Photonuclear Reactions in Iron and Aluminum Bombarded with High-Energy Electrons*

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Previously reported evidence for photofission of iron is investigated by a more extensive study. Thin foils of iron were bombarded with beams of 1.5-, 3-, 5-, and 16-GeV electrons. γ-ray spectroscopy and radiochemical measurements were used to measure yields of radionuclides produced in the targets. The yields of 25 nuclides were measured for the bombarded iron foils; these include eight nuclides in the mass region $22 \leq A \leq 35$. A thin aluminum target was bombarded with 16-GeV electrons, and the yields of seven radionuclides were measured. The yield of ^{24}Na in a thick iron target was measured as a function of target thickness and compared with that of radionuclides produced by cascade-evaporation reactions. The experimental evidence obtained in this series of experiments indicates that nuclides of mass <35 produced in the iron targets are the result of a fissionlike process. For cascade-evaporation reaction products, there is a decrease in the variation of yield with ΔZ as the bremsstrahlung energy is increased.

I. INTRODUCTION

When high-energy electrons pass through matter, photonuclear reactions are induced.[1] One method of studying these reactions is to measure the yields of radioactive nuclides produced. A number of such studies have been reported for bombarding energies of up to 320 MeV.[2-8]

The work reported here is part of a study of yields of radionuclides from photonuclear reactions produced by bombarding energies in the GeV energy region. In a preliminary report,[9] evidence for photofission of iron was presented. The present paper deals with a more extensive investigation of this phenomenon.

II. EXPERIMENTAL

Bombardments at energies of 1.5, 3.0, and 5.0 GeV were made at the Cambridge electron accelerator (CEA). The targets, metal foils about $0.015X_0$ thick (X_0 = radiation length), were positioned in air on the downstream window of a secondary-emission beam current monitor. The material upstream from the target consisted of a stainless-steel window (thickness $4.2\times10^{-3}X_0$) and the secondary-emission monitor ($9.6\times10^{-3}X_0$). During each bombardment, a record was made of the integrated beam current, which ranged from 3 to 15 $\times10^{14}$ electrons. After bombardment, the targets were transferred to the Oak Ridge National Laboratory via air freight. At times ranging from 9 to 40 h after bombardment, initial measurements of the γ-ray spectra were made with a Ge(Li) detector. Sufficient measurements were made to permit identification of nuclides by half-life as well as by γ-ray energy. Spectra were recorded on a multichannel pulse-height analyzer and listed and plotted by a computer. Each spectrum and data list was used to measure the counts per unit time in the photoelectric peaks of interest. The absolute detection efficiency and published decay-scheme data were used to convert peak intensities to source strengths for each radionuclide.

During the course of this work, it was deemed desirable to measure yields of some short-lived reaction products. Since an appreciably higher beam intensity is available at the Stanford linear accelerator, a few bombardments were made with a 16.0-GeV beam at the Stanford Linear Accelerator Center. The targets were positioned in air about 100 ft upstream from a water-tank beam

dump. Bombardments were made with a beam that, far upstream, had passed through a ~$0.01X_0$-thick target set up for other experimenters. Direct beam current monitoring was not available, but estimated integrated beams of ~10^{15} electrons were used for each bombardment. The available beam intensity permitted these bombardments to be made in about one minute. γ-ray spectra were measured with a Ge(Li) spectrometer at times as short as six minutes after bombardment. This permitted the detection of short-lived nuclides that could not be observed in the case of the CEA bombardments.

In a few instances chemical separations and β counting were necessary to measure yields of nuclides that decay by β emission without accompanying γ rays. Chemical separations were also made to verify the atomic numbers of some of the nuclides detected by γ-ray spectral measurements.

III. RESULTS AND DISCUSSION

The yields of several nuclides produced in iron targets at various electron bombarding energies are plotted in Fig. 1 as a function of the nuclidic mass. The abscissa scale at the top is based on the assumption that the target is ^{56}Fe, although that isotope makes up only 91.7% of natural iron. The ordinate in Fig. 1 is indicated as a cross section. For the 1.5-, 3-, and 5-GeV data, plotted values are based on the number of electrons in the integrated beams used in each bombardment. As mentioned previously, beam current monitoring was not available for the 16-GeV data; instead these yields are normalized to the CEA data for nuclei with masses ≥ 43.

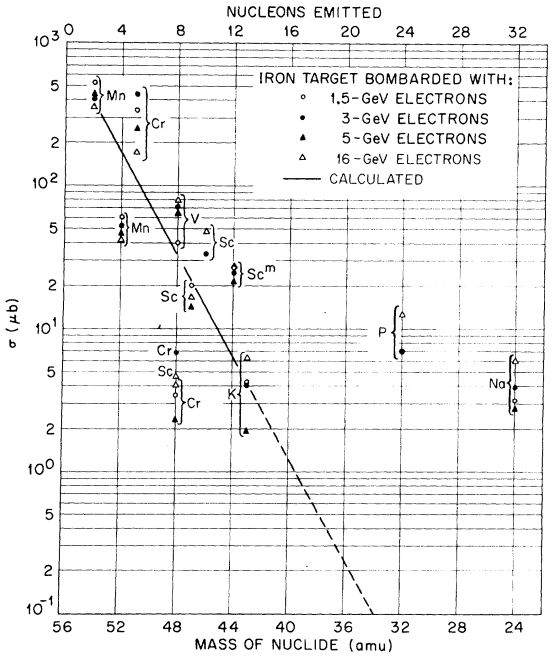

FIG. 1. Yields of radionuclides identified in iron samples that were exposed to beams of 1.5- to 16-GeV electrons. The values of σ are based on the integrated electron beam intensity. The "calculated" line is, in reality, a "calculated slope" as is described in the text.

In the region of $A \geq 43$ the yields exhibit an exponential decrease with the number of nucleons emitted from the target nucleus. The yields of ^{24}Na and ^{32}P are, however, much larger than would be expected unless the mechanism for production of these isotopes is different from that for nuclides with $A \geq 43$. The curve shown in Fig. 1 is an exponential drawn through a plot of calculated cross sections determined from the cascade-evaporation calculations of Bertini[10] for the production of residual nuclei by nucleon-induced reactions. Cross sections calculated at 50-MeV intervals were then multiplied by the intensity of the bremsstrahlung spectrum[11] at each energy. The resulting products were summed for each mass number to yield the plot through which the curve, again normalized to the experimental data for $A \geq 43$ of Fig. 1, was drawn. Although the absolute magnitude of the calculated cross section for nucleon-induced reactions is about three orders of magnitude larger than that of the measured cross sections in Fig. 1, the falloff of yield with mass of the nuclide is in reasonable agreement with that of the data down to mass 43.

The ^{24}Na yield is more than a factor of 10^3 larger than that expected from extrapolation of the curve based on the cascade-evaporation calculations. The ^{32}P yield is also much larger than expected. The reaction mechanism for production of nuclides in the mass region of ^{24}Na and ^{32}P is apparently different from that for production of nuclides of mass ≥ 43 in iron targets. A spectrochemical analysis of a piece of the iron foil used as target material indicated the presence of impurities that can account for less than 1% of the ^{24}Na production if it is assumed that impurities near mass 24 have cross sections for production of ^{24}Na comparable to that of aluminum.[12] The most reasonable explanation for the relatively large yields of ^{24}Na and ^{32}P appears to be that they result either from fragmentation or from photofission of iron nuclei. If the mechanism were fragmentation, a corresponding high yield of ^{7}Be, the one measurable nuclide of low mass, would be expected. On the contrary, no ^{7}Be was detected. It has been pointed out that any nucleus will undergo fission if it is supplied with sufficient excitation energy.[13,14] The fission barrier for iron is ~50 MeV,[15] therefore many photons of the bremsstrahlung spectrum from electrons in the GeV energy region have enough energy

to produce fission in iron.

The data of Fig. 1 with 3-GeV electrons were obtained first in the work reported here. Subsequent measurements were motivated in part by an interest in further exploring the evidence for fission of iron.

In an earlier paper[16] we reported measurements of yields of nuclides in thick targets bombarded with 3-GeV electrons. For an iron target it was observed that yields of nuclides for $A > 43$ peak at target thicknesses that decrease with the number of charged particles emitted from the target nucleus. Those results, on the basis of shower theory,[17] imply that the dominant photon energy increases with the ΔZ of the reaction. This is in agreement with the nuclear star measurements of Roos and Peterson,[18] which showed the prong number of stars (i.e., number of charged particles emitted) to increase with bremsstrahlung energy.

Measurements were made of the ^{24}Na yield in a thick iron target bombarded with 3-GeV electrons. Its yield as a function of target thickness is compared in Fig. 2 with that for four other nuclides produced in a thick iron target. As argued above, nuclides of mass ≥ 43 are presumed to be produced by cascade-evaporation reactions. The ^{24}Na yield curve is similar in shape to that of the other nuclides; it peaks, however, at a target thickness

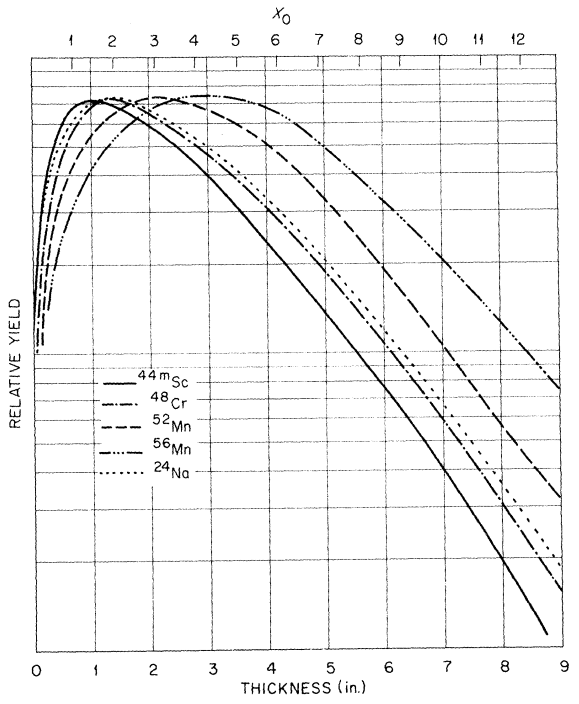

FIG. 2. Relative yields of some nuclides produced in a thick iron target bombarded with 3.0-GeV electrons. The curves are normalized to the same ordinate value at the peaks.

larger than that of the ^{44}Sc yield curve. The production of ^{44}Sc from an iron target involves the emission of five protons, while the nuclear charges of iron and sodium differ by 15 protons. If the curve for ^{24}Na in Fig. 2 were to follow the pattern of peaking at a thickness that decreases with ΔZ and ΔA observed for the manganese, chromium, and scandium isotopes, it should peak at a very small target thickness. In addition, the yield curve for ^{24}Na at larger target thicknesses should fall below those of the other products shown in Fig. 2. The observed ^{24}Na yield curve, however, falls close to that of ^{48}Cr. This is further evidence that ^{24}Na is produced in iron targets by a reaction mechanism different from cascade evaporation.

For the bombardments at the Stanford linear accelerator, it was possible to obtain γ-ray spectra of the targets soon after bombardment; thus yields of some of the shorter lived nuclides were measured. Some of the data obtained in the 16-GeV bombardments of iron are shown in Fig. 3. The data are displayed in a manner which was used by Halpern *et al.* for 320-MeV data,[19] designed to exhibit their regularity. The family of curves shown in Fig. 3 may be regarded as a yield surface for cascade-evaporation reactions. The construction of the family of curves is described in Appendix A. Some of the features are as follows: (1) There is one curve for each element, i.e., each value of ΔZ; (2) The curves peak ~0.2 mass units on the neutron-deficient side of the valley of β stability; (3) Nuclides near the β-stability line are produced with the largest probability; (4) Yields for various ΔZ lie on similar curves; and (5) Corresponding points on the curves are separated by a constant factor estimated to be about 1.6 in yield for each unit of ΔZ. This yield-surface description agrees well with data for $A \geq 44$. For nuclides of lower mass and $\Delta Z \geq 5$, the observed yields are larger than those indicated by the yield-surface plot. In particular, the yields for ^{32}P and ^{24}Na show a sharp departure from the pattern.

In Fig. 4 yields of radionuclides obtained from a 16-GeV bombardment of aluminum[12] are plotted in the same manner as that used for the iron target (Fig. 3). The observed sodium and fluorine yields fit the yield-surface plot while the yields of lighter elements fall above the plotted curves. An exception is ^{13}N whose excited states are unstable with respect to particle emission. For this reason its measured yield is believed to be low. The relatively high yields of ^{15}O and ^{11}C may be regarded as possible evidence of photofission of aluminum. The high relative yield of ^{7}Be suggests, however, that these nuclides might also be formed by fragmentation. The aluminum-target data are also presented in this discussion principally to call

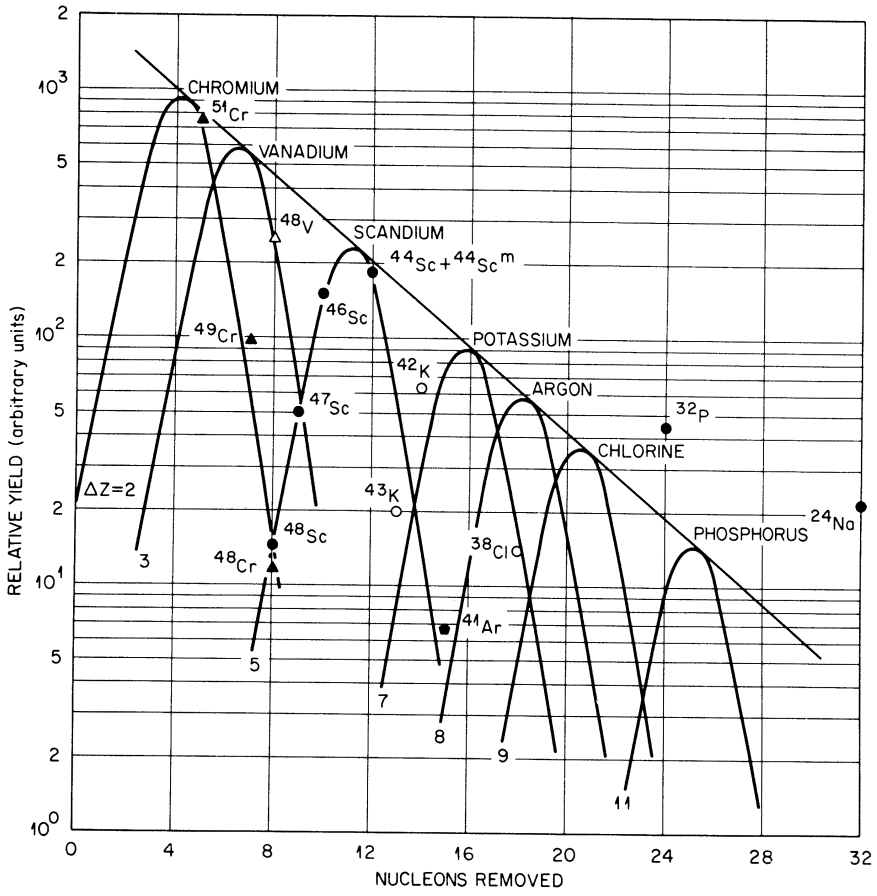

FIG. 3. Yields of radionuclides observed in a thin iron target bombarded with 16.0-GeV electrons versus the number of nucleons removed. The abscissa scale refers to ^{56}Fe.

attention to the relative yields of ^{24}Na and ^{22}Na. These nuclides are presumably produced in aluminum targets by cascade-evaporation reactions. Sodium 22 is nearer the valley of β stability, and its observed yield is indeed slightly larger than that of ^{24}Na.

The relative yields of all nuclides observed in 16-GeV bombardments of iron foils are plotted in Fig. 5. An effort was made to measure the yields of as many nuclides as possible in the mass region $A<35$. The value shown in Fig. 5 as an upper limit for ^{7}Be is the largest observed and was not reproducible. It could be ascribed to reactions in the oxygen impurity of the target foil. The upper limit for ^{7}Be produced in iron targets observed in the present work is in agreement with observations[18] of nuclear stars produced by bremsstrahlung-induced reactions in photographic emulsions. In that work, most of the star prongs were protons, a few were α particles, and none were reported for heavier particles.

An important result in the data presented in Fig. 5 is the relative yields of ^{22}Na and ^{24}Na. In contrast to the data obtained from aluminum, the yield of ^{24}Na is larger than that of ^{22}Na. This is further evidence that the yields of sodium isotopes in iron targets are not due to target impurities and are produced by reactions different from the cascade-evaporation reactions that produce sodium isotopes in aluminum targets (see Fig. 4). The low upper limit of the ^{7}Be yield is evidence that the sodium isotopes do not result from emission of fragments of $A=5-10$ from the target nucleus. The curve shown in Fig. 5 was obtained by calculations based on the compound-nucleus evaporation theory of Dostrovsky, Fraenkel, and Friedlander.[20] Reasonable agreement is obtained for nuclides of $A\gtrsim 40$. The yields of nuclides in the sodium to phosphorus mass region are appreciably larger than is predicted by the evaporation theory. It should be noted that in the preliminary report of this work,[9] the phosphorus yield was attributed to ^{32}P and ^{33}P. Subsequent measurements of decay curves of chemically separated phosphorus yields showed the yield is principally ^{32}P. This result agrees with the yield-surface description shown in Fig. 3. If that discussion is applicable, then the yield of ^{32}P relative to that of ^{33}P should be about the same as that of ^{46}Sc to ^{47}Sc, i.e., ~3 to 1. In summary then, the evidence presented in Figs.

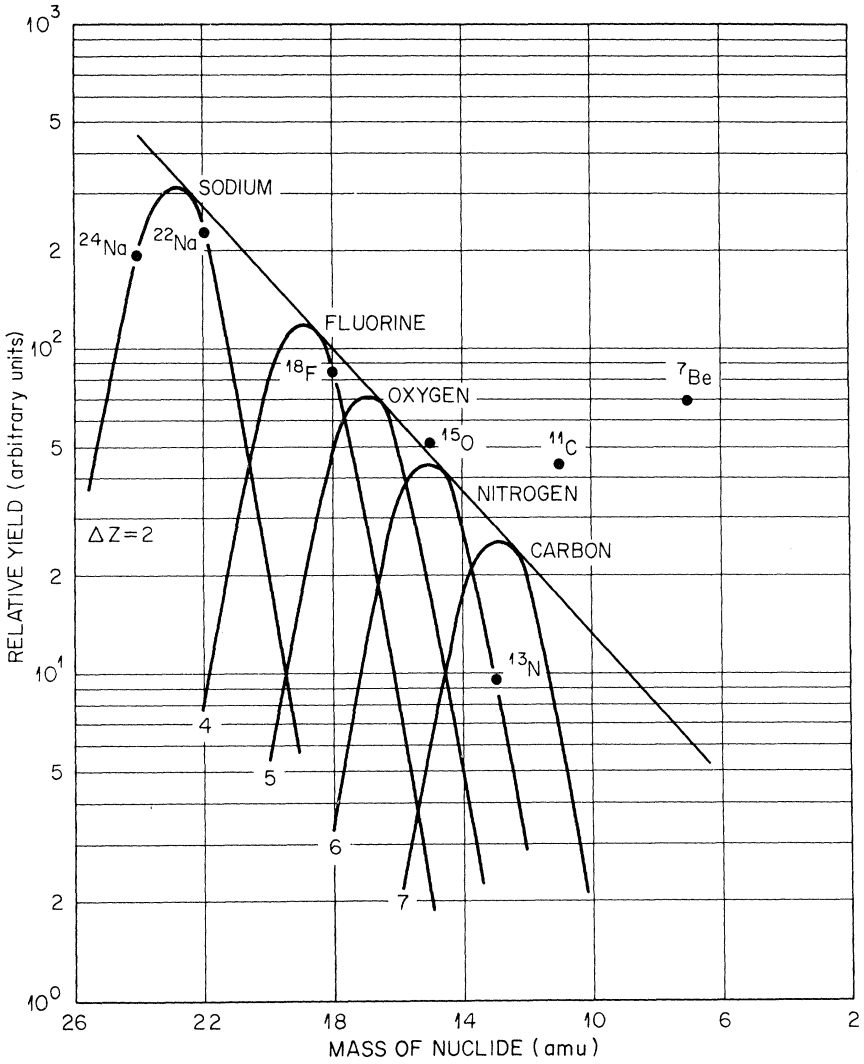

FIG. 4. Yields of radionuclides observed in a thin aluminum target bombarded with 16.0-GeV electrons versus mass of nuclides.

1–5 shows that the yields in the Na-P mass region observed in iron targets do not result from cascade-evaporation reactions.

As was pointed out above, reactions resembling fission would appear to provide the most reasonable explanation for the observed yields in the sodium-phosphorus region. Since the fission barrier for iron is ~50 MeV,[15] a large number of photons in the bremsstrahlung spectrum from GeV electrons have sufficient energy to induce fission in iron. With this explanation in mind, let us examine the yields in the mass region of $A < 40$ (Fig. 5).

Although ^{35}S is displaced only one half mass unit from the valley of β stability, the measured yield is low, perhaps because of the fact ^{35}S is on the upper wing of the fission yield curve. Similarly the low yield of ^{22}Na relative to that of ^{24}Na suggests that ^{22}Na is on the low wing of the fission yield curve. Its position on the upper wing of the fission yield curve may be another factor in the low yield of ^{33}P relative to that of ^{32}P.

The nuclide ^{28}Al is near the β-stable valley, and its yield is as large as that of any of the "fission products" observed in this work. Symmetric fission of ^{56}Fe without neutron emission, however, would yield two ^{28}Al fragments; therefore, for comparison with the other nuclides in this mass region, the ^{28}Al yield should probably be reduced by a factor of ~2. The nuclides ^{27}Mg, ^{28}Mg, and ^{29}Al are all one mass unit or more removed from the valley of β stability, and their observed yields are lower than those of ^{24}Na, ^{28}Al, and ^{32}P. If the yield of ^{28}Al is divided by 2 and compared with that of ^{29}Al, the ratio is about the same as that of the ratio of ^{47}Sc to ^{48}Sc, in agreement with the yield-surface description discussed above. The yields of ^{27}Mg and ^{28}Mg are almost equal, even though ^{28}Mg is about half a mass unit further re-

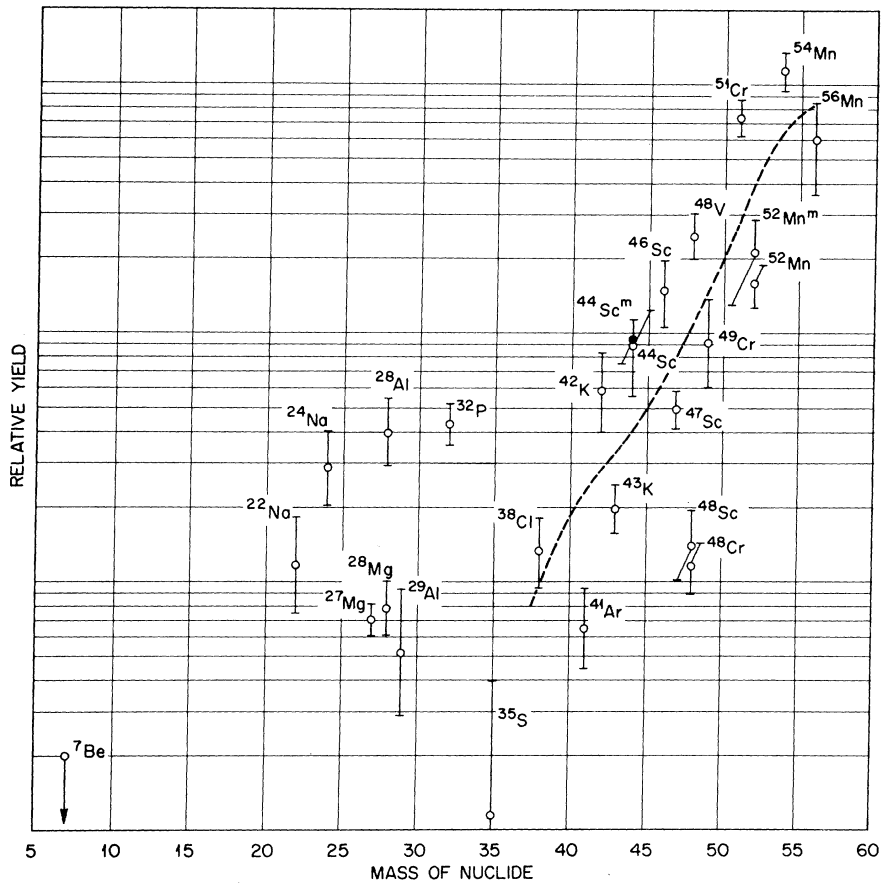

FIG. 5. Yield of radionuclides observed in a thin iron target bombarded with 16-GeV electrons versus mass of the nuclide. The dashed curve is the prediction of a nuclear evaporation theory (Ref. 20).

moved from the valley of β stability than ^{27}Mg. Apparently another effect is involved. Q values calculated by Schmitt[21] with the Wing-Fong mass formula[22] for fission of ^{56}Fe show fission to be energetically favored when the neutron numbers of both fragments are even. Thus, there are two effects involved: One favors a larger yield of ^{27}Mg while the other favors a larger yield of ^{28}Mg. While our data are not an exhaustive study, it would appear that the two effects are balanced for this case.

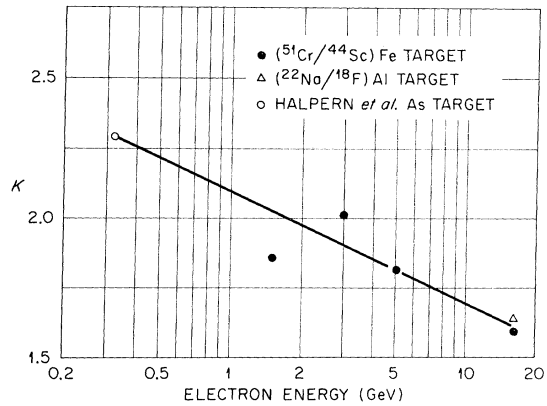

FIG. 6. Values of the yield-surface-ridge slope, K, as a function of bremsstrahlung energy.

Measurements of radionuclide yields from the interaction of 3- and 29-GeV protons with silver by Katcoff, Fickel, and Wyttenbach[23] showed an increase of the yield below $A \approx 35$. The yields in the region $15 < A < 35$ were attributed by the authors as probably arising from a "fissionlike" process. In contrast to our results where a very small upper limit for production of ^{7}Be in iron targets is observed, the yield of ^{7}Be reported for proton-induced reactions in Ref. 23 is relatively large. For proton-induced reactions the energy of the incident particle is involved in the reaction, while for bremsstrahlung-induced reactions a photon spectrum with $\sim 1/E$ distribution is involved. Also high-energy protons can produce rotational motion of the target nucleus. It has been shown[24] that rotational motion of the compound nucleus is an important factor in the emission of heavy fragments such as ^{7}Be.

Another explanation of the differences in the results of the present studies and those reported in Ref. 23 may be that iron is more fissionable than silver. An evaluation by Nix and Sassi[25] of fission barriers and particle binding energies based on the liquid-drop model indicated that fissilities of elements lighter than about silver should increase with decreasing Z^2/A. Their calculations show iron to be at least an order of magnitude more fissionable than silver. A large fission probably would result in fewer fragmentation reactions and therefore account for a low upper limit of the yield of ^{7}Be.

It was our hope that a sufficient number of yields could be measured to determine whether the apparent fission of iron is predominantly symmetric or asymmetric. The calculations of Schmitt[21] indicate that the fission modes energetically favored would yield stable fragments. These are not measurable by the technique used in the present work. Our data do indicate, however, that the fission mass yield for iron is largely in the region $A \sim 20$ -35.

IV. ENERGY DEPENDENCE

It was pointed out by Halpern *et al.*,[19] that the value of the slope K of the yield-surface ridge is difficult to determine accurately because the ridge is near the β-stability line where the yields are not observable by the techniques used in Ref. 19 or in the present work. The value $K \cong 1.6$ obtained for our 16-GeV data is somewhat lower than the value of 2.3 deduced by Halpern *et al.*[19] for data obtained from 320-MeV bremsstrahlung-induced reactions in arsenic. This difference suggests that K may be energy dependent. It is of interest to examine our data at other energies for evidence of this. The nuclides ^{51}Cr and ^{44}Sc are about equally displaced from the valley of β stability and hence are equally displaced from the ridge of the yield surface. The ratios of the yields ^{51}Cr/^{44}Sc for the bombarding energies included in our data are listed in Table I. For the 1.5-, 3.0-, and 5.0-GeV data, the 4.0-h ^{44}Sc yield was not measured. In the 16-GeV data (Fig. 5), the yield of ^{44}Sc and ^{44}Sc^m are about equal. Thus, the numbers shown in the table for 1.5, 3, and 5 GeV are the ratios of the ^{51}Cr yield divided by twice the ^{44}Sc^m yield. The resulting values of K are listed in the table and plotted in Fig. 6.

The nuclides ^{22}Na and ^{18}F are about equally displaced from the valley of β stability. Thus, the ratio of the two yields is used to obtain a value of K for the 16-GeV aluminum data. This is also listed in the table and plotted in Fig. 6. The values of K thus obtained for iron and aluminum for the 16-GeV data are in good agreement. We believe that the data plotted in Fig. 6 provide evidence of a decrease of the slope of the yield-surface ridge with increase of bremsstrahlung energy. The straight line drawn through the points plotted in Fig. 6 combined with the shape of the yield curve for a single value of ΔZ (Fig. 3) provide a means for estimating cross sections as a function of energy for targets in the Al-As mass region.

APPENDIX A

The family of curves shown in Fig. 3, which represent a yield surface for nuclides produced by the cascade-evaporation process, was constructed in the following manner. Yields were plotted as a function of the number of nucleons removed from the target nucleus (assumed to be ^{56}Fe). A smooth curve, symmetrical about its peak, was drawn through the scandium yield points. A curve of identical shape was then drawn through the chromium yield points. The peak of the scandium curve was observed to be a factor of ~0.25 lower than that of the chromium curve. This corresponds to a K (decrease of yield per unit increase of ΔZ) of ~1.6. To determine the positions of curves for other values of ΔZ, a straight line was drawn tangent to the chromium and scandium curves. This line is near and parallel to the ridge of the yield surface. Curves for other values of ΔZ were then drawn tangent to the straight line at intervals corresponding to $K = 1.6$.

TABLE I. Ratio of yields of nuclides used to determine values of K. ^{44}Sc yields include 4.0-h ^{44}Sc and 2.4-day ^{44}Sc^m, which for the 1.5-, 3.0-, and 5.0-GeV data were assumed to be twice the yield of ^{44}Sc^m.

Energy (GeV)	Target	Nuclides	Ratio of yields (experimental)	$\Delta Z_1 - \Delta Z_2$	K
1.5	iron	^{51}Cr/^{44}Sc	6.2	3	1.86
3.0	iron	^{51}Cr/^{44}Sc	8.4	3	2.03
5.0	iron	^{51}Cr/^{44}Sc	5.95	3	1.82
16	iron	^{51}Cr/^{44}Sc	4.0	3	1.59
16	aluminum	^{22}Na/^{18}F	2.7	2	1.64
0.32	arsenic		(Halpern *et al.*)[a]		2.3

[a] See Ref. 19.

The family of curves shown in Fig. 4 were drawn in the same manner as those of Fig. 3. A curve whose shape is the same as those of Fig. 3 was drawn through the sodium yield points. Curves for other values of ΔZ were then drawn such that K corresponds to a value of 1.6.

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Excitation Functions for the Reactions $^{34}S(p, n)^{34}Cl$ and $^{31}P(\alpha, n)^{34}Cl$†

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Excitation curves for $^{34}S(p,n)^{34}Cl$ and $^{31}P(\alpha,n)^{34}Cl$ were measured from the observed thresholds at 6438 ± 10 and 6360 ± 10 keV, respectively, to 1.6 MeV above threshold by detecting the residual radioactivity. Two prominent resonant structures were found in both reactions at the same excitation energies, 12.9 and 13.9 MeV, in the compound nucleus ^{35}Cl. Excitation curves for $^{31}P(\alpha,\alpha_0)$ were also measured in the region of these resonances, but no similar prominent resonances were found. Arguments are presented that indicate a $T=\frac{3}{2}$ assignment for these resonances. Absolute values of the total cross section were determined for the above (p,n) and (α,n) reactions and also for $^{34}S(p,n)^{34}Cl^m$ and $^{31}P(\alpha,n)^{34}Cl^m$.

I. INTRODUCTION

Proton scattering as well as proton-induced reactions have become a well-established tool for the observation of isobaric analog states. These states are generally studied by analyzing the compound-nucleus resonances which appear in excitation curves. The analog states observed in the proton-plus-target system have isospin $T_0+\frac{1}{2}\equiv T_>$, where T_0 is the target isospin. These states are