effect due to ionized clouds being ejected from the sun, then we might expect the density of ionized matter in the vicinity of the earth to change during a solar cycle also. Besides correlations with magnetic activity, one might expect to find correlations of cosmic-ray intensity with other phenomena which depend on the presence of ionized matter in the vicinity of the earth.

Although the solar influence is probably the most important, the possibility of other modulating mechanisms must be borne in mind. The situation of August, 1940, where 10% more cosmic-ray energy was coming to the earth than during the solar minimum of 1954, is cited as a case in point. This was not the solar-flare type of increase for it not only lasted for a number of days but it was measured at Huancayo, Peru also.

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Spallation of Yttrium by 240-Mev Protons*

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The individual cross sections of 36 nuclides resulting from the inelastic interaction of 240-Mev protons on yttrium were determined. The results were found to be in agreement with the current theories of high-energy nuclear reactions. By means of interpolated cross sections for the stable nuclei produced in the reactions, it was possible to estimate the total inelastic cross section for yttrium with 240-Mev protons. By use of this value and application of the optical model, r_0 for Y^{89} was found to 1.21×10^{-13} cm and the nuclear transparency about 17%.

INTRODUCTION

INVESTIGATIONS of the interaction of high-energy particles with complex nuclei are made to seek answers to the following: (1) the mechanism by which high-energy reactions take place, (2) the total inelastic cross section, (3) the distribution of energy transferred.

Two experimental approaches may be used. One is the study of the particles emitted in these reactions by means of emulsions, cloud chambers, and counter telescopes. The second is the radiochemical approach whereby the products formed are isolated chemically and identified.

In this study, yttrium was bombarded with 240-Mev protons, the reaction products were separated and their yields determined by the radiochemical approach. Inherent in this approach is the disadvantage that the primary process is not observed directly but must be deduced from the end products. However, it has the advantage that it is possible to detect the effects of the emission of neutral particles.

Other studies on the interaction of high-energy particles with complex nuclei, as approached by the radiochemical method, are given in a recent review.[1]

EXPERIMENTAL

Yttrium in the form of spectroscopically pure yttrium oxide[2] was irradiated in the University of Rochester 130-in. synchrocyclotron at a radius corresponding to 240-Mev protons. Each target was prepared by wrapping 100 mg of yttrium oxide in a 5-mil aluminum foil envelope, 3 mm×3 mm×3 cm, the latter dimension perpendicular to the radial direction of the beam.

Two 1-mil aluminum foils were placed before and after the target foil, of which the two inside foils served to monitor the integrated proton beam through use of the known cross section for the reaction $Al^{27}(p,n)Na^{24}$. A value was used for the cross section for this reaction of 11.0 mb, which is based on an interpolation of the values determined by Hintz and Ramsey[3] at 110 Mev

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[1] D. H. Templeton, *Annual Review of Nuclear Science* (Annual Reviews, Inc., Stanford, 1953), Vol. 2, pp. 93–104.

[2] The yttrium oxide, supplied through the courtesy of Ames Laboratory, Iowa State College, Ames, Iowa, was better than 99.9% pure.

[3] N. M. Hintz and N. F. Ramsey, Phys. Rev. 88, 19 (1952).

and Marquez at 420 Mev.[4] Each 1-mil aluminum foil was dissolved in 1.0 ml conc. HCl and a 0.1-ml aliquot absorbed on a filter paper disk 1.8 cm in diameter. The filter paper disk was then cemented to a stainless steel disk, which was used for counting. The 15-hour Na^{24} was determined in each case. In one run, a uniform mixture of the oxides of aluminum and yttrium wrapped in a 5-mil aluminum foil envelope, was bombarded, and the Na^{24} produced from the aluminum oxide was chemically separated. It was thus possible to compare the ratio of the yield of Na^{24} to that of various products from yttrium by these two different targeting arrangements. From the second method a correction can be obtained for the decrease in radial penetration of the proton beam through the target, as will be explained later.

The yttrium oxide target was placed in a distilling flask which contained known quantities of inert carriers of the elements niobium, zirconium, strontium, rubidium, bromine, selenium, arsenic, and germanium. The flask was connected to an all-glass line which had provision for flowing helium through the system. The target was dissolved by adding conc. nitric acid. The bromide carrier was oxidized to bromine, which was carried by the helium and absorbed in a CCl_4 trap held at 0°C. Radioactive krypton, produced from the yttrium, was carried by the helium through a series of dry ice traps and was finally adsorbed in a charcoal-filled trap at liquid nitrogen temperature.

The chemical separations for the remaining elements were based on modifications of standard procedures.[5] The fraction of the inert carrier recovered was determined in most cases by gravimetric analysis. Samples of each of the recovered elements were prepared by filtering a convenient form of the element through a glass chimney onto filter paper disks. Each disk was then glued to a stainless steel disk and covered with a few drops of Duco solution (2–3 g/cm^2).

All samples were counted with a methane-flow beta proportional counter. When the nature of the radiations permitted, a sodium iodide scintillation counter and an x-ray proportional counter were also used.

Each radioactive nuclide produced was identified by (a) its half-life from resolution of decay curves, (b) maximum beta-spectrum end points obtained with a survey beta spectrometer, and (c) chemical isolation experiments in certain cases.

Each counting rate was converted into a disintegration rate by correction for solid geometry and use of the decay scheme of the particular nuclide. For beta emitters, correction factors were applied for back scattering, self-absorption and self-scattering, and air and window absorption. The back scattering factors used in this work were determined experimentally by use of standard techniques.[6] The self-absorption and self-scattering factors were determined by use of the procedure outlined by Baker and Katz.[7] The factor for air and window absorption was determined in each case by the empirical relationship of Gleason, Taylor, and Tabern.[8] The solid angle of the source with respect to the counter was determined both by the use of Blachman's equation[9] and experimentally by use of calibrated sources. Also, by use of calibrated sources the sensitive volume of the counting chamber was determined and this effect was incorporated in the geometry factor.

The counting efficiency of the proportional counter for beta particles was taken as 100%, which was in agreement with calculations made on standard sources. The efficiency for gamma radiations was 1.5%, obtained from a calibrated source of Co^{60}. Finally, the x-ray counting efficiency was calculated both by using mass absorption coefficients and by intercalibration with the x-ray proportional counter and pulse-height analyzer.

All disintegration rates were calculated to infinite bombardment time and this saturation disintegration rate was compared with the saturation disintegration rate of Na^{24} from the aluminum monitor. From this comparison and taking into account the weight of the targets, the cross section was calculated for each product.

RESULTS AND DISCUSSION

The experimental values of the cross sections of the various products obtained from the spallation of yttrium at[10] 240 Mev are given in Table I. The results are an average of three to six separate determinations and the precision estimate is the average deviation of these multiple determinations. Of the 63 radioactive nuclides between zirconium and germanium that could feasibly be produced by the spallation of yttrium, 36 were definitely observed. This leaves 27 nuclides not found because of low cross sections or too long or too short half-lives, of which 65% were on the neutron excess side of stability.

Several general features are apparent from a study of Table I. First, the products observed lie predominantly on the neutron deficient side of stability. Comparison of the cross section of Ge^{77} with the other germanium isotopes indicates the low probability of making neutron excess nuclides, even when they are many mass numbers removed from the target. Secondly, it is seen that in the mass region between three and ten mass

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[6] L. Zumwalt, U. S. Atomic Energy Commission Report MDDC-1346 1947 (unpublished).
[7] R. G. Baker and L. Katz, Nucleonics **11**, No. 2, 14 (1953).
[8] Gleason, Taylor, and Tabern, Nucleonics **8**, No. 5, 12 (1951).
[9] B. P. Burtt, Nucleonics **5**, No. 2, 28 (1949).
[10] The interaction of yttrium with protons of 60, 100, 150, and 175 Mev will be presented in a forthcoming paper.

TABLE I. Cross sections for the formation of nuclides from the bombardment of yttrium with 240-Mev protons.

Nuclide	Gross reaction	Cross section (mb)
Zr^{89}	(p,n)	6.1 ± 0.7
Zr^{88}	$(p,2n)$	25.6 ± 2.0
Zr^{87}	$(p,3n)$	9.2 ± 1.0
Zr^{86}	$(p,4n)$	0.8 ± 0.2
Y^{88}	(p,pn)	93.0 ±15
Y^{87}	$(p,p2n)$	23.0 ± 4
Y^{87m} and Y^{86} [a]	$(p,p2$ or $3n)$	28.2 ± 5
Y^{85}	$(p,p4n)$	15.8 ± 3
Y^{84} and Y^{83} [a]	$(p,p5$ or $6n)$	2.3 ± 0.5
Y^{82}	$(p,p7n)$	0.08 ± 0.04
Sr^{87m}	$(p,2pn)$	5.0 ± 0.5
Sr^{85}	$(p,2p3n)$	59.5 ±10
Sr^{83}	$(p,2p5n)$	27.5 ± 4
Sr^{82}	$(p,2p6n)$	9.3 ± 5
Rb^{86}	$(p,3pn)$	4.3 ± 1
Rb^{84}	$(p,3p3n)$	71.4 ±12
Rb^{82} and Rb^{81} [a]	$(p,3p5$ or $6n)$	37.2 ±15
Kr^{79} [b]	$(p,4p7n)$	23
Kr^{77} [b]	$(p,4p9n)$	11.4
Kr^{76} [b]	$(p,4p10n)$	3.5
Br^{80}	$(p,5p5n)$	2.7 ± 0.7
Br^{77}	$(p,5p8n)$	4.3 ± 1.0
Br^{76}	$(p,5p9n)$	4.4 ± 1.0
Br^{75}	$(p,5p10n)$	3.8 ± 1.0
Se^{75}	$(p,6p9n)$	0.95 ± 0.2
Se^{73}	$(p,6p11n)$	3.5 ± 1.0
Se^{72}	$(p,6p12n)$	0.96 ± 0.3
As^{74}	$(p,7p9n)$	0.77 ± 0.3
As^{72}	$(p,7p11n)$	2.3 ± 0.5
As^{71}	$(p,7p12n)$	1.5 ± 0.5
Ge^{77}	$(p,8p5n)$	0.007± 0.005
Ge^{71}	$(p,8p11n)$	2.1 ± 1.0
Ge^{69}	$(p,8p13n)$	0.016± 0.01
Ge^{68}	$(p,8p14n)$	0.021± 0.01
	Total	483.5 ±86.5

[a] Value represents the sum for the two species. Separation difficult due to similar half-lives.
[b] Values based on the yield of Br^{76} grown in from a purified krypton fraction.

numbers from the target, the cross sections in general do not decrease rapidly.

Another interesting feature is found in a comparison of several isobaric pairs of nuclides. Yields of Rb^{82} are larger than those of Sr^{82}, and As^{72} yields are larger than those of Se^{72}. If the mechanism for the production of nuclides many mass numbers removed from the target involves the evaporation of one or more nucleons, especially in the last step, then nucleon binding energies, Coulomb barriers, and the level density of the product nuclides will be important factors in determining their yield. Since the level density[11,12] of the even-even nucleus is lower than that of the odd-odd nucleus, the odd-odd nucleus might be expected to have the higher cross section of the two nuclei comprising the isobaric pair, other factors being equal.

The general features of the spallation product distribution can be summarized as follows: (1) The maximum yield for an isotope of a given element is apparently one to two mass units on the neutron deficient side of stability, increasing slightly with increasing distance from the target, (2) From 3 to 7 units of Z lower than the target, the cross sections drop off smoothly. (3) At many nucleons removed from the target, the yield distribution includes nuclides on the neutron excess side of stability as well.

From these general features it is possible to obtain approximate values of the cross sections of the stable isotopes formed by spallation from interpolation of cross section *vs* mass number for a given Z, and cross section *vs* Z for a given isobaric chain. Smooth distributions, with maxima from 1.5 to 2 mass units on the neutron-deficient side, were assumed in each case except for Sr^{88}. For the latter, owing to the probably different mechanism for its production, a cross section equal to 1/1.28 times the Y^{88} cross section was assumed. This will be discussed later. The total cross sections in millibarns for each element, including the interpolated values, are as follows: Zr, 42; Y, 162; Sr, 250; Rb, 200; Kr, 82; Br, 21; Se, 8; As, 6; Ge, 2. Thus, the total inelastic cross section for 240-Mev protons on yttrium is 773±177 mb. The estimated error emphasizes the approximate nature of the value. It may be noted that the strontium isotopes account for the largest fraction of the cross section when the estimated yields of stable nuclei are included, though the largest observed activity is in the yttrium fraction.

As was mentioned previously, the ratio of the production of Na^{24} to various products from yttrium was measured by two different target monitoring arrangements, i.e., one with aluminum foil on the outside of the envelope and the other with aluminum oxide mixed uniformly with the yttrium oxide target. These gave a difference of about 11% in the yields. By means of the empirical relationship developed by Clark[13] for the radial penetration of the proton beam in the Rochester cyclotron, it was possible to calculate this effect on the target arrangement which had the external aluminum foils as monitors. This calculation gave a value of 14% for the correction, which is in good agreement with the value of 11% obtained from the internal monitor.

Comparison of Results with Optical Model

It is of interest to compare this experimental plus interpolated value of the total inelastic cross section with a calculation by the optical model[14] as has been done by Belmont and Miller.[12] The results of these calculations are given in Table II. From this table it can

[11] S. N. Ghoshal, Phys. Rev. **80**, 939 (1050).
[12] E. Belmont and J. M. Miller, Phys. Rev. **95**, 1554 (1954).
[13] D. L. Clark, Ph. D. thesis, Department of Physics, University of Rochester, 1952 (unpublished).
[14] Fernbach, Serber, and Taylor, Phys. Rev. **75**, 1352 (1949).

be seen that the experimental value of σ_a obtained in this work corresponds to an r_0 of 1.21×10^{-13} cm and a nuclear transparency of about 17%.

This value of 17% for the nuclear transparency of Y^{89} nuclei with 240-Mev protons compares with 13% obtained by Perry[15] for AgBr nuclei irradiated with 240-Mev protons, 35% for AgBr emulsions, obtained by Bernardini *et al.*,[16] with 380-Mev protons, and the values of 53%, 41%, and 31% for carbon, copper, and lead, respectively, irradiated with 270-Mev neutrons by De Juren.[17]

TABLE II. The absorption cross section, σ_a, for yttrium with 240-Mev protons calculated by means of the optical model as a function of r_0. The geometric cross section, ($\sigma_G=\pi r_0^2 A^{\frac{2}{3}}\times10^{-26}$ mb), and the percent nuclear transparency are also listed.

$r_0\times10^{13}$ (cm)	σ_a(mb)	σ_G(mb)	% transparency
1.0	553	625	11
1.1	660	757	13
1.2	755	904	16
1.3	870	1055	18
1.4	946	1220	22
1.5	1070	1400	24

Model for High-Energy Reactions

The over-all results of the spallation of yttrium are in agreement with current theories of high energy reactions.[18] The primary process is thought to consist of two phases: the knock-on phase, which gives rise to an internal nucleonic cascade and deposits a distribution of energies in the nucleus, followed by the evaporation phase, whereby the excited nucleus evaporates protons and neutrons until thermal equilibrium is attained. This mechanism explains the distribution of yields given in Table I. The maximum in the yields should be expected to occur close to the target element, as actually found, since the probability of an extended nucleonic cascade is low. The small amount of neutron-excess nuclides found is explained by the higher probability of evaporating neutrons compared to protons, forming neutron-deficient nuclides.

The knock-on mechanism explains the high cross section for the production of Y^{88}. The incident proton knocks out a neutron from Y^{89} leaving an Y^{88} nucleus with less than 10 Mev of excitation. This occurs in about 10% of the cases. By this mechanism, the ratio of the cross section for Y^{88} to Sr^{88} should be approximately equal to the neutron-to-proton ratio in Y^{89}. For this reason, the interpolated value of the Sr^{88} cross section was chosen as 1/1.28 times the Y^{88} cross section, as was mentioned previously. For the purposes of interpolation, differences in *pn* and *pp* scattering angular distributions were ignored.

About 5% of all reactions lead to the production of neutron-deficient zirconium nuclides. To produce these the incident proton knocks out one or more neutrons and is captured by the target nucleus. Part of the yield of the zirconium nuclides can be accounted for by charge exchange processes. Table I shows the trend in the yields of these nuclides as the number of neutrons lost increases; Zr^{88} is produced in highest yield while the nuclides on both sides are lower. This is probably due to the energy spectrum left behind in the excited nucleus remaining after the knock-on process. Thus, the yields fall off as one, two or three neutrons are evaporated, as might be expected from energy requirements. The effect of secondary particles on these yields was investigated in two experiments where the target mass was reduced by a factor of 5. There was no measurable effect.

The production of yttrium nuclides involves the loss of one proton, i.e., the incident proton and one or more neutrons up to seven for Y^{82}. A similar loss of many neutrons from the target element has been observed in other cases.[18] From Table I and the estimated total yields (including stable nuclides) it is seen that despite the large yield of Y^{88} and the large number of yttrium nuclides observed, the total yttrium cross section is less than that of either strontium or rubidium and actually amounts to only about 20% of the total inelastic cross section. Approximately one-half of the total yttrium cross section is accounted for by Y^{88}, which must be predominantly a knock-on product. The yield of the remaining yttrium nuclides shows a maximum at about Y^{87} and decreases with increasing loss of protons. This means that it is more probable for the excited yttrium nucleus to evaporate at least one or more protons rather than six or seven neutrons and no protons.

To account for the yield distribution of products shown in Table I, it is necessary to conclude that the residual nucleus, after the nucleonic cascade, is left with a wide spectrum of excitation energies. Based on calculations made by Le Couteur[19] it appears that it requires from 10 to 20 Mev of excitation energy for each particle evaporated. With this assumption, the results of this work indicate that the most probable amount of energy transferred to the struck nucleus is less than half the incident energy.

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