Excitation functions and isomeric cross-section ratios for the $^{90}Zr(n,p)^{90}Y^{m,g}$ and $^{91}Zr(n,p)^{91}Y^{m,g}$ processes

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Excitation functions were measured for the $^{90}Zr(n,p)^{90}Y^{m+g}$ and $^{91}Zr(n,p)^{91}Y^{m+g}$ reactions over the neutron energy range of 6.5 to 10.6 MeV. Use was made of the activation technique in combination with radiochemical separation and "low-level" β^- counting. From the available experimental data isomeric cross-section ratios were determined. Statistical model calculations taking into account precompound effects were performed for the formation of both the isomeric and ground states of the product. The calculational results on the total (n,p) cross sections agree well with the experimental data. The measured and calculated cross sections for the formation of the isomeric states also agree up to an incident neutron energy of 10 MeV; at higher energies, however, the calculated values are appreciably higher. In the case of isomeric cross-section ratios good agreement was found between experiment and theory up to 10 MeV; at higher energies the calculated values are consistently higher.

I. INTRODUCTION

Studies of excitation functions of neutron threshold reactions on medium mass nuclei are of considerable significance for testing nuclear models. Furthermore, isomeric cross-section ratios are of fundamental interest for studying the spin dependence of the formation of isomeric states. The available information on the latter, especially as a function of incident neutron energy, is often unsatisfactory. We chose to investigate the isomeric pairs $^{90}Y^{m,g}$ and $^{91}Y^{m,g}$ formed in (n,p) reactions on ^{90}Zr and ^{91}Zr, respectively.

II. EXPERIMENTAL

Cross sections were measured radiochemically over the neutron energy range of 6.5–10.6 MeV. The relevant techniques are described below.

A. Irradiations, neutron flux monitoring, and γ-ray spectroscopy

High purity ZrO_2 (specpure, Johnson Matthey, U.K.) of natural isotopic composition was packed in PVC capsule (1.1 cm $\phi \times 2.6$ cm; wall thickness $=0.1$ cm), and the monitor foils (Al or Fe, each 200 μm thick) were attached in front and at the back of the capsule. The sample was irradiated in the 0° direction with quasimonoenergetic neutrons produced via the $^2H(d,n)^3He$ reaction on a D_2 gas target at the Jülich variable energy compact cyclotron CV28. The characteristics of the neutron source and the method of calculation of mean neutron energies effective at the sample have been described in detail earlier (cf. Refs. 1 and 2). The neutron flux density in the energy region up to 7.7 MeV was determined via the $^{56}Fe(n,p)^{56}Mn$ monitor reaction (cf. Ref. 3) and between 8.2 and 10.6 MeV via the $^{27}Al(n,\alpha)^{24}Na$ reaction (cf. Ref. 4). The mean neutron flux densities ranged between 8.2×10^6 and 2.7×10^7 cm^{-2}s^{-1}.

Each irradiated sample was counted on a Ge(Li) detector and the cross sections for the $^{90}Zr(n,p)^{90}Y^m$ and $^{91}Zr(n,p)^{91}Y^m$ reactions were determined. The results have been reported in detail in Ref. 5.

B. Radiochemical separation and source preparation

The products $^{90}Y^g$ ($T_{1/2}=64.1$ h; $E_\beta=2.3$ MeV; $I_\beta \geq 99\%$) and $^{91}Y^g$ ($T_{1/2}=58.5$ d; $E_\beta=1.54$ MeV; $I_\beta=100\%$) are pure β^- emitters. It was therefore mandatory to separate them radiochemically. The irradiated ZrO_2 ($\sim$5 g) was digested with concentrated HF in a plastic beaker, diluted with water, and heated on a water bath until complete dissolution occurred. The acid was then evaporated, the residue dissolved in 0.5M HF, and 200 mg each of Y and Sr carriers added. After vigorous shaking the mixture was allowed to stand for several hours, and the YF_3 and SrF_2 precipitates centrifuged off. The precipitates were then taken up in conc. H_2SO_4, transferred to a teflon beaker, and HF fumed off. The residue was diluted and the $SrSO_4$ precipitate centrifuged off. Thereafter the solution was made alkaline by adding excess of 20% NaOH solution, the precipitated $Y(OH)_3$ collected, washed with water, dissolved in dilute HCl, and reprecipitated as oxalate. The yttrium oxalate was then converted to Y_2O_3 by heating at 900 °C. Thereafter it was transferred to an aluminum planchet, spread over a surface of 4 cm ϕ, fixed with glue, and used for counting. The chemical yield was about 80%. It was determined gravimetrically, and after the end of experiment, via

thermal neutron activation analysis. The two results agreed within 5%.

C. Measurement of β^- activity

Each separated Y_2O_3 ($^{90}Y+^{91}Y$) sample was subjected to "low-level" β^- ray counting. The measurement was started several days after the end of irradiation in order to allow complete decay of $^{90}Y^m$ and $^{91}Y^m$ to $^{90}Y^g$ and $^{91}Y^g$, respectively. Counting was done for about one year. In the decay curves two components with half-lives of 64 h and 58.5 d could be unambiguously analyzed, and are attributed to $^{90}Y^g$ and $^{91}Y^g$, respectively. The count rates were corrected for chemical yield, absorption in the counter window, finite-size geometry, and efficiency of the detector.

D. Calculation of cross sections and errors

The decay rates of both $^{90}Y^g$ and $^{91}Y^g$ were corrected for contributions from background neutrons (gas out–gas in results and breakup of deuterons on D_2 gas) as described for other reactions.[1,2,5] From the corrected decay rates and the mean neutron flux densities the cross sections were calculated using the well-known activation equation. The principal sources of error and their magnitudes in our activation measurements have been described in detail earlier.[1,2] The present studies involving radiochemical separations and β^- counting have somewhat larger errors. Combining the individual errors in quadrature, the total error for each cross-section value was obtained.

III. NUCLEAR MODEL CALCULATIONS

Calculations were performed in the frame of the statistical model, utilizing the exciton model formalism for preequilibrium particle emission, the width-fluctuation corrected Hauser-Feshbach formula for first chance emission from the equilibrated system, and the evaporation formula for higher chance emission. The choice of parameters was consistent with that for earlier calculations on neutron induced reactions on ^{93}Nb (Ref. 6) and ^{92}Mo (Ref. 7).

We generated the particle transmission coefficients in the spherical optical model using the potential of Delaroche *et al.*[8] for neutrons (with the modifications described in Ref. 6) and of Mani *et al.*[9] for protons. The γ-ray strength functions were derived in a completely analogous way to those in calculations on Nb and Mo,[6,7] with a giant resonance form for electric dipole radiation, and energy independent strength functions according to the Weisskopf model for $M1$, $E2$, $M2$, $E3$, and $M3$ radiation. At the neutron binding energy, the $M1/E1$ ratio was prescribed according to the work of McCullagh *et al.*[10] whereas the normalization of the strengths of the other radiation types relative to $E1$ was according to Weisskopf's estimate. An overall normalization of all γ-ray strength functions was carried out, applying a factor which had been obtained by averaging the individual normalization constants required for a proper description of experimental neutron capture cross sections for several nuclei in the mass region $A=88-103$.

The features of the exciton model formalism are summarized in the description of the computer code STAPRE used;[11] we chose the constant in the squared matrix element for internal transitions to be 300 MeV^3 and used a pairing energy shift in the exciton state densities.

In the equilibrated nuclei, the excited states were described by means of the discrete level information as far as possible, i.e., considering 19 levels in ^{91}Zr (up to 2.694 MeV; Ref. 12), 16 levels in ^{91}Y (up to 2.279 MeV; Ref. 12), 18 levels in ^{90}Zr (up to 4.28 MeV; Ref. 13), and 14 levels in ^{90}Y (up to 1.760 MeV; Ref. 13). The branching ratios for γ transitions between the discrete levels are well known for ^{91}Y; for ^{90}Y, the corresponding information is rather scarce. However, Ref. 13 gives for each energy level the final states populated by γ rays starting from that initial level. If this information is correct, the branching ratios of only one level out of the 14 considered are of any influence on the isomeric ratio, in that its γ transitions lead to both the isomeric and the ground state. All the others feed either the isomeric or the ground state. Above the region of discrete levels, the excited states of all relevant nuclei were treated as a continuum described by the back-shifted Fermi gas model. The choice of level density parameters was guided by the compilation of Dilg *et al.*[14] The parameters were verified by checking the reproduction of cumulative level densi-

TABLE I. Fast neutron induced activation cross sections.

Mean neutron energy effective at ZrO_2 sample[a] (MeV)	Reaction cross section (mb) $^{90}Zr(n,p)^{90}Y^{m+g}$	$^{91}Zr(n,p)^{91}Y^{m+g}$
6.48±0.19	3.1±0.5	3.0±1.0
7.18±0.13		3.7±0.7
7.70±0.13	5.6±0.6	4.9±0.7
8.21±0.13	11.2±1.2	6.7±0.9
8.72±0.14	9.7±1.2	8.0±1.3
9.19±0.19	15.7±2.2	9.9±1.5
9.64±0.22	15.2±2.2	12.2±2.0
10.13±0.23	20.5±2.3	14.5±2.4
10.61±0.22	21.7±2.4	

[a]The deviations do not give errors; they show energy spreads due to angle of emission.

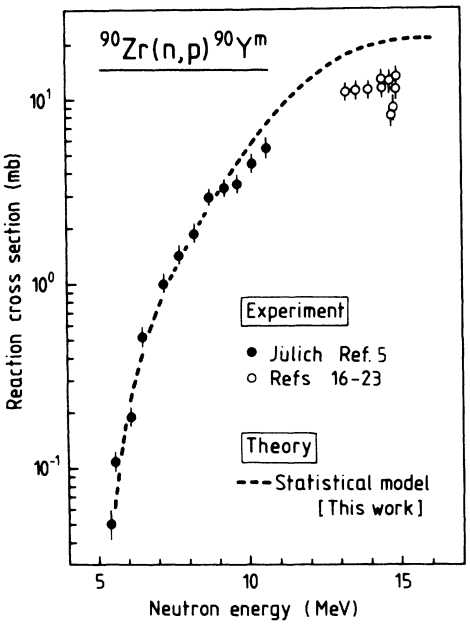

FIG. 1. Excitation function of the $^{90}Zr(n,p)^{90}Y^{m}$ reaction.

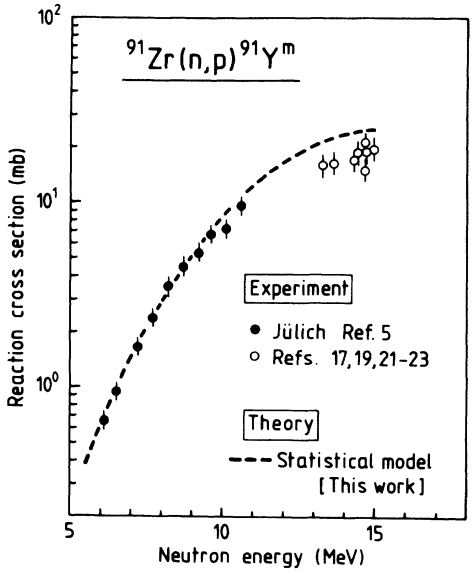

FIG. 3. Excitation function of the $^{91}Zr(n,p)^{91}Y^{m}$ reaction.

ties and resonance spacings. If some nuclei encountered in the calculations on Nb (Ref. 6) and Mo (Ref. 7) were relevant to the present calculations, the parameter values chosen were maintained. For ^{91}Zr, the compilation by Mughabghab *et al.*[15] contains a larger value for the *s*-wave neutron resonance spacing than that in the work of Dilg *et al.*,[14] and in addition a value for the *p*-wave neutron resonance spacing. Hence, the *a* parameter of ^{91}Zr was reduced with respect to the value given in Ref. 14.

IV. RESULTS AND DISCUSSION

The results of measurements are presented in Table I. The total error in the $^{90}Zr(n,p)^{90}Y^{m+g}$ reaction cross section amounts to between 11 and 16%, and that in the $^{91}Zr(n,p)^{91}Y^{m+g}$ cross section between 33% (near the threshold) and 17% (at 10 MeV).

The experimental excitation functions of the four processes under consideration, viz., $^{90}Zr(n,p)^{90}Y^{m}$, $^{90}Zr(n,p)^{90}Y^{m+g}$, $^{91}Zr(n,p)^{91}Y^{m}$, and $^{91}Zr(n,p)^{91}Y^{m+g}$, are shown in Figs. 1–4, respectively. The data for the

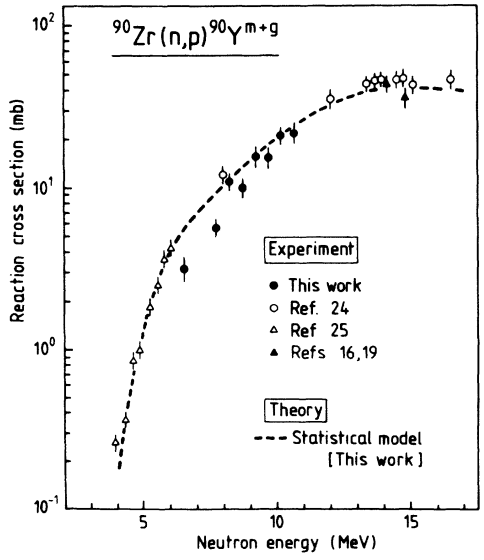

FIG. 2. Excitation function of the $^{90}Zr(n,p)^{90}Y^{m+g}$ process.

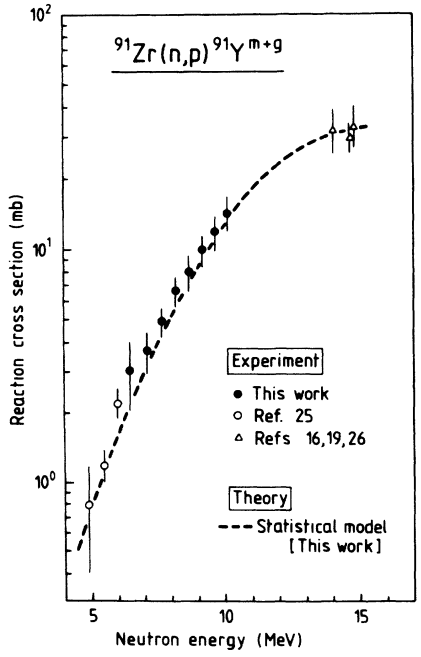

FIG. 4. Excitation function of the $^{91}Zr(n,p)^{91}Y^{m+g}$ process.

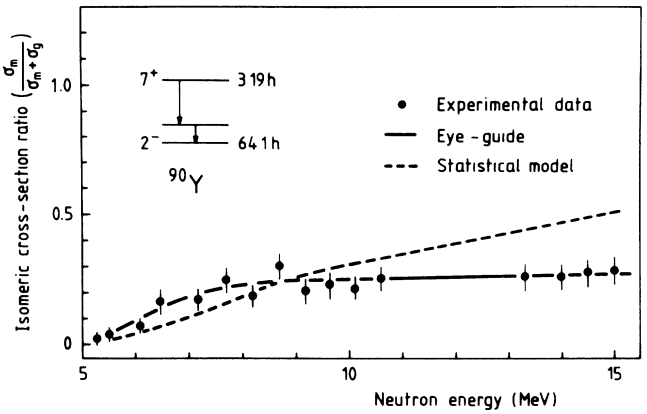

FIG. 5. Isomeric cross-section ratio for the isomeric pair $^{90}Y^{m,g}$ [formed via (n,p) reaction on ^{90}Zr] plotted as a function of incident neutron energy.

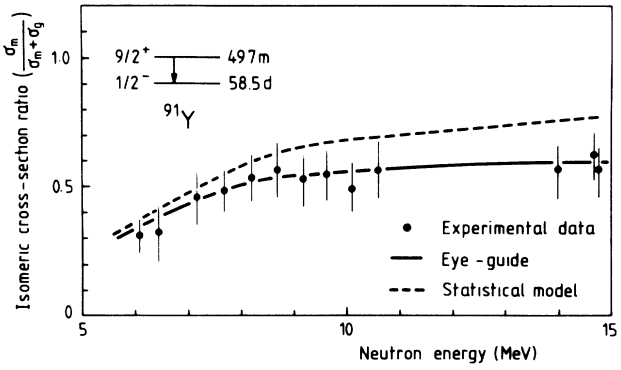

FIG. 6. Isomeric cross-section ratio for the isomeric pair $^{91}Y^{m,g}$ [formed via (n,p) reaction on ^{91}Zr] plotted as a function of incident neutron energy.

isomeric states were reported earlier, in the energy region up to 10.6 MeV from Jülich[5] and around 14 MeV by several groups.[16–23] A few literature data were available for the formation of the ground states (cf. Refs. 16, 19, and 24–26). Our measurements provide a more extensive data base. The results of nuclear model calculations are also shown in Figs. 1–4.

The measured cross sections for the $^{90}Zr(n,p)^{90}Y^{m+g}$ and $^{91}Zr(n,p)^{91}Y^{m+g}$ reactions are reproduced well by our calculation over the whole energy range (cf. Figs. 2 and 4). The fit to the $^{90}Zr(n,p)^{90}Y^{m}$ and $^{91}Zr(n,p)^{91}Y^{m}$ cross sections is acceptable from threshold up to an incident neutron energy of about 10 MeV; above this energy, however, the isomeric cross sections are severely overestimated (cf. Figs. 1 and 3).

The isomeric cross-section ratios $(\sigma_m/\sigma_m+\sigma_g)$ were deduced from the experimental cross-section data. For those energy points where we measured both σ_m and $\sigma_m+\sigma_g$ (Ref. 5 and this work), the ratio was obtained directly. In other energy regions, smooth curves were drawn through the experimental points and therefrom the ratios were estimated. The values are shown in Figs. 5 and 6, and are compared with the results of model calculations. The theoretically calculated ratios are too high for energies above 10 MeV. Varying the branching ratios of the discrete levels of ^{90}Y and ^{91}Y (within the limits of their uncertainties mentioned above) does not lead to any better agreement as it affects the excitation function in a similar manner over the whole energy range. Likewise, changes in the absolute normalization of the γ-ray strength functions and the relative $M1/E1$ normalization were not suited to adjust the calculated (n,p) excitation functions (of the metastable states) to the measurements.

The good reproduction of the excitation functions for the combined formation of the isomeric and ground states [total (n,p) cross section] indicates that the amount of preequilibrium proton emission is described correctly as a function of incident energy. Its spin distribution, however, may be approximated poorly by the formulation used, which assumes the relative weights for each spin value to be equal in preequilibrium and equilibrium emission. The expected preference of preequilibrium decay to populate states of somewhat lower spin would reduce the activation cross section of the high spin isomers. However, even if 40% of the preequilibrium contribution to the cross section for the formation of $^{91}Y^{m}$ were transferred to the ground state, this would only halve the deviation between the calculated and experimental isomeric cross-section ratio.

A correction of the absorption cross section for losses to direct reactions as applied in Refs. 6 and 7 was not done. Evidently, it would slightly improve the description of the excitation functions for the isomers, somewhat deteriorate the reproduction of the excitation functions for the ground states, and leave the isomeric cross-section ratios unchanged.

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