Neutron Activation Cross Sections at 14.8 MeV for Rubidium, Strontium, Zirconium, and Niobium†

L. HUSAIN,* A. BARI, AND P. K. KURODA
Department of Chemistry, University of Arkansas, Fayetteville, Arkansas 72701
(Received 15 October 1969)

The activation cross sections for 18 (n, p), (n, α), and $(n, 2n)$ reactions have been measured with 14.8-MeV neutrons. The radioactivity measurements were carried out with a Ge(Li) detector. The cross sections measured, in millibarns, are ^{27}Al$(n, p)^{27}$Mg, 73±5; ^{85}Rb$(n, 2n)^{84m}$Rb, 714±50; ^{85}Rb$(n, 2n)^{84g}$Rb, 621±75; ^{85}Rb$(n, p)^{85m}$Kr, 6±1; ^{87}Rb$(n, 2n)^{86m}$Rb, 584±40; ^{87}Rb$(n, 2n)^{86g}$Rb, 833±60; ^{87}Rb$(n, p)^{87}$Kr, 7±1; ^{84}Sr$(n, 2n)^{83}$Sr, 395±75; ^{86}Sr$(n, 2n)^{85m}$Sr, 360±30; ^{86}Sr$(n, p)^{86m}$Rb, 9±1; ^{88}Sr$(n, 2n)^{87m}$Sr, 344±30; ^{88}Sr$(n, p)^{88}$Rb, 20±2; ^{90}Zr$(n, 2n)^{89m}$Zr, 143±15; ^{90}Zr$(n, 2n)^{89g}$Zr, 342±25; ^{90}Zr$(n, p)^{90m}$Y, 13±1; ^{90}Zr$(n, \alpha)^{87m}$Sr, 4.6±0.5; ^{93}Nb$(n, 2n)^{92m}$Nb, 455±25; and ^{93}Nb$(n, \alpha)^{90m}$Y, 5.8±0.5. The measured cross sectons are compared with those calculated by evaporation theory. The $(n, 2n)$ measured cross sections agree well with the calculated values but the agreement between calculated and measured values for (n, p) reactions is poor.

INTRODUCTION

RECENT surveys[1–6] of 14–15-MeV neutron activation cross sections for $(n, 2n)$, (n, p), and (n, α) reactions indicate the need for more accurate determinations of cross sections for these reactions, especially for medium and heavy nuclei. The cross-section values measured by various investigators are generally in poor agreement with each other. The data become quite sparse when either the natural abundance of the target nucleus is low or when the half-life of the product nucleus is either very short or very long.

In the present investigation, a total of 18 (n, p), (n, α), and $(n, 2n)$ reaction cross sections at 14.8 MeV for rubidium, strontium, zirconium, and niobium isotopes were measured. Natural and enriched isotope samples were used. The reaction yields were determined by an absolute measurement of the γ activities of the product nuclei. A Ge(Li) detector was used for the measurement of the γ activities.

If the incident neutron flux ϕ remains constant during the irradiation time T, the cross section σ, for a parent activity, is defined by the basic equation

$$\sigma = A^0/[1-\exp(-\lambda T)]\phi n, \qquad (1)$$

where n is the number of the target nuclei in the sample, A^0 is the activity of the product at the end of the irradiation, and λ is the decay constant of the activity produced in the reaction. The irradiation conditions were selected so that Eq. (1) could be applied with only minor corrections, i.e., flux was kept as constant as possible for the short-lived product nuclei, and for the long-lived products T was such that λT was small. The monitor reactions chosen were such that the half-life of the monitor product nucleus differed as little as possible from that of the product nuclei in the reaction under study.

We also report some measurements of isomer ratios. Here the cross section for production of the metastable state is given by an equation like Eq. (1). The ground state, however, includes the contribution from the metastable states. The relevant equation for the number of ground-state atoms, $N_g(T, t)$, measured at a time t after the end of an irradiation of duration T, is

$$N_g(T,t) = \{N_g^0 - [N_m^0 B\lambda_m/(\lambda_g-\lambda_m)]\}\exp(-\lambda_g t) + [N_m^0 B\lambda_m/(\lambda_g-\lambda_m)]\exp(-\lambda_m t), \qquad (2)$$

where m and g refer to the metastable and the ground state, respectively, B is the branching ratio, $N_m^0 = n\phi\sigma[1-\exp(-\lambda_m t)]/\lambda_m$, and

$$N_g^0 = n\phi[(B\sigma_m+\sigma_g)/\sigma_g][1-\exp(-\lambda_g T)] + n\phi[B\sigma_m/(\lambda_g-\lambda_m)][\exp(-\lambda_g T)-\exp(-\lambda_m T)].$$

EXPERIMENTAL PROCEDURE

Neutrons were produced via the ^{3}H$(d, n)^4$He reaction by bombarding a thick tritium-titanium target with 400-keV deutrons in the University of Arkansas Cockroft-Walton accelerator. The samples, which were of the order of 1 cm^2 in area, subtended an angle of ±30°. For this arrangement 90% of the neutron beam lies in the energy range of 14.8±0.2 MeV. The neutron flux, which was of the order of $3\times10^9 n$cm^{-2} sec^{-1}, was fairly constant over the irradiation periods. The corrections for flux variations were made when needed.

Target samples for irradiations were made by thorough mixing of 10–300 mg of spectroscopically pure target material with spectroscopically pure ferric or aluminum oxide. The mixture was sealed in 0.00025-in.-thick Mylar. When enriched isotopes were

† Work supported by the U.S. Atomic Energy Commission, under Contract No. AT-(40-1)-3235.
* Present address: Department of Chemistry, Brookhaven National Laboratory, Upton, N.Y. 11973.
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TABLE I. Radioactive products studied, the decay γ-ray energies, and the branching ratios.

Nuclide	Half-life[a]	Radiation detected[a] (energy in keV)	Branching ratio[a]
^{27}Mg	9.5 min	840	0.70
^{85m}Kr	4.4 h	305	0.13
^{87}Kr	76 min	403	0.84
^{84m}Rb	20 min	216	0.37
		250	0.65
		464	0.32
^{84g}Rb	33 day	880	0.74
^{86m}Rb	1 min	560	1.00
^{86g}Rb	18.7 day	1074[b]	0.088
^{88}Rb	18 min	910	0.13
		1863	0.21
^{83}Sr	32 h	762	0.332[c]
^{85m}Sr	70 min	231	0.85
^{87m}Sr	2.8 h	388	1.00
^{90m}Y	3.2 h	202	0.97
		482	0.91
^{89m}Zr	4.2 min	588	0.87
^{89g}Zr	78.4 h	910	0.99
^{92m}Nb	10.2 day	934	0.99

[a] Taken from Lederer *et al.* (Ref. 7) unless otherwise mentioned.
[b] This work.
[c] Reference 8.

used, the target material was sandwiched between two high-purity aluminum foils.

At the end of the irradiation the product and the monitor activities were measured, without any chemical separation, with a Ge(Li) detector having a 4-cm^2 active area and 7-mm depletion depth and a resolution of 3.7 keV FWHM (full width at half-maximum for 1332 keV). The efficiency of this counter was calibrated with known standards, obtained from Tracerlab, Inc., covering an energy range of 88 to 1373 keV. Selected γ-ray photopeaks were measured as a function of time for at least three half-lives, so that the nuclides were characterized by means of their γ-ray energies and half-lives. The count rates of the samples were such that the statistical errors were of the order of a few percent. Table I lists the product nuclides that were measured, the radiations which were selected for measurements, half-lives, and the branching ratios (taken from Lederer *et al.*,[7] except ^{83}Sr taken from Etherton *et al.*[8]). Decay curves were analyzed by the least-squares analysis to give the activity at the end of the irradiation. After necessary corrections the cross sections were calculated using Eq. (1).

The reaction selected as the primary standard for monitoring the neutron flux was ^{56}Fe$(n, p)^{56}$Mn. The 2.6-h^{56}Mn product has a conveniently measurable 847 keV γ ray emitted in 99% of the decays. The cross section for this reaction was taken to be 105±2 mb, which is the weighted mean of three published values[9–11] computed by standard statistical methods. Since the monitor material was natural iron, the measured ^{56}Mn activity included a contribution from the ^{57}Fe$(n, np)^{56}$Mn reaction. This was corrected for by use of the measured cross section for this reaction, 6.1±2.6 mb, reported by Chittenden *et al.*[12] This correction amounted to less than 1%. The reactions ^{27}Al$(n, \alpha)^{24}$Na and ^{27}Al$(n, p)^{27}$Mg were also used as a monitor in some cases. The cross sections for these reactions were taken as 114±6 mb[13] and 73±5 mb (this work), respectively.

The principal errors in the cross sections measured in this work arise from the uncertainties in the counter efficiency (±5%), the decay schemes, and neutron flux (±5%). The error limits attached to the cross-section values (Table II) are from these sources of error. The cross-section values reported are generally means of two or more measurements.

RESULTS AND DISCUSSION

In Table II the cross sections measured in this work are given, along with the reported values with references in parentheses, and the theoretical values from evaporation theory. We remark the following:

(n, p) reactions. Many measured values have been reported for the ^{27}Al$(n, p)^{27}$Mg reaction.[13–17] Our value is in agreement with four of the six values.[14–17] Also, we report a lower error limit. The cross section for the reaction ^{86}Sr$(n, p)^{86m}$Rb is measured for the first time. Our values for ^{88}Sr$(n, p)^{88}$Rb and ^{90}Zr$(n, p)^{90m}$Y reactions are in good agreement with the values given in the literature,[18–21] and those for ^{85}Rb$(n, p)^{85m}$Kr and ^{87}Rb$(n, p)^{87}$Kr reactions are about 30% higher than the preliminary results of Rao *et al.*[22]

(n, α) reactions. We have measured the ^{90}Zr$(n, \alpha)^{87m}$Sr and ^{93}Zr$(n, \alpha)^{87m}$Sr and ^{93}Nb$(n, \alpha)^{90m}$Y reaction cross

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TABLE II. 14.8-MeV neutron cross sections (mb).

Reaction	σ (expt) This work	σ (expt) Literature	σ (calc)
$^{25}Al(n, p)^{27}Mg$	73±5	71±9(14) 77±8(15) 53±5(13) 82±10(16) 97±10(17) 68±8(22)	
$^{85}Rb(n, p)^{85m}Kr$	6±1	4.1±0.4(22)	2.7
$^{87}Rb(n, p)^{87}Kr$	7±1	4.9±0.5(22)	<1.4
$^{86}Sr(n, p)^{86m}Rb$	9±1		16.5
$^{88}Sr(n, p)^{88}Rb$	20±2	17.7±3.5(18) 18±3(19) 30±2(20)	<1.4
$^{90}Zr(n, p)^{90m}Y$	13±1	12±4(21)	5.7
$^{90}Zr(n, \alpha)^{87m}Sr$	4.6±0.5	3.0±0.2(23) 2.8±0.4(24) 4.2(25) 194±107(18)	
$^{93}Nb(n, \alpha)^{90m}Y$	5.8±0.5	5±2(21) 5.9±2.0(26)	
$^{85}Rb(n, 2n)^{84m}Rb$	714±50	926±61(27) 800±90(28) 478±48(22)	
$^{85}Rb(n, 2n)^{84g}Rb$	621±75	756±161(27) 374±28(28) 414±41(22)	
$^{85}Rb(n, 2n)^{84m+g}Rb$	1335±90	1682±222(27) 1174±94(28) 1420±60(29) 1520±80(30) 680±80(20) 892±63(22)	1209
$^{87}Rb(n, 2n)^{86m}Rb$	584±40	932±150(27)	
$^{87}Rb(n, 2n)^{86g}Rb$	833±60	1619±250(27)	
$^{87}Rb(n, 2n)^{86m+g}Rb$	1417±72	2551±350(27) 1550±160(29) 1200±50(30) 820±130(20) 973±97(22)	1326
$^{84}Sr(n, 2n)^{83}Sr$	395±75	380(20) 180±10(30) 1770±177(31)	260
$^{86}Sr(n, 2n)^{85m}Sr$	360±30	222±25(27) 312±50(20) 246±12(29)	
$^{86}Sr(n, 2n)^{85g}Sr$	323±30[a]	280±10(20)	
$^{86}Sr(n, 2n)^{85m+g}Sr$	683±42	592±51(20)	628
$^{88}Sr(n, 2n)^{87m}Sr$	344±30	330(6) 222±25(27) 215±24(20)	
$^{90}Zr(n, 2n)^{89m}Zr$	143±15	191±15(27) 168±12(32) 142±21(29) 84±12(33)	
$^{90}Zr(n, 2n)^{89g}Zr$	342±25	762±82(27)	
$^{90}Zr(n, 2n)^{89m+g}Zr$	485±29	953±97(27) 502±36(20) 677±51(34) 800±60(38)	471
$^{93}Nb(n, 2n)^{92m}Nb$	455±32	460±25(29) 550±50(36) 310±10(20) 500±100(26)	

[a] Based on an isomer ratio of 1.12±0.18, Ref. 20.

sections, and our values are in good agreement with previously reported values.[21,23–26]

(n, 2n) reactions. We have measured the cross sections for the $^{85}Rb(n, 2n)^{84m}Rb$ and $^{85}Rb(n, 2n)^{84g}Rb$ reactions. There are three previously measured values available for these reactions.[22,27,28] Our values for the cross sections for these reactions are in best agreement with those of Minetti and Pasquarelli.[27] There are several values reported for the $^{85}Rb(n, 2n)^{84(m+g)}Rb$ reaction, but there is quite a spread among them.

We have also measured the cross section for $^{87}Rb(n, 2n)^{86m}Rb$ and $^{87}Rb(n, 2n)^{86g}Rb$. Here the cross-section values measured separately for these reactions are only those of Minetti and Pasquarelli.[27] Our values for these reactions are 50% of those reported by these authors. However, our values for the $^{87}Rb(n, 2n)^{86(m+g)}Rb$ reaction are in good agreement with two of the other four reported values.[20,22,29,30]

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TABLE III. Isomer ratios (σ_m/σ_g).

Reactions	Spin of metastable state	Spin of ground state	σ_m/σ_g (this work)	σ_m/σ_g (literature)
^{85}Rb$(n, 2n)^{84m,g}$Rb	6	2	1.15±0.6	1.22±0.27[a] 1.07±0.05[b] 1.15±0.16[c] 0.33±0.05[d]
^{87}Rb$(n, 2n)^{86m,g}$Rb	6	2	0.70±0.07	0.58±0.13[a]
^{90}Zr$(n, 2n)^{89m,g}$Rb	$\frac{1}{2}$	$\frac{9}{2}$	0.42±0.05	0.25±0.03[a] 0.21±0.04[e]

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[d] Reference 28.
[e] Reference 35.

The cross sections measured in this work for ^{84}Sr$(n, 2n)^{83}$Sr, ^{86}Sr$(n, 2n)^{85m}$Sr, and ^{88}Sr$(n, 2n)^{87m}$Sr are in agreement with at least one of the reported values (Table II).[6,20,27,29–31] The cross section for the ^{90}Zr$(n, 2n)^{89}$Zr reaction have been measured by several authors.[27,29,32–35] Here the spread among the measured values is not as large as for the other reactions, and our value falls in the middle. There is only one value reported for the ^{90}Zr$(n, 2n)^{89g}$Zr reaction; our value is 50% of this measurement.[27] The cross section measured in this work for the ^{93}Nb$(n, 2n)^{92m}$Nb reaction is in good agreement with three of the four previously measured values.[20,26,29,36]

Evaporation-theory calculations. Table II also compares the measured values with those calculated on the basis of evaporation theory.[37] These calculations used Weisskopf's[38] expression for the probability per unit time for the emission of a particle j with kinetic energy between ϵ and $\epsilon+d\epsilon$:

$$P_j(\epsilon)\,d\epsilon=\gamma_j\sigma\epsilon[W(f)/W(i)]d, \qquad (3)$$

where $\gamma_j=(g_jm_j/\pi^2\hbar^3)$, with g_j and m_j the number of spin states and the mass of the particle j, respectively, σ is the cross section for the inverse reaction, and $W(f)$ and $W(i)$ are the level densities of the final and initial nuclei at their respective excitation energies.

The nuclear level density $W(E)$ with excitation energy E is given by

$$W(E)=C\exp\{2[a(E-\delta)]^{1/2}\}, \qquad (4)$$

where C is a constant, δ is the pairing energy, and a is the level-density parameter. Using the author's[37] version but substituting $a=\frac{1}{10}A$, where A is the mass number, we computed the values of P_j.

Once Eq. (3) is integrated, the cross section σ_j for the j-particle emission can be calculated from

$$P_j=\int_{\epsilon_1}^{\epsilon_2}P_j(\epsilon)\,d\epsilon \qquad (5)$$

and

$$\sigma_j=P_j\sigma_T, \qquad (6)$$

where $\sigma_T=\pi R^2$. The value of R was taken as $1.5A^{1/3}\times10^{-13}$cm.

The numerical calculations were done by the Monte Carlo method on a CDC 6600 computer. The calculated values are total cross sections (sum of metastable and ground states, if there is a metastable state) and are included in Table II. The calculated values are in good agreement with the measured values for the $(n, 2n)$ reactions, but the agreement between the calculated and the measured cross sections for the (n, p) reaction is rather poor. In quantitative terms, the comparisons between the calculated and the measured cross sections for (n, p) reactions are not very meaningful because of the low probability for charged-particle emission in evaporation calculations. Here we have included the calculated (n, p) values only to show the general disagreement between the calculated and the measured (n, p) cross sections.

Isomer ratios. In Table III the isomer ratios measured in this work are given along with the reported values.

ACKNOWLEDGMENT

The authors wish to thank Dr. K. L. Chen for the computer calculations and Dr. William Rubinson for constructive criticism of the manuscript. We also wish to thank Philip Pile for carrying out the irradiations.

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