

3.1-Hour Y^{90m}

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A 3.1-hr isomer, Y^{90m} , has been produced by neutron capture in yttrium. The isomeric transition consists of two coincident gamma rays of nearly equal intensity and energies, 203 kev and 480 kev. Mass and atomic number assignment has been made by cross bombardment and chemical separations. Modes of production were $Y^{89}(n,\gamma)Y^{90m}$ (thermal and epithermal neutrons) and $Nb^{93}(n,\alpha)Y^{90m}$, and $Zr^{90}(n,p)Y^{90m}$ (14-Mev neutrons).

INTRODUCTION

THE 3.1-hr isomer of yttrium-90 first observed and reported by Lyon, Eldridge, and Bate¹ recently has been ascribed variously to Y^{92} ,² to Nb^{92m} or Nb^{93m} ,³ an unassigned yttrium isotope,⁴ and most recently to Y^{90m} .⁵

The original mass and atomic number assignment made by Lyon, Eldridge, and Bate was based on neutron activation of highly purified Y_2O_3 in ORNL reactors, chemical separation of yttrium activities, and beta- and gamma-ray measurement. Because of the studies reported in references 2 and 3 above it seemed worthwhile to establish that this activity produced by other methods and ascribed to other nuclides was in fact the 3.1-hr isomer, Y^{90m} . Consequently, the work described below using 14-Mev neutrons was performed. The original studies reported in reference 1 are summarized briefly in Part A below.

EXPERIMENTAL

A. Neutron Activation of Yttrium Oxide

Highly purified yttrium oxide samples were irradiated in the pneumatic tube of the ORNL Graphite Reactor for periods ranging from a few minutes to several hours. Examination of the irradiated yttrium by use of a NaI(Tl) gamma-ray spectrometer equipped with a multichannel analyzer indicated the presence of two gamma rays of energy 203 kev and 480 kev. Chemical separation of yttrium was performed using precipitation techniques and ion-exchange chromatography.⁶ The activity observed followed yttrium chemistry. The irradiations were repeated a number of times using targets of yttrium oxide from different suppliers. In

one series of irradiations electromagnetically-separated yttrium was used. The decay of the two gamma rays was followed and the half-life found to be 3.1 ± 0.1 hr.

The ratio of effective cross section for production of the 64-hr Y^{90} to the effective cross section for production of the 3.1-hr Y^{90m} was calculated for a number of positions and conditions in the ORNL Graphite Reactor and the ORNL Low Intensity Test Reactor (LITR). The activity of the 3.1-hr Y^{90m} was found by integration of the 480-kev gamma-ray photopeak and calculation of the total number of γ rays by use of the method described by Lazar.⁷ One gamma ray per transition was assumed. The activity of the 64-hr Y^{90} was found by beta-counting and aluminum-absorption data by use of a Geiger-Muller counter. The ratio $\sigma_{3.1}/\sigma_{64}$ was calculated:

$$\frac{\sigma_{3.1}}{\sigma_{64}} = \frac{A_{3.1}}{A_{64}} \frac{(1 - e^{-(0.693/3.1)t})}{(1 - e^{-(0.693/64)t})}, \quad (1)$$

where $A_{3.1}$ = activity of 3.1-hr isomer at end of irradiation time t , and A_{64} = activity of 64-hr isomer at end of irradiation time t . This ratio $\sigma_{3.1}/\sigma_{64}$ was found to range from 9.1×10^{-3} for Cd-shielded irradiations in the LITR to 1.4×10^{-3} for irradiations in a thermal area in the ORNL graphite reactor where the ratio of epithermal to thermal neutrons was $\sim 10^{-5}$. These cross section ratios for a particular position were independent of both irradiation time and source- Y_2O_3 material. From these experiments the reaction $(n,2n)$ leading to an isomer of Y^{88} and the possibility of successive neutron capture in Y^{90} to give isomers of Y^{91} or Y^{92} were eliminated. Y^{89} has a 16-sec isomer; it is the known daughter of Zr^{89} . Swann and Metzger⁸ by neutron inelastic-scattering techniques found no excited state of energy below 910 kev in Y^{89} . In studies of the $Y^{89}(n,\gamma)Y^{90}$ reaction, however, Bartholomew, *et al.*⁹ observed a 203-kev level. From the neutron capture experiments in this work and the data reported in references 8 and 9, the assignment to Y^{90m} was made.

Throughout these experiments, and those described in Part B, no evidence was found for the existence of any negatrons of 3.1-hr half-life. Beta-measurements

* Operated for the U. S. Atomic Energy Commission by Union Carbide Corporation.

¹ W. S. Lyon, J. S. Eldridge, and L. C. Bate, Oak Ridge National Laboratory Report ORNL-2866, 1959 (unpublished), p. 41.

² R. W. Fink, Annual Progress Report, Department of Chemistry, University of Arkansas, Fayetteville, January, 1960 (unpublished), p. 7.

³ M. Bocciolini, G. di Caporiacco, L. Foa, and M. Mando, *Nuovo cimento* **16**, 780 (1960).

⁴ D. R. Koehler, W. L. Alford, and C. E. Mandeville, Presented at the Southeastern Section of the American Physical Society Meeting, March, 1961 (unpublished).

⁵ L. Haskin and R. Vandenbosch (private communication, 1961).

⁶ B. H. Ketelle and G. E. Boyd, *J. Am. Chem. Soc.* **69**, 2800 (1947).

⁷ N. H. Lazar, *IRE Trans. Nuclear Sci.* NS-5, No. 3, 138 (1958).

⁸ C. P. Swann and F. R. Metzger, *Phys. Rev.* **100**, 1329 (1955).

⁹ G. A. Bartholomew, P. J. Campion, J. W. Knowles, and G. Manning, *Nuclear Phys.* **10**, 590 (1959).

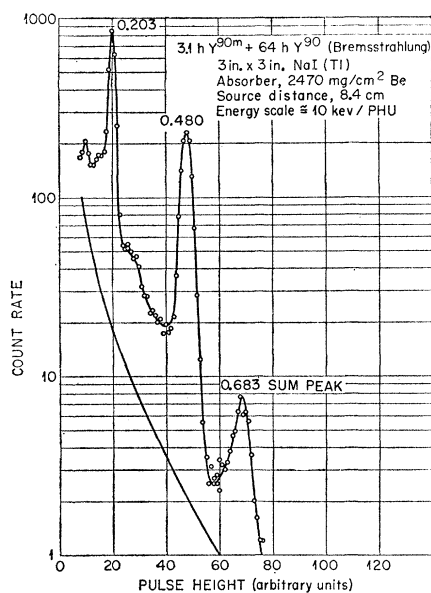


Fig. 1. Gamma-ray spectrum of Y^{90m} produced by $Y^{89}(n,\gamma)Y^{90m}$. Y_2O_3 target in ORNL graphite reactor.

in all these experiments were complicated by the presence of the 2.26-Mev beta group from 64-hr Y^{90} which was produced along with the Y^{90m} . Attempts to establish that the 3.1-hr activity decays to the 64-hr Y^{90} 2- level by observation of the growth of the 64-hr Y^{90} were unsuccessful due to the large amount of 64-hr Y^{90} produced which made it statistically unlikely to observe the slight growth due to the decay of Y^{90m} .

Gamma-gamma coincidence measurements established that the two gamma-rays are in coincidence. Their relative intensities were observed in these experiments to be approximately equal—an exact integration of the 203-keV γ ray was difficult because of the great bremsstrahlung contribution from the 64-hr Y^{90} (Fig. 1). The 683-keV photopeak in Fig. 1 is the coincidence-sum peak as indicated by calculation of the summing effect⁷ and measurement at extended distances from the detector.

B. 14-Mev Neutron Experiments

Workers at the University of Arkansas² reported finding an activity with a gamma-ray of 470 keV and ~ 3.6 -hr half-life which accompanied an yttrium separation from an irradiation of niobium using 14.8-Mev neutrons. This they tentatively assigned to Y^{92} postulating the reaction $Nb^{93}(n,2p)Y^{92}$. Bocciolini and di Caporiacco³ observed both the 203-keV and the 480-keV gamma rays from 14.8-Mev neutrons on niobium and measured the half-life as 3.18 hr. They assigned the activity to either Nb^{92m} or Nb^{93m} . In the present work 0.6 g of 99.99% Nb metal was irradiated with ~ 14 -Mev neutrons ($\sim 1.1 \times 10^8$ n/cm² sec) for 3 hr. The ORNL Biology Division Cockcroft-Walton accelerator and the (d,t) reaction was used. Decay of

the γ -emitting activities produced was followed by use of a 3 in. \times 3 in. NaI(Tl) crystal and a Nuclear Data 256-Channel analyzer. The 203-keV and 480-keV γ rays from Y^{90m} were observed and identified by decay. In addition, the 10-day Nb^{92} activity was identified through decay of its 930-keV γ -ray photopeak. Nb^{92} was produced by the reaction $Nb^{93}(n,2n)Nb^{92}$. Integration of the two gamma photopeaks as described previously⁷ indicated the two gamma rays in Y^{90m} are of nearly equal intensity (Fig. 2). The 64-hr Y^{90} was identified by beta-counting and decay measurements.

These data support the conclusion that the reactions $Nb^{93}(n,\alpha)Y^{90}$ and $Nb^{93}(n,\alpha)Y^{90m}$ occurred, and that the activities observed by Fink² and Bocciolini *et al.*³ were in fact Y^{90m} .

Again no evidence for growth of the 64-hr Y^{90} was observed. However the effect would be expected to be small ($\sim 5\%$ maximum); counting statistics were $\sim \pm 3\%$.

Koehler, Alford, and Mandeville⁴ observed the 3.1-hr activity from 14-Mev neutron irradiation of both niobium and zirconium. They ascribe the activity to an unidentified yttrium isomer.

In the work reported here, 0.8 g of zirconium metal enriched¹⁰ in Zr^{90} to 98.6% was irradiated with 14-Mev neutrons for 30 min at a flux of $\sim 1 \times 10^9$ n/cm² sec. Again the (d,t) reaction and the ORNL Physics Division Cockcroft-Walton accelerator were used. By use of the instrumentation and techniques described above the

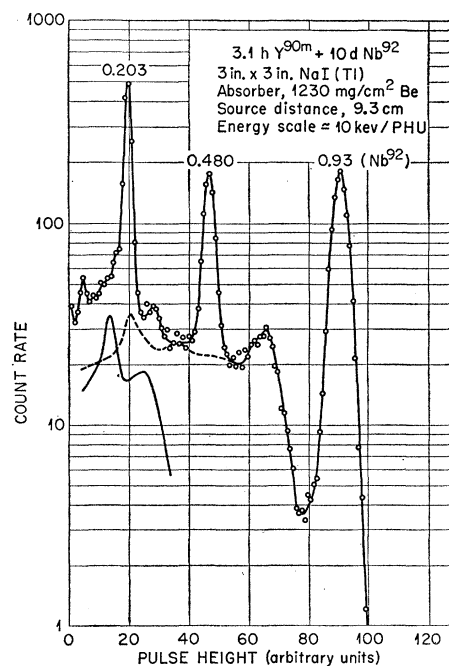


Fig. 2. Gamma-ray spectrum of Y^{90m} and Nb^{92} produced by 14-Mev neutrons on Nb metal.

¹⁰ Enriched Zr^{90} was obtained on loan from the ORNL Isotopes Division.

two gamma-ray photopeaks from Y^{90m} were identified by energy and decay measurements. Seventy-nine hour Zr^{89} was identified by gamma-ray energy and decay measurements. It was produced by $Zr^{90}(n,2n)Zr^{89}$. The Y^{90m} was produced by $Zr^{90}(n,p)Y^{90m}$ and is the activity observed by the workers in reference 4.

DISCUSSION AND CONCLUSIONS

The original assignment¹ of the 3.1-hr activity to Y^{90m} has been confirmed through cross bombardment. The two gamma rays are in coincidence and of nearly equal intensity.

No beta particles or electrons with 3.1-hr half-life were observed. This indicates the 3.1-hr activity is an isomeric transition in Y^{90} . The data reported herein are inconclusive as to whether the 3.1-hr isomer feeds the 64-hr Y^{90} . If it should, the 203-keV level would lie above the 2^- beta-emitting level in Y^{90} and presumably would have the level assignment 3^- as determined by Bartholomew *et al.*⁹ The 203-keV γ transition would thus be M1; the predicted K conversion coefficient as calculated using the tables of Sliv and Band¹¹ is ~ 0.02 which is in agreement with the observation reported above regarding no observed negatrons. Haskin and Vandenbosch⁵ report that the growth of Y^{90} from Y^{90m} was observed. They propose a decay scheme in which the 683-keV level is given a 7^+ assignment. Thus, the 483-keV gamma-transition would be M4; the predicted K conversion coefficient is ~ 0.06 . The calculated

lifetime τ_γ , where

$$\tau_\gamma = T_{\frac{1}{2}}(1 + \alpha_{\text{total}})/\ln 2 \quad (2)$$

(and α_{total} is small), is in agreement with the predicted value as discussed by Goldhaber and Sunyar.¹² τ_γ calculated from Eq. (2) is 1.6×10^4 sec; the theoretical value computed from normalized lifetime energy relations for M4 transitions with spin correction is $\tau_\gamma = 1.1 \times 10^4$.¹¹

An adventitious result of these later experiments has been the measurement of a number of 14-MeV neutron cross sections in this region of the periodic table. These data, which include cross sections for production of both Y^{90} and Y^{90m} , will be presented in a subsequent paper.

Note. Cline *et al.*,¹³ by neutron irradiation of Y^{89} have confirmed the experiments reported in part A above. On the basis of gamma-gamma directional correlation and internal conversion measurements, these workers propose spin and parity assignments for levels in Y^{90m} which are in agreement with those suggested by Haskin and Vandenbosch.⁵

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¹¹ L. A. Sliv and I. M. Band, Leningrad Physico-Technical Institute Report, 1956 [translation: Report 57ICC K1, issued by Physics Department, University of Illinois, Urbana, Illinois (unpublished)].

¹² M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 906 (1951).

¹³ J. E. Cline, R. L. Heath, C. W. Reich, and E. H. Turk, Bull. Am. Phys. Soc. **6**, 228 (1961).

10-MeV Proton Reaction Cross Sections Compared with Surface and Volume Absorption Optical Models of the Nucleus*

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(p,n) cross sections were measured at 9.85 MeV for self-supporting thin targets of Al, Ti, Fe, Co, Ni, Cu⁶³, Cu⁶⁵, Rh, Ag, Sn, Ta, and Au. $(p,2n)$ contributions were calculated using the statistical model of the nucleus for Rh, Ta, Ag, and Au. Charged-particle emission was assumed negligible in Ta and Au because of small Coulomb penetrabilities. Approximate proton reaction cross sections were obtained by adding (p,n) and $(p,2n)$ cross sections to (p,p') and (p,α) cross sections previously reported by Meyer and Hintz. These results were compared with volume absorption and surface absorption optical-model calculations of proton reaction cross sections. The parameters for both model calculations were obtained prior to this work by fitting proton elastic scattering and polarization data. The results indicate a surface-absorption potential rather than a volume-absorption potential.

IN order to obtain approximate reaction cross sections for the nuclei, Al, Ti, Fe, Co, Ni, Cu⁶³, Cu⁶⁵, Rh, Ag, Sn, Ta, and Au, (p,n) cross sections were measured at

9.85 MeV and added to (p,p') and (p,α) cross sections previously measured.¹ The Rh, Ta, Ag, and Au reaction cross sections were corrected for $(p,2n)$ contributions

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¹ V. Meyer and N. M. Hintz, Phys. Rev. Letters **5**, 207 (1960).