Cross Sections for Formation of the Isomeric Pair In^{117} and In^{117m} in the $Sn^{120}(p,\alpha)$ Reaction

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The excitation functions and yield ratio (σ_g/σ_m) for the isomeric pair In^{117} and In^{117m} produced from Sn^{120} by the (p,α) reaction were measured for energies from threshold to 22 MeV. The yield ratio vs energy is large at low energies, drops to a minimum at 13 MeV, and then rises linearly to 22 MeV. The measured half-lives of the excited and ground state of In^{117} were found to be 117.5±1.0 and 44.6±0.6 min, respectively.

INTRODUCTION

THE ratio of the yields of isomeric pairs as a function of energy is a further tool for the investigation of the mechanism of nuclear reactions. In particular, studies of this kind stress the importance of the conservation of angular momentum and of the dependence of the cross section on nuclear spin. When the reaction can be considered to proceed through the compound-nucleus mechanism, the isomer yield ratio will depend on the spin distributions brought about by the following processes: (1) formation of the compound nucleus, (2) formation of the residual nucleus in its excited states through particle emission, and (3) gamma-ray de-excitation of the energy levels of the residual nucleus.

The spin distribution in the compound nucleus is dependent on the energy of the bombarding particle via the energy dependence of the transmission coefficients for various l values; that of the excited residual nucleus will depend on the transmission coefficients for the various l values of the outgoing particles and on the distribution in J of the energy levels in the residual nucleus; and that brought about by the gamma-ray cascade will depend on the distribution in J of the energy levels. The details of these processes have been discussed by Huizenga and Vandenbosch.[1]

Aside from the energy considerations that hold for particles emitted in the near vicinity of threshold, it is expected that at low bombarding energy the state with spin closest to that of the target nucleus will be favored, while, for compound nucleus reactions, the population of the previously unfavored state will increase with increasing bombarding energy.

The present investigation is a study of the reaction $Sn^{120}(p,\alpha)In^{117,117m}$. This reaction should be of interest not only from the standpoint of isomer ratio determination but also for its intrinsic value as a (p,α) reaction. Fulmer and Cohen[2] and Fulmer and Goodman[3] have made an extensive study of (p,α) reactions by observing the outgoing alpha particles, but few measurements have been made on (p,α) activation reactions. In contrast to previously published works on isomer ratio cross sections in proton-induced reactions,[4,5] the present study represents a reaction which makes up only a small fraction of the total compound nuclear reaction cross section. For such reactions, the initial spin distribution is likely to be considerably fractionated as a result of competing reactions. In this paper we present only the experimental details and results of this investigation; a theoretical analysis will be given in a subsequent paper.[6]

In non-compound-nucleus reactions the shapes of the isomer ratio vs energy curves are expected to differ considerably from the ones obtained through the formation of a compound nucleus. Thus, Zherebtsova *et al.*[7] showed that there are marked differences in isomer ratio curves for the reactions $Zn(d,p)Zn^{69,69m}$ and $Ga^{69}(n,p)Zn^{69,69m}$. The former is an example of a stripping reaction, and, as expected, the state requiring small spin changes is favored at all times; this yields an isomer-ratio curve which does not change greatly with energy.

During the course of the present work, it became evident that the data could not be reconciled with the decay scheme reported in the *Nuclear Data Sheets*[8] (in particular with the reported half-life of 66 min for In^{117}). The latter is based essentially on the work of McGinnis.[9] More recent data on the decay of In^{117} have been obtained by Gleit[10] and by Wolfe and Hummel[11] who

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have reported, respectively, the values 57 and 43 min for the half-life of In^{117}. In view of these circumstances, the half-lives of In^{177} and In^{117m} were redetermined.

EXPERIMENTAL

Metal foils of tin enriched to 97% in Sn^{120} were bombarded in the external beam of the ORNL 86-in. cyclotron. The foils were separated with aluminum absorbers to give the desired energy. The energy at the midpoint of each foil was determined with the aid of stopping-power curves interpolated from the data of Aron, Hoffman, and Williams[12] and from the range-energy curve for protons in aluminum of Bichsel, Mosley, and Aron.[13] The maximum energy of the cyclotron beam was found (by absorption methods) to be 22.4 MeV, with a beam spread of ± 0.1 MeV and a day-to-day variation of ± 0.3 MeV. During bombardment, the stack of tin foils was located at the rear of a Faraday cup, 7.5 in. long and 3.5 in. in diameter; no bias was applied to the cup. Previous measurements, in which the solid angle available for electron escape was varied and a magnetic field was applied, showed that there was no loss of secondary electrons from the cup under the conditions of the bombardment.

The intensity of the beam was determined with the aid of a current integrator which produced an output pulse whenever a small capacitor was charged to about one volt. The integrator was absolutely calibrated once with precision resistors and a standard cell and thereafter calibrated relatively by using an internally generated current. The stability of the current integrator over a period of two months was $\pm 1\%$. The effect on the cross sections of variations in the beam current during bombardment was less than 1%.

After irradiation, the tin foils were dissolved in concentrated hydrochloric acid, antimony and indium carriers were added, and the antimony and tin were precipitated as sulfides. Indium was then removed as the hydroxide, redissolved, and additional tin and antimony carriers were added. The *p*H was adjusted to 3.5, and indium was again precipitated as sulfide, leaving tin and antimony in solution. This procedure was repeated twice, and indium was finally precipitated with 8-hydroxyquinoline. The precipitate was collected on a preweighed filter paper by a filter chimney arrangement, dried, weighed, and mounted in a standardized manner.

The activities of the indium samples were determined by means of lead-shielded end-window Geiger counters and also with a gamma-ray scintillation spectrometer and single-channel pulse-height analyzer. The sensitive unit of the scintillation counter was a 3 in.$\times$3 in. NaI(Tl) crystal mounted on a DuMont 6393 photomultiplier tube. A wide channel (22%), centered about

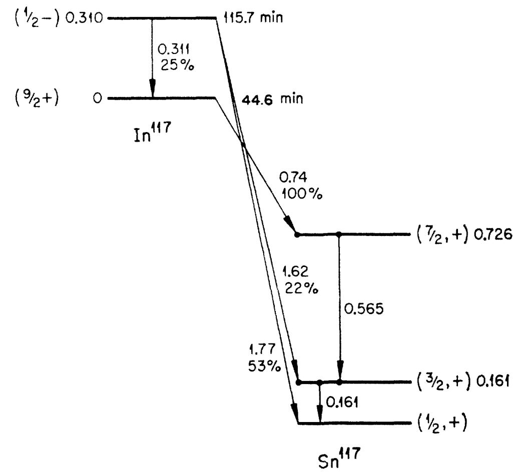

FIG. 1. In^{117} decay scheme as modified by the present work.

the 565-keV gamma ray emanating from the decay of the ground state of In^{117} was counted. The Geiger counters were intercompared by counting a standard Na^{22} source in each, and one was calibrated absolutely with a NBS-RaDEF source. The usual corrections[14] for backscatter, window and air absorption, and source scattering and absorption were applied.

These data were consistent with the published half-life for In^{117m} but were inconsistent with that for In^{117}. For the determination of the latter, 30 mg of CdO, enriched to 93.8% in Cd^{116}, was irradiated for 16 h in the Oak Ridge Reactor. The sample was allowed to decay for 19 h and then was dissolved in 6*N* HCl. Indium carrier was added, and the indium activity that had grown in since irradiation was separated from the cadmium by repeated sulfide precipitations of cadmium, with indium carrier added at each step. The purified cadmium was then allowed to decay for another hour, and indium carrier was again added. The indium activity that had grown in during the hour was separated from the cadmium sample by sulfide precipitation, purified by repeated sulfide precipitation of added cadmium carrier, and then removed as the hydroxide. The hydroxide was dissolved in dilute HCl, and a few drops of the solution was placed on a filter disk for gamma counting by means of a 400-channel RIDL pulse-height analyzer. Correction for dead time was made by the analyzer. Counting extended over a period of 10 h.

RESULTS

The counting data, uncorrected for background, were subjected to a least-squares analysis on an IBM 704. The iteration program used yields both half-lives and magnitudes. Any arbitrary number of half-lives can be fixed, all subject to the limitation that there be at least two variables left. The print-out includes the best estimates of the standard deviation of the variables.

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[13] H. Bichsel, R. E. Mosley, and W. A. Aron, Phys. Rev. **105**, 1788 (1957).

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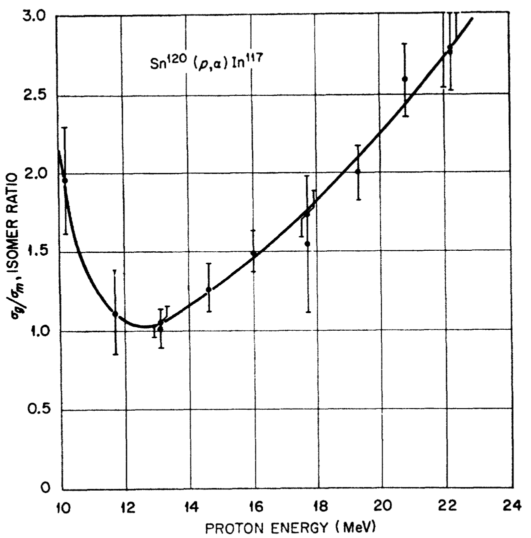

FIG. 2. Ratio of σ_g/σ_m as a function of proton bombarding energy.

Decay of In^{117}

From three samples that had been beta counted for periods of 12 to 15 h and for which the counting had started after the ground-state contribution had died away, the value for the half-life of the isomeric state of In^{117} was determined to be 117.5±1.0 min. This value is in good agreement with a published[8] value of 114±3 min.

With this half-life for the excited state fixed, the decay of the 565- and 161-keV lines of the reactor sample were analyzed for the half-life of the ground state. The value obtained was 44.6±0.6 min, which compares favorably with Wolfe's[11] value of 43 min. Further support for this new value comes from the fact that it is consistent with the values obtained when the single-channel decay data of the 565-keV gamma ray from the cyclotron-produced samples were analyzed for the ground-state half-life.

Also present in the reactor sample was In^{115m} produced by the decay of Cd^{115}. The presence of a strong 330-keV line from this activity obscured the 311-keV line from In^{117m} so that a determination of the correction to the 161-keV photopeak due to the Compton continuum of the 311-keV gamma ray could not be made. An upper limit of 0.35 was obtained for the value of the fraction of decays of the isomeric level that populate the ground state. This is to be compared with 0.23±0.03 found by McGinnis[15] and 0.28 reported by Wolfe.[11]

Figure 1 shows the decay scheme adopted for the calculations in this paper.

Isomer Ratio

The ratio of the production cross sections for the ground state and for the isomeric state, σ_g/σ_m, was obtained from the single-channel gamma-counting data of the 565-keV gamma-ray decay of In^{117} produced in the cyclotron bombardments. This gamma ray is produced only by the decay of the In^{117} ground state; its counting rate will, therefore, have the same time dependence as the ground state.

The population of the ground state at the end of a bombardment of duration T is given by

$$N_g^0=\frac{K_g+AK_m}{\lambda_g}(1-e^{-\lambda_g T})+\frac{AK_m}{\lambda_g-\lambda_m}(e^{-\lambda_g T}-e^{-\lambda_m T})\,; \quad (1)$$

the population of the ground state at a time t after cyclotron shutdown is given by

$$N_g=\left(N_g^0-\frac{N_m^0 A\lambda_m}{\lambda_g-\lambda_m}\right)e^{-\lambda_g t}+\frac{N_m^0 A\lambda_m}{\lambda_g-\lambda_m}e^{-\lambda_m t}. \quad (2)$$

Here, the subscripts m and g refer to the isomeric and ground states, respectively; the K's are production rates (proportional to σ); the λ's are the decay constants; and A is the branching ratio for decay of the isomeric to the ground state. Equation (2) can be rewritten as

$$N_g=C_g\exp(-\lambda_g t)+C_m\exp(-\lambda_m t). \quad (3)$$

The least-squares fit to the decay data at each energy yielded the C's (except for a scaling factor). Where the ratio of coefficients $C_g/C_i\equiv R$, the isomer ratio is

$$\frac{\sigma_g}{\sigma_m}=\frac{A}{\lambda_g-\lambda_m}\left(R\lambda_g\frac{1-\exp(-\lambda_m T)}{1-\exp(-\lambda_g T)}+\lambda_m\right). \quad (4)$$

Table I gives the isomer ratio as obtained in this experiment; the same data are shown plotted in Fig. 2. As is seen, the isomer ratio is large at low energies, drops to a value of ~1.0 to 12 MeV, and rises linearly to a value of 2.75 at 22 MeV. The errors shown represent the best estimate of the probable error obtained from propagation of the fitting errors in λ_g, λ_m, and R, and an assumed 10% error in A.

TABLE I. Ratio of σ_g to σ_m as a function of proton bombarding energy.[a]

Energy (MeV)	σ_g/σ_m
22.2	2.75±0.23
22.2	2.75±0.22
20.8	2.59±0.23
19.3	2.01±0.16
17.7	1.74±0.14
17.7	1.55±0.42
16.0	1.50±0.13
14.6	1.27±0.10
13.1	1.02±0.09
13.1	1.06±0.10
11.7	1.12±0.17
10.2	1.96±0.24

[a] The error indicated is probable error in the ratio determined by propagation of the fitting errors in R, λ_g, and λ_m, and an assumed 10% error in A.

[15] C. L. McGinnis (private communication).

Cross Sections

The cross section for the formation of the isomeric state was calculated from the Geiger counter data. The experimental data represent the decay of both the ground and isomeric states. The ground state decays with emission of a 0.75-MeV beta particle and the isomeric state with emission of both 1.77- and 1.62-MeV beta particles. The backscatter effect is the same for all beta particles, and the inscattering from the walls is expected to be the same. It is assumed that the air and window absorption and the source self-absorption and self-scattering effects together are the same for all beta particles. Then the decay of the total beta activity can be written as

$$\lambda_g N_g{}^0 e^{-\lambda_g t} 1-\left(\frac{A\lambda_m N_m{}^0}{(\lambda_g-\lambda_m)N_g{}^0}\right) +\lambda_m N_m{}^0 e^{-\lambda_m t}\left(1+\frac{A\lambda_m}{\lambda_g-\lambda_m}\right). \quad (5)$$

The values of $\lambda_m N_m{}^0[1+A(\lambda_m/\lambda_g-\lambda_m)]$ were obtained from least-squares fits to the data. The cross sections were then computed in the standard way; the results are shown in Fig. 3. The absolute errors in the cross section depend on the errors in A, beam integration, chemical efficiency, and counter calibrations; they are estimated to be $\pm 25\%$. The relative errors can be judged from the scatter of the duplicate points. The error bars shown represent only the errors involved in the fitting of the decay curves.

The ground-state cross sections were calculated from the curves of σ_m and σ_m/σ_g and were added to the curve for σ_m to get the total (p,α) cross section. This is also plotted in Fig. 3.

The behavior of the isomer ratio vs energy curve may be explained qualitatively by considering the spins of target and final nuclei and taking into account the law of conservation of angular momentum. The large value at low energies is undoubtedly due to the difference in threshold energies for formation of ground and excited states. Near the threshold, the ground state will always be favored, regardless of spin. The dip in the vicinity of 12 MeV shows preference for the state with spin near that of the target nucleus, which is the excited state. As the energy is increased, the compound nucleus is formed in states of higher spin, resulting in an increase in population of the high-spin state, which is the ground state. An analysis of these results in the manner of Huizenga and Vandenbosch[1] will be given in following paper.

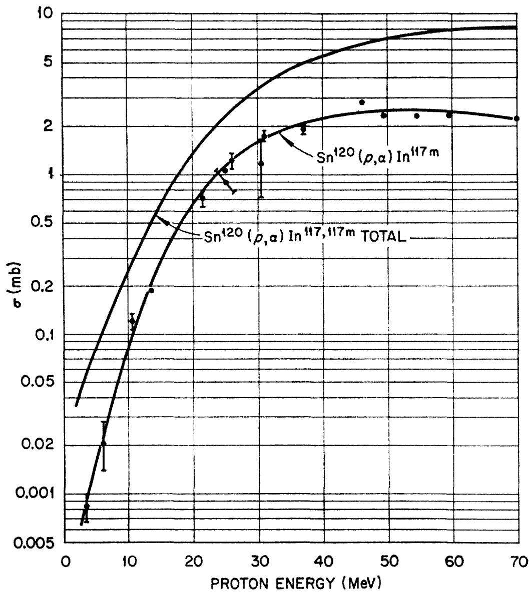

FIG. 3. σ_m, for the reaction $Sn^{120}(p,\alpha)In^{117m}$ and total $Sn^{120}(p,\alpha)In^{117,117m}$ cross section as a function of proton bombarding energy.

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