Measurement of 14-MeV neutron cross sections for ^{88}Zr and ^{88}Y

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We have measured (n,2n) and (n,np+n,pn) cross sections for the radioactive nuclides ^{88}Y and ^{88}Zr. The results are, for the (n,2n) reaction on ^{88}Y at 14.19±0.04 MeV, 1140±50 mb; and at 14.8±0.1 MeV, 1180±50 mb. The ^{87}Y isomer ratios (m/total) are 0.70±0.05 and 0.74±0.05, respectively. For ^{88}Zr at 14.8±0.1 MeV, the (n,2n) cross section is 467±23 mb, and the (n,np+n,pn) cross section is 253±25 mb. The ^{87}Y isomer ratio for the latter reaction is 0.90±0.06. The results are compared to calculations which are based on a combined statistical-preequilibrium model. Half-lives for ^{88}Zr, ^{87}Zr, ^{87}Y^m, and ^{87}Y^g are presented. Branching ratios for the decay of ^{87}Zr to ^{87}Y^{m,g} were determined and the photon intensity of the 484.9-keV gamma ray from the decay of ^{87}Y^g was measured.

NUCLEAR REACTIONS ^{88}Y(n,2n)^{87}Y^{m,g}, E=14.2, 14.8 MeV; ^{88}Zr(n,2n)^{87}Zr, E=14.8 MeV; ^{88}Zr(n,2n + n,np)^{87}Y^{m,g}, E=14.8 MeV; measured $\sigma(E)$. Comparison with combined statistical and preequilibrium model.
RADIOACTIVITY ^{87}Y^m, ^{87}Y^g, ^{87}Zr, ^{88}Zr; measured half-life. ^{87}Zr, measured branching decay. ^{87}Y^g, measured I_γ.

I. INTRODUCTION

Experimental data are needed for neutron-induced reactions in the energy range around 14 to 15 MeV in order to verify the accuracy of nuclear models used in the calculation of cross sections. We rely upon nuclear model calculations to provide information on neutron-induced reactions in energy regions where measurements are incomplete or lacking. The calculation codes receive a more meaningful test when their results can be compared with measurements for a number of target isotopes for a given element. A relative wealth of experimental data exists for stable target nuclides, but is almost completely lacking for unstable targets. We have measured cross sections for reactions on 106.6-d ^{88}Y and 82.6-d ^{88}Zr and compare the results with those from recent calculations using the statistical model of nuclear reactions. These are the first measurements reported for cross sections of this type using relatively short-lived nuclides as target materials. Even though other neutron energies are of interest, we emphasize the range from 14 to 15 MeV as it is only here that the neutron fluxes available are sufficiently high for this type of measurement to be made.

The yields of reactions on short-lived targets such as these are difficult to measure, not only because of obvious handling problems, but also because the intense target radioactivity interferes with the measurement of the radioactive product nuclides. It is generally necessary to physically separate the product from the target by means of an isotope separator, for example, or to chemically separate the daughter nuclide of the product. The general technique used for these experiments was to make several separations of ^{87}Sr^m as a function of time from the ^{88}Y target and from the yttrium and zirconium fractions of the ^{88}Zr target.

We report here the results of measurements for the (n,2n) reaction on ^{88}Y at 14.2 and 14.8 MeV, and the (n,2n) and (n,np) reactions on ^{88}Zr at 14.8 MeV. The (n,np) reaction refers to the sum of the reactions (n,np), (n,pn), and (n,d).

II. EXPERIMENTAL

The target material was produced by long bombardments of molybdenum targets with ~750-MeV protons at the Clinton P. Anderson Meson Physics Facility (LAMPF) of the Los Alamos National Laboratory. The ^{88}Zr was produced by spallation reactions as a by-product from the production of ^{82}Sr for the Medical Isotopes Program. The ^{88}Y target material was obtained as the decay product of ^{88}Zr. Both the ^{88}Y and ^{88}Zr were purified radiochemically from other elements before use. In addition, the material was not used until many months after the end of bombardment so that short-lived ^{87}Y produced in the LAMPF bombardment could decay to a negligible level.

In both the ^{88}Y and the ^{88}Zr experiments the target material was purified from its daughter nuclides, aliquotted for assay, and then coprecipitated with $Tm(OH)_3$. These steps were done within 24 h of the start of the irradiation. The $Tm(OH)_3$ precipitate was dried and the powder packaged in a small quartz tube for the irradiation. The ^{169}Tm(n,2n)^{168}Tm reaction served as an internal monitor for the neutron fluence. The cross section for this reaction is essentially constant at 1.98 b in the 14–15 MeV range, and the ^{88}Y and ^{88}Zr cross sections were measured relative to this value.[1]

The neutron irradiations were made at the Lawrence Livermore National Laboratory RTNS-1 facility (ICT).

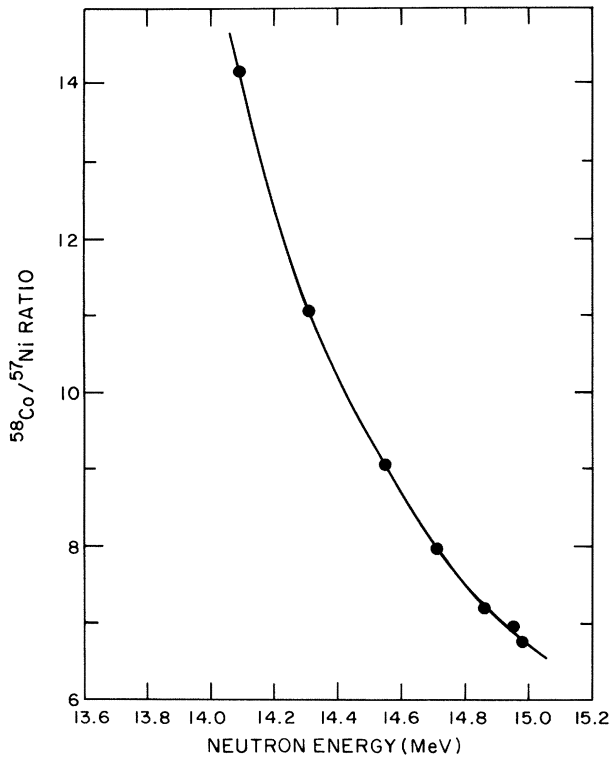

FIG. 1. Plot of the $^{58}Co/^{57}Ni$ activation ratio from nickel foils irradiated with neutrons at various angles to the incident deuteron beam.

This facility provides an intense source of 14-MeV neutrons by the bombardment of a rapidly rotating titanium-tritide target with 400-keV deuterons. The 14-MeV neutron source strength in these experiments was about 2×10^{12} n/sec. The neutron flux density was monitored with a proton-recoil counter so that corrections could be made for the small changes in neutron intensity. The total neutron fluences for these irradiations were on the order of 10^{15} n/cm^2.

A number of irradiations were performed on ^{88}Zr targets ranging from 42 to 141 mCi (2.3 to 7.8 μg). In all ^{88}Zr irradiations, the target material was placed as close to the neutron source as possible at 0° to the deuteron beam for an effective neutron energy of 14.8 MeV. At this position approximately 63% of the neutrons have energies between 14.6 and 15.0 MeV.

Irradiations were also performed on ^{88}Y targets ranging from 10 to 40 mCi (0.75 to 2.9 μg). The first several irradiations were carried out exactly like the ^{88}Zr experiments: at 0° to the deuteron beam and as close to the neutron source as possible. The effective neutron energy was again 14.8 MeV. In the final ^{88}Y experiment we used a special conical-head tritium target designed for sample irradiations at angles up to 120°. A 40 mCi ^{88}Y target was irradiated at an angle of about 83° to the deuteron beam and at a distance of 3 cm from the neutron source to obtain better neutron energy resolution. A small nickel foil was placed at the tip of the quartz tube containing the ^{88}Y target. The $^{58}Co/^{57}Ni$ ratio was measured and used to determine the effective neutron energy incident on the nickel foil and, hence, on the ^{88}Y target. The $^{58}Co/^{57}Ni$ ratio is sensitive to the neutron energy[2] and was calibrated in a separate experiment in which several nickel foils were irradiated at various known angles on the arc of a 20-cm radius circle. A plot of the $^{58}Co/^{57}Ni$ ratio versus neutron energy is shown in Fig. 1. Using this plot, we determined that the average neutron energy in the final ^{88}Y irradiation was 14.19 ± 0.04 MeV, with 67% of the neutrons having energies between 14.1 and 14.3 MeV.

Following the irradiations, the ^{88}Zr targets were dissolved in concentrated HCl, and chemical separations of zirconium, yttrium, thulium, and strontium were made. The Y/Tm/Sr fraction was separated from the ^{88}Zr target material by adsorbing the zirconium onto AG MP-1 anion exchange resin (BioRad Laboratories, Richmond CA) in concentrated HCl. The time of chemical separation, which represents the start of the $^{87}Sr^m$ growth, was noted. Standardized strontium carrier was added to the zirconium fraction and aliquots were taken for ^{88}Zr assay. The zirconium fraction was then split into two or four samples and set aside to allow $^{87}Sr^m$ to grow in from the (n,2n) product, ^{87}Zr. Meanwhile, strontium was removed from the Y/Tm/Sr fraction by hydroxide precipitations of yttrium and thulium. Once again the time of chemical separation was noted. The sample was weighed to determine the amount of Tm_2O_3 present, redissolved, and standardized strontium carrier was added. The Y/Tm sample was then split into two or four fractions, and set aside to allow $^{87}Sr^m$ to grow in from the (n,np) products, $^{87}Y^m$ and $^{87}Y^g$.

At several later times, strontium was separated from the zirconium and Y/Tm samples to measure the amount of $^{87}Sr^m$ that had grown into the samples. The time of each separation was noted since it represented the end of the growth period for $^{87}Sr^m$. Early separations of $^{87}Sr^m$ were necessary to obtain the yield for $^{87}Y^m$, while later separations gave results for the ground state of ^{87}Y. The strontium samples were purified radiochemically by a series of yttrium and zirconium precipitations with NH_4OH and by precipitation as $Sr(NO_3)_2$ and $SrCO_3$. The $^{87}Sr^m$ in the final $SrCO_3$ samples was measured by following the radioactive decay with a NaI(Tl) well-type detector.

After all the strontium separations had been completed, the thulium flux monitor was separated from the yttrium fraction using a high performance liquid chromatography (HPLC) cation exchange column and α-hydroxyisobutyric acid. It was then analyzed for ^{168}Tm.

Similar chemical separations, except for zirconium, were made for the ^{88}Y irradiations. In this case, two techniques were used to obtain the final strontium samples. In the first, strontium carrier was added to the Y/Tm fraction, and aliquots taken for ^{88}Y assay. The Y/Tm fraction was then split into several samples for later strontium separations. In the second method, several consecutive timed strontium separations were made from the entire Y/Tm fraction. Assays were made for ^{88}Y at each step to measure the yttrium loss during each strontium separation.

III. CALCULATIONS

The cross section results were calculated with the aid of a computer program which follows the nuclear reactions

TABLE I. Nuclear decay data used in the cross section calculations.

Nuclide	Half-life	Decay mode	E_γ (keV)	I_γ
^{88}Zr	82.6 d[a]	100% to ^{88}Y	394	0.97[b]
^{88}Y	106.6 d[c]	100% to ^{88}Sr	898	0.937[c]
			1836	0.992[c]
^{87}Zr	1.68 h[d]	98% to $^{87}Y^m$		
		2% to $^{87}Y^g$		
$^{87}Y^m$	13.37 h[d]	98.5% to $^{87}Y^g$	381	0.781[e]
		1.5% to $^{87}Sr^g$		
$^{87}Y^g$	3.317 d[d]	100% to $^{87}Sr^m$	485	0.938[d]
$^{87}Sr^m$	2.81 h[e]	100% to $^{87}Sr^g$	388	0.823[e]
^{168}Tm	93.1 d[b]	100% to ^{168}Er	198	0.522[f]
			816	0.488[f]

[a]Reference 3.
[b]Reference 4.
[c]Reference 5.
[d]This work.
[e]Reference 6.
[f]Reference 7.

occurring during the irradiation and also follows the growth and decay of each of the relevant nuclides. The input includes a list of up to eight nuclides, their half-lives, their decay products, and the branching fraction to each product. A list of nuclear reactions with target and product nuclides and the cross sections is also included. The program operates in small time steps which are chosen depending on the total time to be followed. For example, time steps of 0.02 min each are typically used. During each short time interval the nuclear reactions are calculated using the cross sections provided. The product

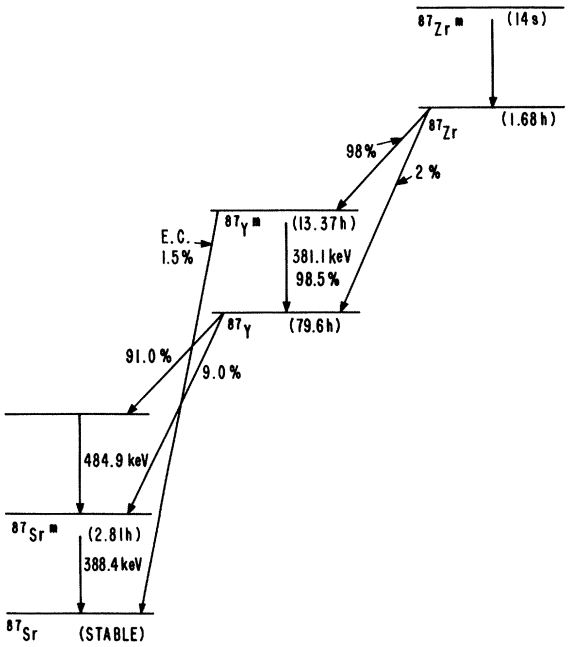

FIG. 2. Sketch of the decay scheme of the mass-87 zirconium, yttrium, and strontium nuclides.

TABLE II. ^{88}Y irradiation results.

Irradiation	mCi of target	E_n (MeV)	Measured σ(n,sn) mb	^{87}Y isomer ratio (m/total)
1	40	14.19	1140±50	0.70±0.05
2	10	14.8	1200±100	0.69±0.14
3	12	14.8	1180±50	0.75±0.05
Averages at 14.8 MeV:			1180±50	0.74±0.05

is increased and the target decreased by the appropriate amount. The decay of each nuclide in the time interval is also accounted for using the half-lives and branching fractions provided. By using sufficiently small time steps, the error in the method can be made negligibly small, usually less than 0.01%. Provision is made for chemical separations in the calculation. That is, the nuclides of any element can be set to zero at given times to correspond to actual Zr/Y/Sr separations.

The calculation starts with the beginning of the neutron irradiation and can be continued an arbitrary length of time past the end of the irradiation. The irradiation history is divided into any number of separate parts, each with a relative neutron flux given by the proton-recoil counter record. The total neutron fluence, as measured by the ^{169}Tm monitor reaction, is apportioned into each irradiation step. The calculation is normally run up to the time of a strontium separation so that the measured $^{87}Sr^m$ in a sample at separation time could be compared directly with the calculated amount. The measured $^{87}Sr^m$ is corrected for chemical yield losses in the separation and purification procedures.

The general mode of operation of the computer program is to assume values of the cross sections to be determined, run the calculation, and compare the calculated yield of $^{87}Sr^m$ at the separation time with the measured yield. Adjustments are then made to the input cross sections and the calculation repeated until the $^{87}Sr^m$ result matches the measured yield. The nuclear decay data used in the calculations are given in Table I. A sketch of the decay scheme of mass-87 zirconium, yttrium, and strontium is shown in Fig. 2.

IV. RESULTS AND DISCUSSION

The results for the ^{88}Y and ^{88}Zr irradiations are given in Tables II and III. Twelve experiments were performed in all. However, significant improvements were made in the chemistry and the overall experimental procedure each

TABLE III. ^{88}Zr irradiation results at $E_n = 14.8$ MeV.

Irradiation	mCi of target	Measured σ(n,2n) mb	Measured σ(n,np) mb	^{87}Y isomer ratio (m/total)
1	42	488±49		
2	36	474±47		
3	141	460±23	253±25	0.90±0.06
Averages:		467±23	253±25	0.90±0.06

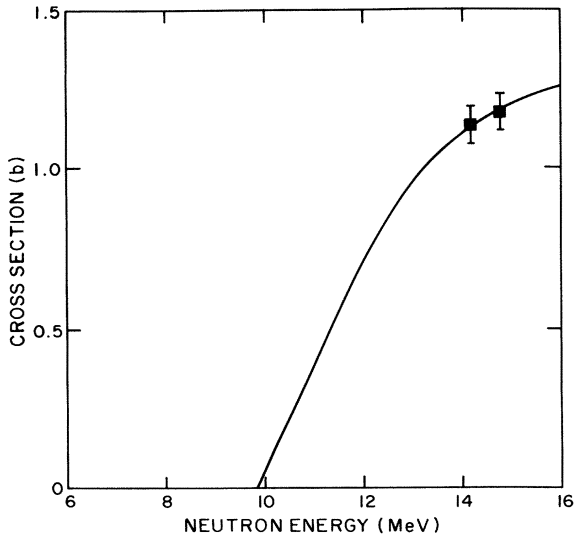

FIG. 3. Excitation function for the $^{88}Y(n,2n)^{87}Y^{m+g}$ reaction. The measured cross sections are shown along with the calculated excitation function of Arthur (Ref. 8).

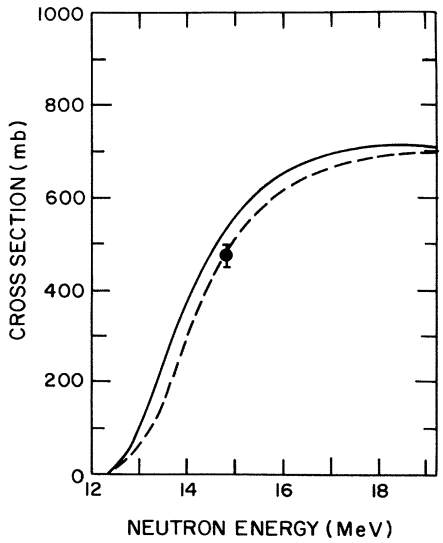

FIG. 4. Excitation function for the $^{88}Zr(n,2n)^{87}Zr$ reaction. The measured cross section is shown along with the calculated excitation functions of Arthur (---) (Ref. 9) and Gardner (——) (Ref. 10).

time an irradiation was performed. For example, the value obtained for the (n,np) cross section in the ^{88}Zr experiment is extremely sensitive to the time of each $^{87}Sr^m$ separation and to the completeness of that separation. We measured the (n,np) cross section in a number of earlier experiments; however, acceptable fits were not obtained with our computer code owing to inaccuracies in timing and insufficient chemical separations. In Table III the values listed for the (n,np) cross section and the isomer ratio are those obtained from the most accurate experiment. That experiment also gave the best computer fit to the data: measured/calculated ratio was unity within experimental error for the data from all of the strontium samples.

The ^{88}Y and ^{88}Zr cross sections were measured relative to the $^{169}Tm(n,2n)$ cross section, which has a value of 1980±40 mb in the 14–15 MeV range.[1] For the $^{88}Y(n,2n)$ reaction at 14.19±0.04 MeV, the measured cross section is 1140±50 mb, with an ^{87}Y isomer ratio (m/total) of 0.70±0.05. At 14.8±0.1 MeV the cross section is 1180±50 mb, and the isomer ratio is 0.74±0.05. The uncertainties represent one standard deviation and include known sources of error such as decay constants (<2%), separation times (0.6%), sample weights and carrier standardization (0.5%), the ^{169}Tm monitor cross section (2%), and the relative uncertainty in measuring $^{87}Sr^m$, ^{88}Y, and ^{168}Tm (2%). Some of the latter errors partially cancel since we actually measure ratios of nuclides. The ^{88}Y cross section is given by

$$\sigma_{88}=\frac{N_{87/88}}{N_{168/169}}(\sigma_{169}),$$

where $N_{87/88}$ is the measured ratio of $^{87}Y/^{88}Y$, $N_{168/169}$ is the measured yield of the internal monitor reaction, and σ_{169} is the cross section for the monitor reaction. In some cases we have increased the errors assigned to the weighted averages in Tables II and III to better reflect the total measurement uncertainty.

The measured ^{88}Zr cross sections at 14.8±0.1 MeV are 467±23 mb for the (n,2n) reaction and 253±25 mb for the (n,np) reaction. The ^{87}Y isomer ratio in the (n,np) reaction is 0.90±0.06.

The measured cross sections are plotted in Figs. 3–5. Shown for comparison are the calculated excitation functions of Arthur[8,9] and Gardner.[10] These calculations employ Hauser-Feshbach statistical model techniques with corrections applied for nonstatistical effects through use of the exciton preequilibrium model. The calculations are sensitive to such factors as the discrete level parameters

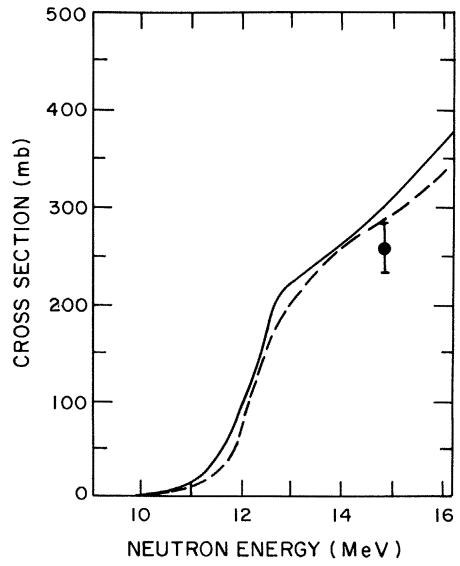

FIG. 5. Excitation function for the $^{88}Zr(n,np+n,pn)^{87}Y^{m+g}$ reaction. The measured cross section is shown along with the calculated excitation functions of Arthur (---) (Ref. 9) and Gardner (——) (Ref. 10).

and the level densities used. Our measured cross sections are in good agreement with these calculated excitation functions.

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APPENDIX

In order to accurately measure the cross sections in this work, it was necessary to have the best possible values for the half-lives of ^{88}Zr, ^{87}Zr, ^{87}Y^{m}, and ^{87}Y^{g}. Although half-lives for these isotopes have been reported in the literature,[6,11–15] recent improvements in counting techniques and computer analyses warranted redetermination of these values. Further, we determined branching ratios for the decay of ^{87}Zr to ^{87}Y^{m,g} and the photon intensity for the 484.9-keV gamma ray from the decay of ^{87}Y^{g}.

A. Determination of the half-life of ^{88}Zr

The half-life for ^{88}Zr used in this study, 82.6±0.2 d, was determined by Bayhurst and confirmed by Butler.[3] The ^{88}Zr was produced on the Tandem Van de Graaff accelerator at the Los Alamos National Laboratory by the ^{89}Y(p,2n)^{88}Zr reaction with 22-MeV protons. After irradiation, the yttrium metal target was dissolved and a very thorough yttrium/zirconium separation was performed. The separation time was recorded, and the ^{88}Zr sample was counted once a week for over a year. Both the decay of ^{88}Zr and the growth and decay of its daughter, ^{88}Y, were followed.

B. The determination of the half-life of ^{87}Zr and the branching to ^{87}Y^{m} and ^{87}Y^{g}

The target for the ^{86}Sr(^{3}He,2n)^{87}Zr reaction was strontium oxide enriched in ^{86}Sr (analysis: ^{86}Sr, 95.72%; ^{84}Sr, 0.02%; ^{87}Sr, 1.24%; ^{88}Sr, 3.01%). The oxide was vacuum evaporated to a thickness of ~300 μg/cm^2 on a 25.4 μm beryllium foil. The target foil was bombarded with 16-MeV ^{3}He^{2+} ions on the Los Alamos Tandem Van de Graaff accelerator for ~100 min at 350 nA.

Following bombardment, the beryllium-backed target was dissolved in 6M HCl, about 20 mg of zirconium carrier was added, and the solution was saturated with HCl gas. The solution was then placed on a BioRad AG 1×8 anion exchange resin column (50—100 mesh; 0.8×10 cm). The zirconium was adsorbed on the column, and beryllium, yttrium, and strontium passed through the column quantitatively. The column was washed two or three times with concentrated HCl and the time of the last wash was recorded. The zirconium was eluted with 3M HCl and the column was then washed with 0.2M HCl. The combined eluates containing the zirconium were divided into two parts and zirconium mandelate was precipitated from each by the addition of 15% aqueous mandelic acid.

One sample of zirconium mandelate was mounted for counting in a trochoidal analyzer (positron counter), and positron decay was followed over a 3-d period. A small number of positrons from ^{89}Zr arising from the ^{88}Sr(^{3}He,2n)^{89}Zr reaction gave a small tail that was removed by means of a least squares analysis of the counting data. For the second zirconium mandelate sample, the 381.1- and 484.9-keV gamma rays from the decay of ^{87}Y^{m,g} were carefully counted at early times in a Ge(Li) counter. The ratio of the photon emission rate for the 381.1-keV gamma-ray process to that for the 484.9-keV gamma-ray process at early times is a very sensitive measure of the branching to the ^{87}Y^{m} and ^{87}Y^{g} states from the decay of ^{87}Zr.

The counting data from the trochoidal analyzer, when treated with a least squares program, gave a half-life of 1.684±0.001 h for ^{87}Zr. An independent determination of the half-life from the GAMANAL (Ref. 16) analysis of the following gamma rays: 511.0, 793.8, 1023.9, and 1209.8 keV, gave a value of 1.667±0.006 h. From these results, a final value of 1.68±0.01 h was chosen. Among previously reported values for the half-life of ^{87}Zr are 1.57 h,[12] 1.6±0.1 h,[13] and 1.733±0.008 h.[6]

A computer program was developed for the calculation of the ratio of the 381.1- and 484.9-keV gamma rays as function of the branching to ^{87}Y^{m} and ^{87}Y^{g}. The best fit to the counting data corresponded to a branching of 0.98±0.006 to ^{87}Y^{m}.

C. The determination of the half-lives of ^{87}Y^{m} and ^{87}Y^{g}

The target for the ^{88}Sr(p,2n)^{87}Y^{m,g} reaction was strontium oxide enriched in ^{88}Sr (analysis: ^{88}Sr, 99.84%; ^{87}Sr, 0.11%; ^{86}Sr, 0.05%; ^{84}Sr, <0.1%). Target preparation and dimensions were identical to those for the ^{86}Sr target. Two target foils were bombarded, one for 5 min and the other for 80 min at ~7 μA with 17.0-MeV protons on the Tandem Van de Graaff accelerator noted above.

The latter sample was counted immediately in a low energy photon spectrometer (LEPS) Ge(Li) counter, 16×4.75 mm, with a resolution of 800 eV at 380 keV. The sample that had been irradiated for 5 min was counted on another Ge(Li) counter, 52×47 mm (~95 cm^3), with an efficiency of 18.5%.

The decay of the 381.1-keV gamma ray from ^{87}Y^{m} was followed on both counters for many days. The data were processed with the GAMANAL computer program,[16] and a least squares analysis gave a half-life of 13.38±0.01 h from the large Ge(Li) counter and 13.36±0.01 h from the LEPS counter. The value of 13.37±0.03 h was taken as the half-life of the isotope. Other values in the literature are 13.2±0.2 h (Ref. 14) and 12.5±0.2 h.[15]

The decay of the 484.9-keV gamma ray from ^{87}Y^{g} plus that of the 388.4-keV gamma ray from ^{87}Sr^{m}, in equilibrium with the 484.9-keV gamma ray, was followed for

about 70 d on a 7.6×7.6 cm NaI crystal. A least squares program was run on the data to remove the small background of ^{88}Y present. A detailed analysis of the data gave a half-life of 79.6±0.2 h for $^{87}Y^{g}$ compared to the previously reported value of 80.3±0.3 h.[14]

D. The determination of relative photon emission rates from ^{87}Y with $^{87}Sr^{m}$ in equilibrium

Gamma rays from samples containing ^{87}Y with $^{87}Sr^{m}$ in transient equilibrium were measured with five Ge(Li) detectors which had been carefully calibrated for efficiency versus gamma-ray energy.[16] The samples also contained ^{88}Y and ^{91}Y, but the amounts were such that they did not interfere with the measurement of the 484.9-keV gamma ray from ^{87}Y or the 388.4-keV gamma ray from $^{87}Sr^{m}$. The relative photon emission rate, $I_\gamma(484.9)/I_\gamma(388.4)$, was found to be 1.100±0.005 for the equilibrium sources. The uncertainty in this result is based on the estimated accuracy with which the relative counting efficiency of these two gamma rays is known. After correcting for parent-daughter decay, the $I_\gamma(^{87}Y)/I_\gamma(^{87}Sr^{m})$ ratio is

$$1.100\times\lambda_{Sr}/(\lambda_{Sr}-\lambda_{Y})=1.140\ .$$

The value of I_γ for the 388.4-keV gamma ray from $^{87}Sr^{m}$ decay is 0.8226±0.0015.[6] We, therefore, obtain $I_\gamma=0.938\pm0.005$ for the 484.9-keV gamma ray from ^{87}Y decay. The value given in Ref. 6 is 0.922±0.010.

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