Spallation of copper by high-energy alpha particles*

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28 radionuclides produced by spallation of natural copper by 720 MeV α particles were investigated by direct γ counting of irradiated targets. Although the isobaric yield distributions and mass yield curve are similar in many respects to those found in proton spallation, a suppression of yields of neutron deficient products is observed. Several interpretations of this latter phenomenon are critically discussed.

[NUCLEAR REACTIONS Cu(α, spallation), E=410, 720 MeV; measured formation cross sections 28 radionuclides, direct γ counting, natural target.]

I. INTRODUCTION

Spallation of complex nuclei by high-energy protons has been under intensive investigation for a considerable number of years. The status of our understanding of the variety of phenomena evident in these reactions is quite satisfactory although not by any means complete. Much of the observed results are believed to be well explained by the two step cascade-evaporation model[1,2] suggested by Serber in 1947. Monte Carlo simulations have been successfully employed in duplicating the cascade-evaporation process at energies below which meson production becomes important (400 MeV).[3-8] The success of these calculations is evidenced by agreement with a multitude of experimental data.

Surprisingly few radiochemical investigations of the interactions of high-energy α particles with complex nuclei have been performed. Aside from several studies on light nuclei, spallations of iodine,[9] niobium,[10] and some very early work on copper[11] are the only investigations existing in the literature other than those of some very specific, selected reactions. For example, α-induced neutron knockout studies have been reported recently by Church.[12] Fragmentation has been examined via ^{24}Na and ^{28}Mg production from several target nuclei by Korteling and Hyde[10] and Crespo *et al.*[13] However, even the aforementioned iodine, niobium, and copper studies encompass only a small number of products not completely representative of the systematic trends such as have been thoroughly revealed in proton-nucleus studies. In fact, no thorough investigation of α-induced spallation has been reported to date. The purpose of the present study is to provide such information and, by comparison with proton results, elucidate further features of high-energy nuclear reactions of complex nuclei.

II. EXPERIMENTAL PROCEDURES

Irradiations were performed at the Space Radiation Effects Laboratory[14] (SREL) synchrocyclotron in Newport News, Virginia. Targets consisted of a stack of three 13.0 μm aluminum foils serving as guard, monitor, and catcher, a single 15.0 or 51.0 μm high purity copper foil, and a final 13.0 μm aluminum catcher foil. Subsequent to irradiation in the internal beam at a radius corresponding to the desired energy, either a circular stack of discs was punched from the target stack 1 to 2 mm from the leading edge or, in the case of aligned target stacks, a 1 cm section was removed. The center aluminum foil was used to monitor the beam flux by means of the ^{27}Al(α, α 2pn)^{24}Na reaction and the forward and backward catchers were used to determine recoil loss corrections.

Both monitor and target from a given irradiation were assayed by direct γ counting under identical geometry on a high-resolution Ge(Li) detector at Carnegie-Mellon University. No products shorter lived than ~3 h were determined. Because of the experimental approach employed, uncertainties associated with chemical yields and counting geometries are eliminated and those associated with relative detector efficiencies are at a generally reduced level. All relevant decay data listed in Table I were acquired from Lederer, Hollander, and Perlman[15] or more recent references where noted.

In a separate study of monitor reactions, irradiated aluminum foils were counted in a calibrated 7.6 cm $\times$ 7.6 cm NaI(Tl) detector at SREL soon after bombardment.

III. RESULTS

A. Monitor reactions

The ^{27}Al(α, α 2pn)^{24}Na cross section has been previously measured up to energies of 380

MeV.[16–18] Church recently reported but did not document a value at 900 MeV.[12] However, the $^{27}Al(\alpha, X)^{18}F$ cross section has been reported at 920 MeV by Radin.[19] A combination normalization-interpolation using new data provides a value at 720 MeV. The $^{24}Na/^{18}F$ production ratio from aluminum has been measured from 410 to 720 MeV and the results listed in Table II. Normalization of the ^{24}Na production data at 410 MeV was accomplished by using the 380 MeV value. The $^{24}Na/^{18}F$ ratio then provides a ^{18}F production cross section value at 410 MeV. This was assumed to connect smoothly to the existing value at 920 MeV. An interpolation of absolute monitor cross sections can now be performed between the 380 and 920 MeV values using the $^{24}Na/^{18}F$ data. Figure 1 shows the relevant cross sections from this work and from other investigations at lower energies.

TABLE I. Relevant properties of nuclides measured (Ref. 15 unless otherwise noted).

Nuclide	Half-life	Radiation measured (keV)	Fractional abundance
^{7}Be	53.6 day	477	0.103
^{22}Na	2.6 yr	1275	1.00
^{24}Na	15 h	1369	1.00
^{43}K	22.4 h	373	0.85
^{42}K	12.4 h	1524	0.18
^{43}Sc	3.9 h	375	0.22
$^{44}Sc^{m}$	2.44 day	271	0.86
^{46}Sc	84 day	889	1.00
		1120	1.00
^{47}Sc	3.35 day	160	0.73
^{48}Sc	1.83 day	1038	1.00
^{48}V	16.2 day	983	1.00
		1313	0.97
^{48}Cr	23 h	116	0.98
^{51}Cr	27.8 day	320	0.098 [a]
^{52}Mn	5.6 day	745	0.88 [a]
		935	0.94 [a]
		1435	1.00
^{54}Mn	303 day	835	1.00
^{56}Mn	2.58 h	1811	0.29
^{52}Fe	8.2 h	165	1.00
^{59}Fe	45 day	1099	0.565 [a]
		1292	0.432 [a]
^{55}Co	18 h	1408	0.18 [a]
^{56}Co	77.3 day	847	1.000
		1238	0.666
^{57}Co	267 day	122	0.87
^{58}Co	71.3 day	811	0.99
^{60}Co	5.26 yr	1173	1.00
		1332	1.00
^{57}Ni	36 h	1378	0.849 [a]
^{61}Cu	3.4 h	284	0.11 [a]
^{64}Cu	12.8 h	1346	0.005
^{62}Zn	9.13 h	597	0.22
^{65}Zn	245 day	1115	0.49

[a] Reference 44.

TABLE II. Aluminum monitor reaction cross sections.

Cross section ratio E(MeV)	$^{18}F/^{24}Na$	Absolute cross section $^{27}Al(\alpha, \alpha 2pn)^{24}Na$	$^{27}Al(\alpha, X)^{18}F$
380–410	0.66 ± 0.04	24.2 ± 0.3 [a]	16.0 ± 2.0 [b]
600	0.74 ± 0.05	19.7 ± 2.4 [c]	14.6 ± 2.1 [c]
720	0.66 ± 0.05	20.9 ± 2 [c]	13.8 ± 1.7 [c]
900–920		22.0 ± 2.0 [d]	12.5 ± 0.5 [e]

[a] Reference 14.
[b] Normalized to ^{24}Na production at 380 MeV.
[c] Interpolated from a, b, and e.
[d] Reference 10.
[e] Reference 17.

Justification for this procedure lies in the generally smooth behavior of the excitation function for nuclear reactions at energies high above their thresholds. This behavior is exemplified by the gently sloped proton monitor cross section excitation functions also shown in Fig. 1. Table II lists the value obtained from this analysis of monitor reaction yield and used for subsequent absolute cross section calculations.

A separate measurement of the production rate of ^{18}F and ^{24}Na from an aluminum stack with and without a 51 μm-thick copper foil was performed to ascertain the magnitude of secondary reaction corrections to the monitor reaction. In Table III are the results of this study which show that, within the uncertainty of the determination, there is no discernible secondary correction to the monitor reaction for the target thicknesses employed in the spallation studies.

B. Spallation reactions

The results for production yields of copper spallation are listed in Table IV. Only one set of de-

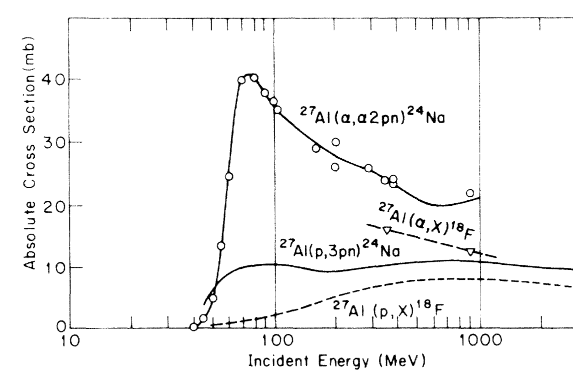

FIG. 1. Aluminum monitor reaction excitation functions. Incident proton data taken from Ref. 20; incident α data taken from Refs. 16–19. $^{27}Al(\alpha, \alpha 2pn)^{24}Na$ and $^{27}Al(\alpha, X)^{18}F$ values interpolated from this work and literature values (see text).

terminations was made at 410 MeV. At 720 MeV duplicate measurements were obtained and the number of independent determinations is listed in parentheses following each cross section value. The quoted uncertainty limits are root-mean-square combinations of individual uncertainties associated with summing corrections, recoil corrections, decay curve analyses,[21] and relative photopeak efficiencies. For multiple determinations, the weighted average cross sections and weighted standard deviations are given.

IV. DISCUSSION

High-energy nucleon-induced reactions[22-24] are generally envisioned as proceeding via the Serber model: a series or cascade of nucleon-nucleon collisions, followed by deexcitation through evaporation, fission, and γ emission. The cascade step is justified by the impulse approximation, that is, for situations where nucleon binding energies are small compared to kinetic energies and when cascade nucleon wavelengths are small and mean free paths large compared to nucleon-nucleon spatial separation within the nucleus. Experimental nucleon-nucleon scattering data are used as input for theoretical calculations as in the Monte Carlo method.[3] For α-induced reactions, experimental α-nucleon scattering data are scarce and the substructure of the incident projectile complicates calculations.

Using existing nucleon-nucleon data, the total reaction cross sections for 590 MeV protons and 720 MeV α particles on ^{64}Cu have been calculated[25] using a method similar to the Fernbach, Serber, and Taylor semiclassical optical model.[26] The nucleon density distribution for ^{64}Cu is analytically approximated by a flattened modified Gaussian density distribution as illustrated in Fig. 2. The half-central density radius and the 50–10% fall off distance have been set equal to the electron scattering result values of $c = 1.07A^{1/3}$ and $\frac{1}{2}t = 1.2$ fm.[27] The root-mean-square radius for natural Cu is 3.88 ± 0.05 fm[28] compared to 3.85 fm in the model. The α particle density distribution was

TABLE III. Secondary monitor effect.

Energy (MeV)	Total thickness Al (μm)	Total thickness Cu (μm)	^{18}F/^{24}Na cross section ratio
720	13	0	0.66 ± 0.04
	13	51	0.65 ± 0.04
410	13	0	0.66 ± 0.05
	13	51	0.65 ± 0.05

TABLE IV. Cross sections (in mb) formed in the α spallation of copper.

Nuclide	$Z - Z_A$	410 MeV	720 MeV
^{65}Zn		3.40 ± 0.018	1.02 ± 0.25 (1)
^{62}Zn			2.24 ± 0.27 (1)
^{64}Cu			33.0 ± 3.0 (2)
^{61}Cu			37.8 ± 2.9 (1)
^{57}Ni	2.175	1.67 ± 0.10	1.37 ± 0.05 (3)
^{60}Co	−0.239	12.7 ± 1.3	15.8 ± 2.0 (3)
^{58}Co	+0.575	63.6 ± 3.5	50.7 ± 1.6 (3)
^{57}Co	+1.175	54.6 ± 3.0	41.2 ± 1.3 (3)
^{56}Co	+1.376	18.1 ± 1.3	14.4 ± 0.5 (3)
^{55}Co	+1.814	2.82 ± 0.45	2.10 ± 0.16 (3)
^{59}Fe	−0.800	2.44 ± 0.16	2.65 ± 0.09 (3)
^{52}Fe	+2.335		0.232 ± 0.018 (1)
^{56}Mn	−0.624		6.72 ± 0.63 (1)
^{54}Mn	+0.353	35.1 ± 1.7	30.8 ± 0.9 (3)
^{52}Mn	+1.335	17.6 ± 1.4	15.6 ± 0.7 (3)
^{51}Cr	+1.130	37.3 ± 2.0	38.4 ± 1.3 (3)
^{48}Cr	+2.442	0.287 ± 0.018	0.373 ± 0.013 (3)
^{48}V	+1.442	13.3 ± 1.0	17.2 ± 0.7 (3)
^{48}Sc	−0.558	0.609 ± 0.048	0.819 ± 0.034 (3)
^{47}Sc	−0.368	2.38 ± 0.13	3.79 ± 0.12 (3)
^{46}Sc	+0.127	5.54 ± 0.34	9.17 ± 0.30 (3)
^{44}Sc^m	+0.850	3.66 ± 0.20	6.52 ± 0.20 (3)
^{44}Sc^g	+0.850		4.80 ± 1.3 (1)
^{43}Sc	+1.244		4.59 ± 0.62 (1)
^{43}K	−0.756	0.57 ± 0.04	1.21 ± 0.05 (3)
^{42}K	−0.265		3.47 ± 0.38 (3)
^{24}Na	−0.605		0.40 ± 0.03 (1)
^{22}Na	+0.638		0.58 ± 0.06 (1)
^{7}Be			3.65 ± 0.53 (1)

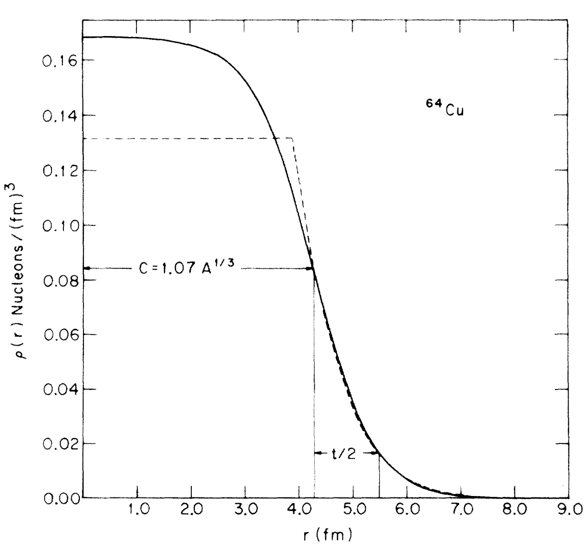

FIG. 2. Nucleon density distribution for ^{64}Cu used in calculating (Ref. 25) total reaction cross sections. The truncated-Gaussian (broken curve) was designed to match the Fermi I distribution density at $r = c = 1.07A^{1/3}$ and at the 10% central density point, thereby reproducing the appropriate nuclear surface texture

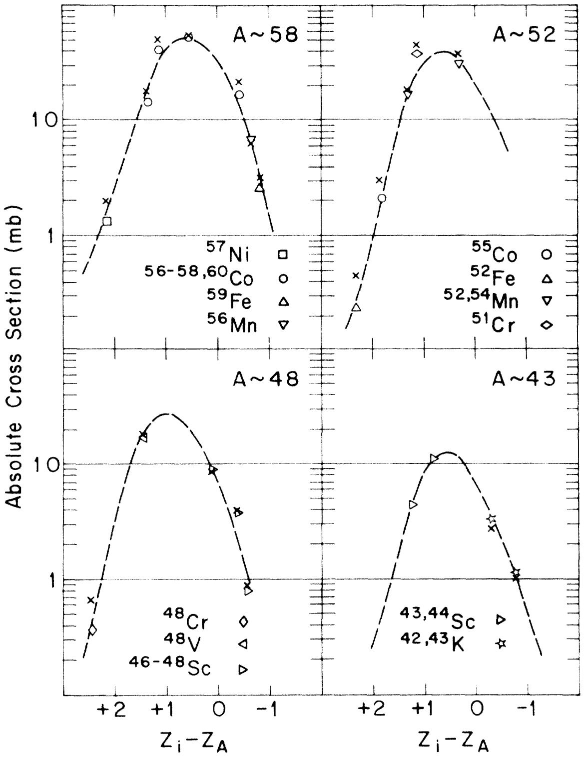

FIG. 3. Absolute isobaric yield distributions as a function of distance from β stability (Z_A) for 720 MeV α particle induced spallation of copper for mass chains A ~58,52,48, and 43. Crosses represent 590 MeV proton data of Refs. 37 and 38 multiplied by a factor of 1.9 for comparison with α results.

assumed to be Gaussian with a root-mean-square radius of 1.63 fm.[29] For 590 MeV protons the total reaction cross section[25] is 690 mb as compared to 667 ± 67 mb at 240 MeV.[30] The calculated total reaction cross section[25] of α particles plus copper amounts to 1300 mb. No comparable literature value is available. The α-to-proton total reaction cross section ratio for a copper target is thus 1.9. This factor is also reflected in the partial cross sections as discussed below.

Monte Carlo results of Gabriel *et al.*[31] for ^{93}Nb using a step density distribution show the α/p total reaction cross section ratio to be 1.9 at 720 MeV. Although their comparison was with 720 MeV protons their cross section ratio will not have been much smaller if 590 MeV protons were considered.

Absolute isobaric yield curves at A ~58, 52, 48, and 43 constructed directly from the tabulated results are illustrated in Fig. 3. The abscissa $Z-Z_A$ was chosen for convenience. Z_A represents the most stable proton number associated with isobars of mass A and was calculated by a parabolic fit to experimental ground state masses for each mass chain. For some lighter masses, it was necessary to resort to semiempirical masses of Garvey *et al.*[32] to substitute for nonexistent data in the Z_A calculation. There is no mechanistic interpretation necessarily implied by this choice of abscissa since the rigorous significance of the position of the stability valley relative to a given spallation reaction product is a complicated, un-

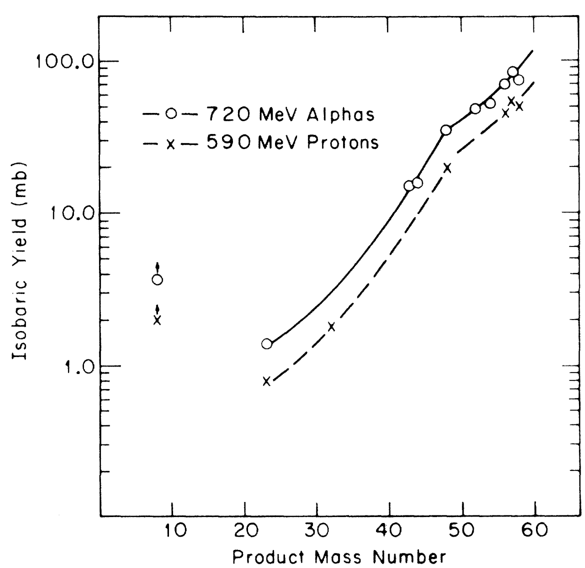

FIG. 4. Mass yield curves obtained from Fig. 3 for 720 MeV α and 590 MeV proton induced spallation of copper.

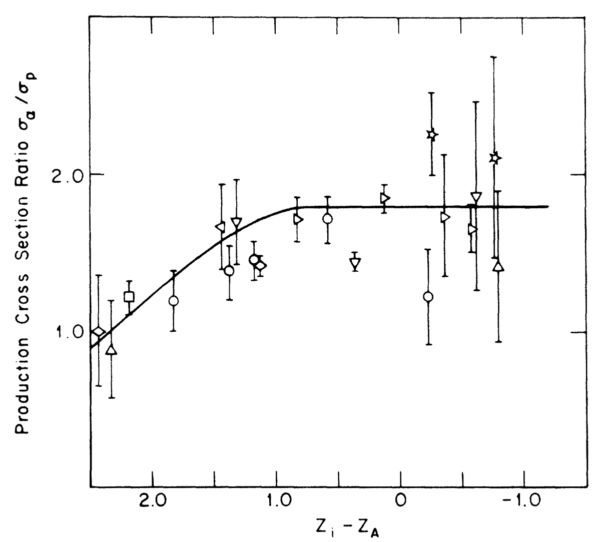

FIG. 5. Ratio of formation cross sections for a given product from copper irradiated with α particles and protons, σ_α/σ_p, as a function of that product's distance from the valley of β stability, $Z-Z_A$.

resolved subject. The values $Z-Z_A$ for each nuclide are listed in Table IV and are in reasonable agreement with Coryell's $Z-Z_A$ values[33] where such comparisons are possible. From the isobaric yield curves a mass yield curve is estimated as shown in Fig. 4. The total reaction cross section of copper with α particles appears to be 1.8 times greater than with protons as estimated from Figs. 3 and 4, in good agreement with a similar estimate based on the $p+{}^{93}Nb$ and $\alpha+{}^{93}Nb$ spallation studies at 320–800 MeV of Korteling and Hyde[10] and with the semiclassical calculation. Included for comparison are 590 MeV proton data data.[37,38,45] Other than magnitude shifts, isobaric yield curves for α and proton reactions seem remarkably similar in shape. A systematic difference is suggested, however, by examination of the formation ratio of α- to p-induced products σ_α/σ_p, as a function of position on the isobaric yield curve represented by distance from β stability. The comparison is illustrated in Fig. 5. Products on the neutron deficient wing of the isobaric yield distributions appear to be suppressed in α-induced relative to proton-induced reactions. This effect is not reflected in the mass yield curves since it is evident only in low-yield species.

Since for products far removed from the target, excitation functions generally are rising quite rapidly up to a few GeV, one asks whether the phenomenon evident in Fig. 5 is an artifact of where on the α and p excitation functions comparison is made. Data in Table IV, however, indicate that neutron deficient yield suppression is conspicuous even for products near the target (e.g. ${}^{55,56}Co$) where the excitation function varies slowly with energy for both α and p although reliable data at other energies are scarce. Continuing with the interpretation of Fig. 5, consideration should be given to the possibility that the cascade phases for α particles and protons on copper are sufficiently different (insofar as the ratios of emitted cascade neutrons to protons are concerned) that the isobaric yield curves are shifted more towards stability in the α cases. Two particulars disfavor this explanation though. First, the effect under consideration is not obvious when comparing proton data over a range of energies sufficiently wide to encompass an assortment of cascade histories.[34-39] Secondly, the effect is also apparently absent in the present α-proton comparison for the neutron excess products. Also related to spallation cascade histories is the investigation by Porile and Church[40] which suggests that the isobaric yield peak position and shape are strongly correlated with the target N/Z (i.e. $Z-Z_A$) value. In particular, the product distribution retains some "memory" of the target structure by being more neutron deficient for targets with low N/Z and *vice versa*. Copper plus an α particle as an initial transient species would have a slightly higher N/Z than copper plus a proton. A naive extension of the arguments of Porile and Church concerning cascade-evaporation memory would therefore predict an increased tendency towards yielding neutron excessive species from α-induced reactions. A very similar interpretation was recently invoked by Garrett and Turkevich[41] for their comparison of π^+-, π^--, and p-induced spallation of copper. The π^--induced spallation, proceeding through an initial species with a relatively large N/Z, resulted in reduced yields of neutron deficient products relative to production by π^+ and protons. Monte Carlo cascade-evaporation calculations seem to corroborate this explanation. In order to establish the import of this interpretation for α-induced spallation, detailed Monte Carlo calculations such as attempted by Gabriel *et al.*[31] would need to be examined.

Finally, an alternative explanation is the effect of differing cascade-residue angular momenta distributions on product charge distribution. The maximum angular momentum for the $\alpha+Cu$ reaction is (classically) ~$85\hbar$ while for $p+Cu$ it is ~$35\hbar$. The Monte Carlo results of Chen *et al.*[7] show that, in the case of ${}^{75}As+378$ MeV protons, the average angular momentum of cascade residues is about half of the maximum possible value. Although no existing calculations provide angular momenta distributions of cascade residuals from both α and proton reactions, it would seem plausible that the α-produced residuals would be left on the average with much more angular momentum than the same proton-produced residual. This being so deexcitation by α-particle (or other heavy ion) evaporation and by γ emission would, for α-produced cascade residues, become much more favorable at the expense of neutron evaporation.[42,43] The net result would be a decreased yield, relative to proton-induced spallation, for the neutron deficient nuclides in agreement with the observed results.

V. SUMMARY

Spallation of copper by high energy α particles has been examined radiometrically. In comparison to proton spallation, the α-induced products are formed with 1.8 times greater yield and similar isobaric yield distributions except for the very neutron deficient products. For these, the $\alpha+Cu/p+Cu$ production cross section ratio approaches unity. Energy dependence of cross sections, isobaric yield curve shifts, fragmentation and angular momentum considerations have all been briefly considered for their relevance to the observed effect.

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