca^{47} were measured by γ counting, while purely electron-capturing nuclides such as V^{49} and Fe^{55} were measured with an x-ray counter.

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III. CROSS-SECTIONS OF THE SPALLATION PRODUCTS

The measured cross sections[14–85] (averages of several determinations) are given in Table I. The relative cross

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sections have been converted to absolute cross sections using the value 8.3±0.5 mb for the reaction $Al^{27}(p,3pn)$-Na^{24} as given by Cumming *et al.*[86] for 28-Gev protons. Since the energy dependence of this cross section is rather small, it can also be used for 24-Gev protons.

In most cases it has not been possible to correct for the growing-in from short-lived parent nuclides. Such corrections are small, however, because the cross section of a parent nuclide is generally much smaller than that of the daughter nuclide for the range of nuclides studied.[11] The cross sections given in the table therefore approximate well the primary yields of the spallation products.

IV. COMPARISON BETWEEN SPALLATION YIELDS OBTAINED AT DIFFERENT IRRADIATION ENERGIES

Cross sections of products from spallation of copper at irradiation energies from 90 Mev to 24 Gev are now available[2–4,6–8,87] and are compared with our results in Table II. The cross sections have been recalculated using recent values of the cross section of the monitor reaction $Al^{27}(p,3pn)Na^{24}$.[88] The table indicates that the cross sections at 24 Gev are in general lower than those at 2.2 and 5.7 Gev.

A simple way to compare different cross-section distributions is to represent them by a suitable analytical formula. A useful empirical formula is the following:

$$\sigma(Z,A)=\exp(PA-Q-R|Z-SA+TA^2|^{\frac{3}{2}}), \quad (1)$$

where $\sigma(Z,A)$ is the formation cross section of the spallation product (Z,A), and P, Q, R, S, and T are parameters depending on the irradiation conditions, i.e., on target and on type and energy of bombarding particle.[89] These parameters can be determined from experimental data by the method of differential corrections. They are given in Table III for a number of distributions obtained in proton-induced spallation of copper. The last column in Table III gives e^{ϵ}, where ϵ is the standard deviation of the difference $(\ln\sigma_{\text{exp}}-\ln\sigma_{\text{calc}})$. Thus e^{ϵ} indicates how well Eq. (1) represents the experimental cross sections. For spallation of copper with 24-Gev protons one finds a value 1.57, showing that on an average the experimental and the calculated cross sections agree within 60%. For this case, the experimental and calculated cross-sections are compared in Table I.

It is evident from Table III that the parameter P is the only one which depends on irradiation energy. This parameter defines the slope of the yield-mass curve. It decreases with increasing irradiation energy below about 1 Gev and then remains approximately constant as also shown in Fig. 1.

The parameter R defines the width of the charge distribution which apparently is independent of irradiation energy. The position of the peak of the charge distribution is Z_p, defined by

$$Z_p=SA-TA^2. \quad (2)$$

(For a narrow mass range one can assume Z_p to be proportional to the mass number. The form chosen is applicable to any mass number, however, and this with a unique set of values of the parameters, or $S=0.487\pm0.003$; $T=0.00042\pm0.00004$.[89]) It is independent of irradiation energy.

Since the yield-mass distribution is very flat in the Gev region, the distance of a nuclide from the line of maximum primary yield Z_p is much more important in deciding the yield than the difference in mass between the nuclide and the target.

V. TOTAL CROSS SECTION FOR SPALLATION

Equation (1) can be used to estimate the total cross section for the formation of spallation products. The cross section for forming products in the mass range

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TABLE I. Experimental and calculated spallation yields.

Nuclide	$\sigma_{exp}/\sigma_{Ni^{57}}$	σ_{exp} (mb)	σ_{calc} (mb)	Number of determinations	Decay scheme references
Be^{7}	13 ± 1	7.0 ±0.8		3	14
F^{18}	2.7 ± 0.1	1.5 ±0.1	1.2	2	15
Na^{22}	2.6 ± 0.3	1.4 ±0.2	1.2	3	16
Na^{24}	3.3 ± 0.2	1.8 ±0.2	1.3	6	11, 17
Mg^{28}	0.47 ± 0.05	0.25 ±0.03	0.14	5	18
P^{32}	5.7 ± 0.6	3.1 ±0.4	3.6	1	19, 20
P^{33}	2.0 ± 0.2	1.1 ±0.1	1.6	1	21
S^{35}	1.6 ± 0.2	0.87 ±0.13	2.2	1	19, 22
S^{38}	0.022± 0.012	0.012±0.006	0.027	2	23
Cl^{34m}	0.37 ± 0.08	0.20 ±0.05		2	24
Cl^{38}	4.1 ± 1.1	2.2 ±0.6	1.0	3	23, 25
Cl^{39}	0.49 ± 0.25	0.26 ±0.13	0.23	3	26, 27
K^{42}	5.0 ± 0.5	2.7 ±0.3	2.1	6	28, 29
K^{43}	1.8 ± 0.2	1.0 ±0.1	0.6	6	30, 31
Ca^{45}	1.1 ± 0.2	0.60 ±0.12	0.90	5	32, 33
Ca^{47}	0.050± 0.009	0.027±0.005	0.033	5	34, 35
Sc^{43}	3.2 ± 1.0	1.7 ±0.6	2.3	2	36, 37
Sc^{44}	4.5 ± 0.4	2.4 ±0.3	5.4	2	37, 38, 39
Sc^{44m}	4.4 ± 0.3	2.4 ±0.3		3	11, 38, 39
Sc^{46}	6.6 ± 0.8	3.6 ±0.5	4.4	4	40, 41
Sc^{47}	3.2 ± 0.2	1.7 ±0.2	1.4	3	42, 43
Sc^{48}	0.61 ± 0.10	0.33 ±0.06	0.32	3	44, 45
Ti^{45}	3.9 ± 0.5	2.1 ±0.3	2.0	2	46, 47
V^{47}	3.3 ± 0.7	1.8 ±0.4	1.7	1	11, 48
V^{48}	14 ± 2	7.6 ±1.2	5.6	3	11, 49
V^{49}	38 ±10	20 ±6	12	1	50, 51
Cr^{48}	0.50 ± 0.05	0.27 ±0.03	0.27	1	52, 53
Cr^{49}	2.6 ± 0.4	1.4 ±0.2	1.4	2	11, 54
Cr^{51}	22 ± 4	12 ±2	12	2	55, 56
Mn^{51}	0.86 ± 0.09	0.47 ±0.06	1.1	1	11, 57
Mn^{52}	7.4 ± 0.5	4.0 ±0.4	4.3	2	58, 59
Mn^{52m}	1.5 ± 0.3	0.8 ±0.2		1	11, 58
Mn^{54}	12 ± 2	6.5 ±1.2	16	1	50, 60
Mn^{56}	3.2 ± 0.3	1.7 ±0.2	2.5	3	61, 62
Fe^{52}	0.18 ± 0.02	0.10 ±0.01	0.15	3	11, 63
Fe^{55}	26 ± 3	14 ±2	11	1	50, 64
Fe^{59}	2.4 ± 0.1	1.3 ±0.1	1.0	3	65, 66
Co^{55}	1.4 ± 0.1	0.76 ±0.09	0.65	5	67, 68
Co^{56}	14 ± 1	7.6 ±0.9	3.0	3	69, 70
Co^{57}	17 ± 1	9.2 ±0.9	10	5	71, 72
Co^{58}	25 ± 3	14 ±2	22	3	56, 73
Co^{61}	7.1 ± 0.9	3.8 ±0.6	1.8	3	11, 74
Ni^{57}	1.00	0.54 ±0.04	0.48	5	11, 75
Ni^{65}	0.083± 0.003[a]	0.14 ±0.01[b]		5	76, 77
Cu^{61}	21 ± 2	11 ±1	7.4	2	11, 78, 79
Cu^{64}	36 ± 2	61 ±6[b]		3	77, 80
Zn^{62}	0.21 ± 0.03	0.11 ±0.02		7	81
Zn^{65}	0.26 ± 0.06[a]	0.45 ±0.10[b]		2	41, 82
Ga^{66}	0.055± 0.006[a]	0.030±0.004		3	11, 83
Ga^{67}	0.041± 0.025[a]	0.022±0.013		2	11, 84
Ga^{68}	0.036± 0.003[a]	0.019±0.002		1	11, 85

[a] Target consisting of 0.05 mm Cu +0.06 mm Al.
[b] Cross sections based on isotopic abundance of Cu^{65}.

A_1-A_2 is obtained as

$$\sum \sigma(Z,A) \sim \int_A \int_Z \sigma(Z,A)\,dA\,dZ \sim \frac{1.79\,e^{-Q}}{R^{\frac{2}{3}}} \frac{}{P}(e^{PA_2}-e^{PA_1}), \quad (3)$$

where the isobaric yield has been obtained by integration between the limits $-\infty$ to $+\infty$. This is permissible because the charge distribution falls off rapidly at the wings. In the case of copper, the upper limit of integration can be put equal to 61. The lower limit is not well defined, but since Eq. (1) holds at least down to F^{18}, a lower limit of 18 can be used. For light fragments (cf. Be^{7}) the yields increase again. It is to be expected, however, that a heavier product is also formed at the same time as these light fragments. Therefore the reactions leading to such fragments are already contained in the integrated cross section.

With the upper limit of integration equal to 61, the cross sections of all possible products with mass number from 62 to 65 must be estimated. These products are $Zn^{62}-Zn^{65}$, $Cu^{62}-Cu^{64}$, $Ni^{62}-Ni^{64}$, $Co^{62}-Co^{63}$, and Fe^{62}.

TABLE II. Copper spallation cross sections in millibarns.

Nuclide	90 Mev[a]	190 Mev[a]	340 Mev[b,c]	680 Mev[d]	980 Mev[e]	2.2 Gev[f,c]	5.7 Gev[g]	24 Gev[h]
Be^{7}						12	17	7.0
F^{18}					1.8	1.2	3.4	1.5
Na^{22}			0.036	0.043		2.1	2.9	1.2
Na^{24}			0.029	0.32		3.7	4.8	1.3
Mg^{28}						0.47	0.65	0.25
P^{32}			0.12	0.63		7.4	7.8	3.1
P^{33}						1.0	2.2	1.1
S^{35}				0.24		1.1	2.0	0.87
S^{38}								0.012
Cl^{34m}			0.018	0.027	3.8	1.0	0.73	0.20
Cl^{38}			0.058	0.062	0.8	0.77	2.0	2.2
Cl^{39}			0.0092	0.014	0.6	0.46	0.60	0.26
K^{42}				0.36	2.3	4.3	5.4	2.7
K^{43}				0.16	2.8	1.5	1.1	1.0
Ca^{45}			0.16	0.44		1.3	1.2	0.60
Ca^{47}			0.0092	0.063		0.11	0.086	0.027
Sc^{43}							3.8	1.7
Sc^{44}							4.6	2.4
Sc^{44m}			1.4			5.8	4.7	2.4
Sc^{46}			1.7	1.9		7.5	7.8	3.6
Sc^{47}			0.75	2.9		4.0	3.0	1.7
Sc^{48}				2.0		2.8	0.38	0.33
Ti^{45}			1.0	2.5		2.9	3.3	2.1
V^{47}				3.1	2.6	3.1	3.1	1.8
V^{48}	0.02	2.6	0.73	2.2	26	11	10	7.6
V^{49}				11		31	13	20
Cr^{48}						0.6	0.22	0.27
Cr^{49}	0.01	0.7	0.94		3.1	2.5	2.8	1.4
Cr^{51}			7.1	20		22	14	12
Mn^{51}		0.3	1.6			1.6	2.2	0.47
Mn^{52}			7.1	6.5		6.2	5.2	4.0
Mn^{52m}	0.1	0.4		2.1		1.8	1.9	0.8
Mn^{54}			12	10		14	16	6.5
Mn^{56}	0.9	1.6	2.5	3.6		2.7	2.8	1.7
Fe^{52}	0.008	0.10	0.18	0.21	0.44	0.22	0.13	0.10
Fe^{55}			11			14	17	14
Fe^{59}			0.78	0.50		0.7	0.77	1.3
Co^{55}	0.6	1.4	2.3	3.1		1.7	1.8	0.65
Co^{56}			3.4	8.0			5.1	7.6
Co^{57}							24	9.2
Co^{58}			58	27			33	14
Co^{61}	2	2	4.8	12		4.2	3.3	3.8
Ni^{57}	0.9	1.3	1.8	0.41		0.7	0.72	0.54
Ni^{65}				0.27[i]			0.22[i]	0.14[i,j]
Cu^{61}	65	30	23	38	10	7.5	15	11
Cu^{62}		42	48	50	22	20	32	
Cu^{64}	150[i]	77[i]	74[i]	45[i]	66[i]	27[i]	71[i]	61[i]
Zn^{62}	8	2	0.80			0.18–0.7	0.20	0.11
Zn^{63}		2	1.1			0.5–0.9	0.87	
Zn^{65}			0.74[i]				3.1[i]	0.45[i,j]
Ga^{66}			0.01			0.05	0.061	0.030[j]
Ga^{67}			0.006			0.07		0.022[j]
Ga^{68}			0.01			0.05	0.064	0.019[j]

[a] Reference 2. [b] Reference 3. [c] Reference 87. [d] Reference 4. [e] Reference 6. [f] Reference 7. [g] Reference 8. [h] This work.
[i] Cross section based on the isotopic abundance of Cu^{65}. [j] Target consisting of 0.05 mm Cu + 0.06 mm Al.

Of these products, Zn^{62} and Zn^{65} have been measured (Table I), and Zn^{63} has been measured at 2.2 and 5.7 Gev (Table II). All these cross sections are low, and the sum of the formation cross sections of Zn^{62}—Zn^{65} will contribute only about 1 mb to the total cross section at 24 Gev.

The cross section of Cu^{62} has been measured to be 20 mb at 2.2 Gev and 32 mb at 5.7 Gev. The cross section of Cu^{64} (from natural copper) is 19 mb at 24 Gev. Estimating the cross section of Cu^{63} to be 10 mb, one obtains the total cross section of Cu^{62}—Cu^{64} to be about 60 mb at 24 Gev.

Strohal and Caretto[90] have shown that the ratio between the yields of the (p,pn) reaction and the $(p,2p)$ reaction in the same target is fairly independent of target nucleus and of irradiation energy. Using their ratio at 440 Mev, 1.62, the sum of the cross sections of Ni^{62}—Ni^{64} can be estimated to be about 40 mb at 24 Gev.

The yields of Co^{62}, Co^{63}, and Fe^{62} should be smaller than the cross sections of the copper and nickel isotopes mentioned above by more than one order of magnitude,

[90] P. P. Strohal and A. A. Caretto, Jr., Phys. Rev. 121, 1815 (1961).

TABLE III. The parameters P, R, S, and T of Eq. (1). The errors given are standard deviations.

Irr. energy	P	R	S	T	e^ϵ
50 Mev	0.90 ±0.15	1.4±0.5	0.43±0.08	−0.0007±0.0013	1.96
90 Mev	0.46 ±0.15	1.8±1.0	0.46±0.07	−0.0001±0.0011	2.14
190 Mev	0.22 ±0.10	2.2±0.5	0.47±0.04	0.0000±0.0007	1.73
340 Mev	0.25 ±0.02	1.9±0.2	0.48±0.01	0.0002±0.0001	1.73
680 Mev	0.16 ±0.02	1.7±0.3	0.50±0.01	0.0006±0.0002	2.44
980 Mev	0.052±0.035	2.0±0.3	0.49±0.02	0.0004±0.0003	1.72
2.2 Gev	0.066±0.014	1.8±0.2	0.49±0.01	0.0005±0.0001	1.66
5.7 Gev	0.052±0.006	2.0±0.1	0.49±0.01	0.0004±0.0001	1.43
24 Gev	0.065±0.008	2.0±0.1	0.49±0.01	0.0004±0.0001	1.57

and they can therefore be neglected. The sum of the cross sections of the products above mass number 61 is then estimated to be about 100 mb. The integrated cross section for products of mass number ranging from 18 to 61 is 430 mb. Thus the total cross section for formation of spallation products should be about 530 mb.

This result can be compared to the nuclear absorption cross-section measured by Ashmore *et al.*[91] Their result for copper, 740±20 mb, is about 40% higher, which seems to be outside the error of the present treatment. The difference could possibly be accounted for by attributing a high cross section to inelastic scattering processes, leaving the target nucleus with so low excitation energy that neutron evaporation is impossible. In such a case no radioactive product is formed, and the event escapes detection. Another possible explanation is that the cross sections of stable unmeasured products are larger than expected from Eq. (1). A systematic investigation does not indicate abnormally large yields close to the peak of the charge distribution, however.[89]

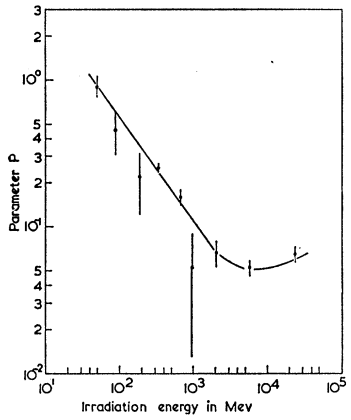

FIG. 1. The parameter P as a function of the irradiation energy.

VI. CONCLUSION

The present investigation indicates that no drastic changes in the spallation yield pattern occur when the irradiation energy is raised. On the contrary, there is a smooth variation in the yield distribution of the spallation products all the way from 50 Mev to 24 Gev with practically no changes at all above 1 Gev.

ACKNOWLEDGMENTS

The authors wish to express their gratitude to the Machine Group of the CERN proton synchrotron for carrying out the irradiations, and especially to Dr. K. H. Reich and Dr. T. Sluyters for their kind assistance in arranging the experiments.

[91] A. Ashmore, G. Cocconi, A. N. Diddens, and A. M. Wetherell, Phys. Rev. Letters **5**, 576 (1960) and personal communication by A. N. Diddens.