Cross Sections for Nuclear Reactions Involving Nuclear Isomers*

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Cross sections for the reactions $Cr^{52}(p,n)Mn^{52m}$, $Cr^{52}(p,n)Mn^{52}$, $Ag^{107}(p,pn)Ag^{106m}$, $Ag^{107}(p,pn)Ag^{106}$, and $Cu^{63}(p,n)Zn^{63}$ were determined as a function of energy with the 20.6-Mev proton beam of the U.C.L.A. synchrocyclotron. The relative yields of Sr^{85}, Sr^{85m}, and Sr^{87m} obtained in the proton bombardment of rubidium were also determined. In all cases the isomer yield ratios varied with energy over the entire energy range studied. The results are discussed in terms of conservation of angular momentum, assuming compound nucleus formation.

INTRODUCTION

ALTHOUGH much evidence has been cited in recent years to indicate that "direct interaction" is a much more probable mechanism in nuclear reactions than was previously assumed,[1–5] it is generally believed that below 30 Mev nuclear reactions proceed mainly through the formation of a compound nucleus. Underlying the theory of compound nucleus formation is the assumption that the compound nucleus decays independently of the way it is formed. When energetically possible, the compound nucleus will emit one or more particles and leave a residual nucleus, which may still be highly excited. Eventually, the residual nucleus will decay to the ground level by a gamma cascade, or, in the case of isomers, to the isomeric states.

When the incoming particle has a low energy it must have low orbital angular momentum in order to penetrate the centrifugal barrier, and so the compound nucleus will be formed with angular momentum not very different from that of the target nucleus. Since angular momentum must be conserved in the overall reaction, the isomer with spin closest to target nucleus will be favored at low energies. Recent experiments confirm this rule.[6–9]

At high energies transfer of angular momentum can be achieved by particle emission and gamma cascading (although particles are unlikely to transfer large amounts of angular momentum if a Maxwellian distribution of outgoing particles is assumed), and so conservation of angular momentum is expected to place no restrictions on the random decay of the nucleus, and the two isomeric states will be populated in constant ratio dependent on the statistical weights of their respective spins. No such limiting ratios were found in the recent work on isomer cross sections[10,11] in the region of compound nucleus formation.

In the investigation reported here the ratio of the cross sections for formation of several pairs of isomers produced by different nuclear reactions has been determined as a function of bombarding proton energy up to 21 Mev. Included in this work are also yield ratios of neighboring isotopes differing by two mass units. The results are interpreted in terms of spin changes and conservation of angular momentum.

EXPERIMENTAL

All targets were wrapped in a single ($\sim$5 mg/cm^2) sheet of aluminum foil and irradiated with the 41-inch U.C.L.A. synchrocyclotron. The targets were either salts or metal foils. Salts were always bombarded with the circulating beam of the cyclotron; the proton energy was adjusted by varying the radial distance of the target from the origin of the beam. Metal foils were irradiated in the deflected beam.

Chromium was bombarded as chromic oxide when a relative yield determination was sought and as a mixture of chromic and cupric oxide (in a definite proportion) when it was intended to determine absolute cross sections. The reaction $Cu^{63}(p,n)Zn^{63}$ was used to monitor the $Cr^{52}(p,n)Mn^{52m}$ reaction since foils of chromium were not available. The $Cu^{63}(p,n)Zn^{63}$ excitation function was determined, independently, by the method of stacked foils and was in agreement with the results of Ghoshal.[12]

Rubidium was bombarded as the chloride and silver as silver oxide and also as the metal.

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TABLE I. Cross sections for the reactions $Cr^{52}(p,n)Mn^{52m}$ and $Cr^{52}(p,n)Mn^{52}$.

Energy (Mev)	(Mn^{52m}) (millibarns)	(Mn^{52}) (millibarns)
6	2	...
7.5	128	21.2
10	358	116
10	312	104
12.5	410	203
16	265	197

For the bombardment of stacked foils a special shielded target holder was used. It consisted of an aluminum box ($2\frac{1}{4}$ inch$\times 1\frac{1}{3}$ inch$\times\frac{1}{2}$ inch) with a one-inch diameter opening in front, in which the foils were placed. The box was completely insulated with Lucite except for the opening above the foils and the whole assembly wrapped in a thin aluminum shielding foil which was grounded. The target holder was attached to a current-reading probehead, which was connected to a vibrating-reed electrometer current integrator.

After being irradiated, all targets other than the silver and copper foils were subjected to chemical separations. Carriers were added, and after suitable separations, the radioactive products strontium, silver, manganese, and zinc were recovered as strontium oxalate, silver chloride, manganese dioxide, and $ZnHg(SNC)_4$, respectively. The activities of the 38-minute Zn^{63} and the 24-minute Ag^{106} obtained in the bombardment of copper and silver foils were measured directly from the foils by using aluminum absorbers to decrease unwanted activities. After the 24-minute Ag^{106} had died out, however, the silver foils were also dissolved; the silver was separated from other elements and precipitated as AgCl before the activities of the 8-day isomer were determined.

All precipitates were mounted, dried, and weighed in a standardized manner in order to correct for chemical yield.

The activities were determined by means of a Geiger counter or a scintillation counter. The counting unit of the Geiger counter was a lead-shielded end-window G-M tube with a thin mica window used in conjunction with a scaler of conventional type. The sensitive unit of the scintillation counter was a one-inch by one-inch NaI(Tl) crystal mounted on a DuMont 6292 photomultiplier tube. The pulses were shaped, amplified, and then fed into a single-channel Atomic Instrument Company Model 510 pulse-height discriminator whose output was connected to a scaler. The scintillation counter was used to measure the manganese activities produced from chromium and to determine the absolute disintegration rate of the 8-day Ag^{106} activity.

RESULTS

In all cases relative yield curves were first obtained by determining the ratio of activities as a function of proton energy. For those reactions where absolute cross sections were determined, the relative yields were normalized to the ratio of cross sections at one particular energy.

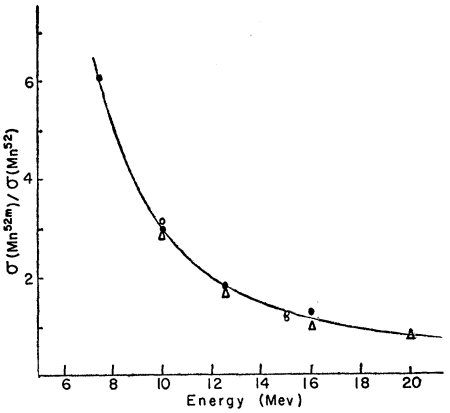

FIG. 1. Ratio of cross sections of Mn^{52} isomers. ○ determined from absolute cross sections; all other points from relative yields, normalized at 10 Mev.

1. Cr(p,n)Mn

For the determination of the $Cr^{52}(p,n)Mn^{52m}$ cross sections, chromium and copper oxides were mixed in the target and the ratio of Zn^{63} to Mn^{52m} was determined at several different energies. Zn^{63} was particularly suitable to monitor this reaction since both Zn^{63} and Mn^{52m} decay by the emission of positrons with about the same energy. The cross sections for Mn^{52} were then calculated from the ratio of activities after correction for self-absorption and assuming 35% positron emission for Mn^{52}. Table I gives the results of these measurements.

Figure 1 shows the ratio of these cross sections and also other values obtained from relative yield curves normalized to the cross-section ratio at 10 Mev.

All points were determined with the Geiger counter except the yield ratio at 20 Mev, which was measured with the scintillation spectrometer. At high energies the formation of the 44-minute Mn^{51} by the $(p,2n)$ reaction made it impossible to resolve the Geiger-counter decay curves. Since the 44-minute Mn^{51} is a pure positron emitter while the two Mn^{52} isomers both have high-energy gamma-rays, decay curves of the gamma-rays were easily resolved into the 21-minute and 5.8-day components. The activities obtained with the scintillation counter were calibrated in terms of Geiger counter readings by following the decay of a manganese sample produced at 15 Mev with both counters.

The (p,n) cross sections on chromium have been determined by others[7] in the energy range below 6.7 Mev. Their data indicate a value of about 40 millibarns for the $Cr^{52}(p,n)Mn^{52m}$ cross section at 6 Mev as compared to 2 millibarns obtained in the present work. However, just below 6 Mev the cross section decreases very rapidly and in view of the large energy spread of

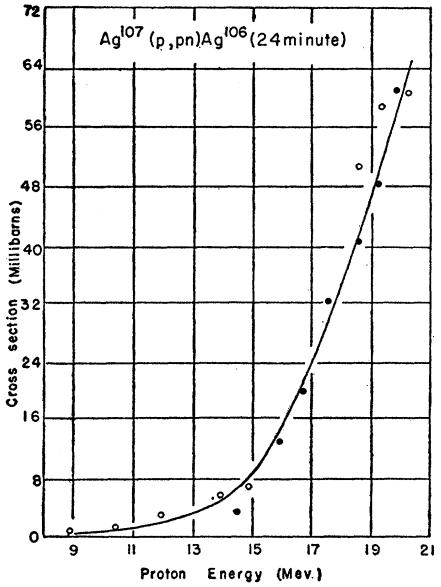

FIG. 2. Cross section for 24-minute Ag^{106}. ○ 14-mg/cm² foils; ● 56-mg/cm² foils.

the circulating beam with which our samples were irradiated, the discrepancy cannot be considered serious.

2. $Ag^{107}(p,pn)Ag^{106}$ (24-minute) and $Ag^{107}(p,pn)Ag^{106}$ (8-Day)

The excitation function for the reaction $Ag^{107}(p,pn)$-Ag^{106} (24-minute) is shown in Fig. 2. For the determination of the 24-minute activity the foils were not subjected to chemical separation. The activities were measured with the Geiger counter. The branching ratio was taken from the data of Bendel, Shore, Brown, and Becker[13] who reported that 56% of the 24-minute activity decays by 1.96-Mev positrons, 7% by positrons of 1.45 Mev, the rest by electron capture. In the present work, the cross sections were calculated on the basis of a 63% positron decay (all of 1.96 Mev) and 37% decay by electron capture; the somewhat smaller over-all counting efficiency of the 1.45-Mev positron was ignored. A probable error of 30% was estimated for the silver reaction.

The general shape of the excitation function agrees with the data of Cohen, Newman, Charpie, and Handley; but the cross sections these authors obtained are larger than ours by a factor of about two. A possible explanation for this discrepancy is the use of a different electron capture branching ratio.

Figure 3 shows a plot of the $Ag^{107}(p,pn)Ag^{106}$ (8-day) excitation function. The silver samples were chemically separated from cadmium and palladium.

All silver samples were counted with the Geiger counter, then one silver chloride sample was counted in two different ways: by the Geiger counter and by the scintillation counter. The sum of the emission rates of the two gamma-rays at 1.55 Mev and 0.511 Mev[10] was taken as the absolute disintegration rate of the 8-day Ag^{106}.

Large deviations were introduced in the determination of the absolute disintegration rate of the 8-day Ag^{106}, where, because of Compton scattering, the area under the 0.511-Mev peak could not be precisely determined. An error of 30–40% was estimated for this reaction.

Although the 24-minute Ag^{106} was obtained in good yield at much lower energies, the counts of the 8-day isomer were too low to permit an accurate determination of its activity below a proton energy of 16 Mev. The ratios of the activities of the two isomers and the approximate cross-section ratio are shown in Fig. 4. The point at 15 Mev represents a lower limit; the actual ratio could not be determined with certainty.

3. $Rb(p,n)Sr$

In Fig. 5 a plot is shown of the ratio of the activities of Sr^{85} to Sr^{87m}, Sr^{85m} to Sr^{87m}, and Sr^{85m} to Sr^{85} as a function of the bombarding proton energy. The 2.8-hour Sr^{87m} and the 65-day Sr^{85} were easily resolvable from the decay curves, but some difficulties were experienced in resolving the 70-minute Sr^{85m} since it was largely masked by the 2.8-hour Sr^{87m}. The error indicated in the graph is the maximum possible error in the yield ratios.

DISCUSSION

The yield ratio curves appear to follow the rule previously cited, at least as far as the low-energy portion of these curves is concerned.

In the case of the $Cr^{52}(p,n)Mn^{52m}$, Mn^{52} reaction, Cr^{52} has a spin of 0, Mn^{52m} a spin of 2, and Mn^{52} a spin of 6. At low excitation energies the small change in spin with

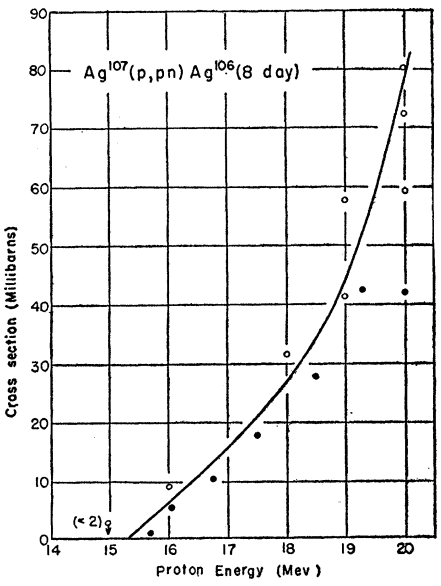

FIG. 3. Cross section for 8-day Ag^{106}. ● stacked foils; ○ indirectly from relative yields.

[13] Bendel, Shore, Brown, and Becker, Phys. Rev. **90**, 888 (1953).

formation of Mn^{52m} would favor formation of this isomer and the ratio of Mn^{52m} to Mn^{52} should decrease as the bombarding particle energy is increased. Figure 1 shows that this is the observed behavior.

The spins of the 24-minute and 8-day Ag^{106} isomers are 2 and 6, respectively. The low spin of the target nucleus, Ag^{107}, should then cause a decreasing ratio of the 24-minute to 8-day activities as is shown in Fig. 4.

In the case of $Rb^{85}(p,n)Sr$, the spins are Rb^{85}, 5/2; Sr^{85m}, 1/2; Sr^{85}, 9/2, with an excited state in the Sr^{85} with spin 7/2. A spin of 5/2 is intermediate between 1/2 and 7/2 (and 9/2), so that at low excitation energies both isomers can be reached, except of course, near the threshold where only the ground state is possible. At higher energies, the total angular momentum of the compound nucleus increases on the average and the formation of Sr^{85m} decreases relative to Sr^{85} but not to the extent observed in the other cases discussed above.

The ratio of the activities of the two Sr^{85} isomers to the activity of Sr^{87m} can also be related to the spins of the respective nuclides. (This, of course, does not apply to the region near the threshold.) The spins of Sr^{87m} and Sr^{87} are 1/2 and 9/2; Sr^{87}, however, is not radioactive. Since the spins of the target nuclides Rb^{85} and Rb^{87} differ but little (Rb^{87} has a spin of 3/2 as compared to the spin of 5/2 of Rb^{85}), we can expect the ratio of the activities of Sr^{85m} to Sr^{87m} to remain constant and the ratio of the activities of Sr^{85} to Sr^{87m} to increase with increasing energy. Figure 5 shows this to be so.

Thus, in every case tested, it is seen that low-spin target nuclei produce nuclides with low spin at low excitation energy and that with increasing energy, product nuclides with high spin become increasingly probable. There is, however, no indication of a high-energy limiting ratio as one would expect on the basis of the statistical theory. According to the simple statistical theory of nuclear reactions,[14] particles are

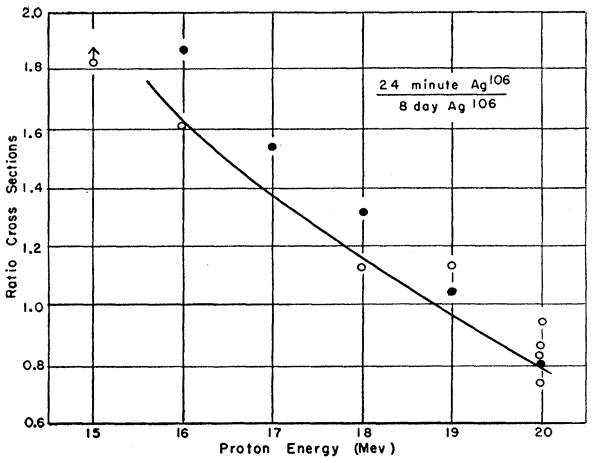

FIG. 4. Ratio of cross sections for Ag^{106} isomers. ● from smooth curves of Fig. 2 and Fig. 3; ○ from experimental points.

[14] J. M. Blatt and V. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952).

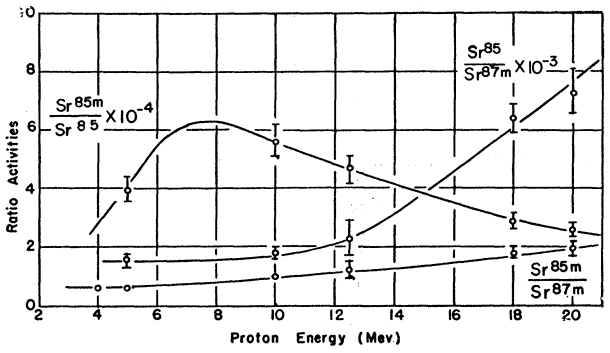

FIG. 5. Relative yields of strontium activities from the bombardment of rubidium with protons.

emitted from the compound nucleus in a Maxwellian distribution. Further de-excitation takes place through a gamma cascade with no correlation between the emitted particles and gamma rays. This investigation shows that some correlation exists, even at relatively high energies. This is not altogether surprising. The number of compound nuclei having high spin states increases very rapidly with increasing energy. De-excitation by gamma emission does not transfer a great deal of angular momentum since, on the average, only 2.5 gammas are emitted in a cascade below the threshold for particle emission,[15] and transitions involving gamma-rays of high multipole order are rare. Apparently, many of the high-spin states must de-excite by emitting particles of high angular momentum. But if this is so, there will be a shift of the distribution of emitted particles toward higher energy.

This shift toward higher energies is easily seen by the following qualitative argument: Consider a particular state of the compound nucleus which decays by particle emission to the various states of the residual nucleus by emission of particle b. The emitted particles will then have an energy distribution, ϵ_b, up to a certain maximum, $\epsilon_{b\,\max}$. Because of the centrifugal barrier some of the possible states within the interval $d\epsilon_b$ are not accessible but only a fraction, $f(\epsilon_b)$, which is an increasing function of ϵ_b. The distribution of emitted particles is then given by

$$I_b(\epsilon_b)d\epsilon_b = \text{const}\,\epsilon_b\sigma_c(\epsilon_b)f(\epsilon_b)W(E)d\epsilon_b,$$

where $W(E)$ is the true level density irrespective of spin and $f(\epsilon_b)W(E)$ is an "effective level density."

Since a shift in the distribution toward higher energies will increase the probability of charged particle emission relative to neutron emission, reactions in which charged particles are emitted become more probable than when the conventional Maxwell distribution is assumed. This may, in addition to "direct interaction," partly account for the large cross sections which are found in reactions where charged particles are emitted.

[15] C. O. Muehlhause, Phys. Rev. **79**, 277 (1950).