14-MeV-Neutron Production of Isomeric States for Several Rare-Earth Elements

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(Received 23 March 1965; revised manuscript received 21 May 1965)

Fast-neutron cross sections have been determined for the following reactions, using 14-MeV neutrons from the $H^3(d,n)He^4$ reaction: $Y^{89}(n,n'\gamma)Y^{89m}$, 400 mb; $Nd^{142}(n,2n)Nd^{141m}$, 545 mb; $Sm^{144}(n,2n)Sm^{143m}$, 400 mb; $Tb^{159}(n,2n)Tb^{158m}$, 160 mb; $Er^{168}(n,2n)Er^{167m}$, 190 mb. Possible sources of error affecting the cross-section measurements have been discussed and evaluated numerically. The over-all accuracy was determined to be about ±12%.

INTRODUCTION

IN the course of developing suitable neutron-activation, direct-instrumental-analysis techniques for determining the individual rare-earth elements in various alloys and compounds, several fast-neutron reactions with the rare-earth nuclides, of unexpected magnitude, were observed. Although fairly extensive cross-section data have been reported for the rare-earth elements,[1–3] the cross sections for some of their isomeric states have not been determined. Alford and Koehler[4] observed the isomeric state of Sm^{143m} in measuring the total $(n,2n)$ reactions with Sm^{144}, but only mentioned that it accounted for about half of the total cross section. Accordingly, a systematic study was undertaken to determine the cross sections for any previously unreported reactions.

EQUIPMENT

A model 9500 Texas Nuclear neutron generator utilizing the $H^3(d,n)He^4$ reaction provided the source of fast neutrons. All irradiations were made with the sample positioned directly in front of the target, and subtending only a small angle to the deuterium beam.

Neutron-flux monitoring was accomplished with a BF_3 detector situated outside the concrete generator shield and connected to a scaler in the counting room. The on-off function of the scaler was automatically controlled through the irradiation timer. Samples were counted in a 3- by 3-in. NaI(Tl) well-type crystal coupled to a EMI 9578B photomultiplier tube, and signals from this tube were fed into a 400-channel analyzer and associated scaler.

A pneumatic shuttle system was used to transfer the sample between the neutron generator and the counting laboratory. The samples, contained in 1.4 cm diam by 5.3 cm long polystyrene vials, were precisely positioned a minimum distance in front of the generator target assembly by a tight-fitting sleeve in the plastic transfer pipe. Irradiation time and the delay time prior to counting were regulated to within a fraction of a second by a pair of precision timers and associated controls.

The sample specimens were 1-g amounts of the various rare-earth oxides and, as contained in the plastic vials, occupied a volume 1.1 cm in diam by about 0.8 cm in length. The oxides were prepared at this Center and had a purity greater than 99.9%.

EXPERIMENTAL

Each rare-earth oxide sample was irradiated in a flux of about 5×10^7 $n/cm^2/sec$. This flux level was determined by direct comparison to the count rate obtained from the BF_3 neutron monitor. During each run the beam current was carefully stabilized manually to ensure that an essentially constant neutron flux was maintained.

The BF_3 monitor was calibrated by equating the count rate to the neutron flux as determined by irradiating a 1-g sample of SiO_2 positioned in an identical geometry to that used for the rare-earth irradiations. Silica was chosen for a standard because it is readily available in pure form, its cross section[5] of 243±22 mb for the $Si^{28}(n,p)Al^{28}$ reaction is quite well accepted, and its Al^{28} product decays with a conveniently short half-life. Furthermore, the effects of competing (n,α) and (n,p) reactions on other Si isotopes, which would indicate a higher than actual flux, could be readily minimized by short irradiations and by gamma spectrometry.

Irradiation time was kept to a minimum, commensurate with the half-life of the rare-earth isotope being produced. Actual times varied from 6 sec for Er^{167m} having a 2.5-sec half-life, to 30 sec for the 64-sec Sm^{143m}.

The sample activity as detected by the NaI crystal was corrected for background and converted to disintegrations by applying the calculated efficiencies given by Verheijke[6] for a 3- by 3-in. well crystal. Corrections were also made for gamma-ray attenuation in the sample and photofraction efficiency of the crystal when required. No attempt was made to correct for the slight difference in neutron attenuation between the rare-earth oxides and the SiO_2 standard. The samples were counted for time periods appropriate to their respective half-lives. Delay and irradiation times were adjusted as

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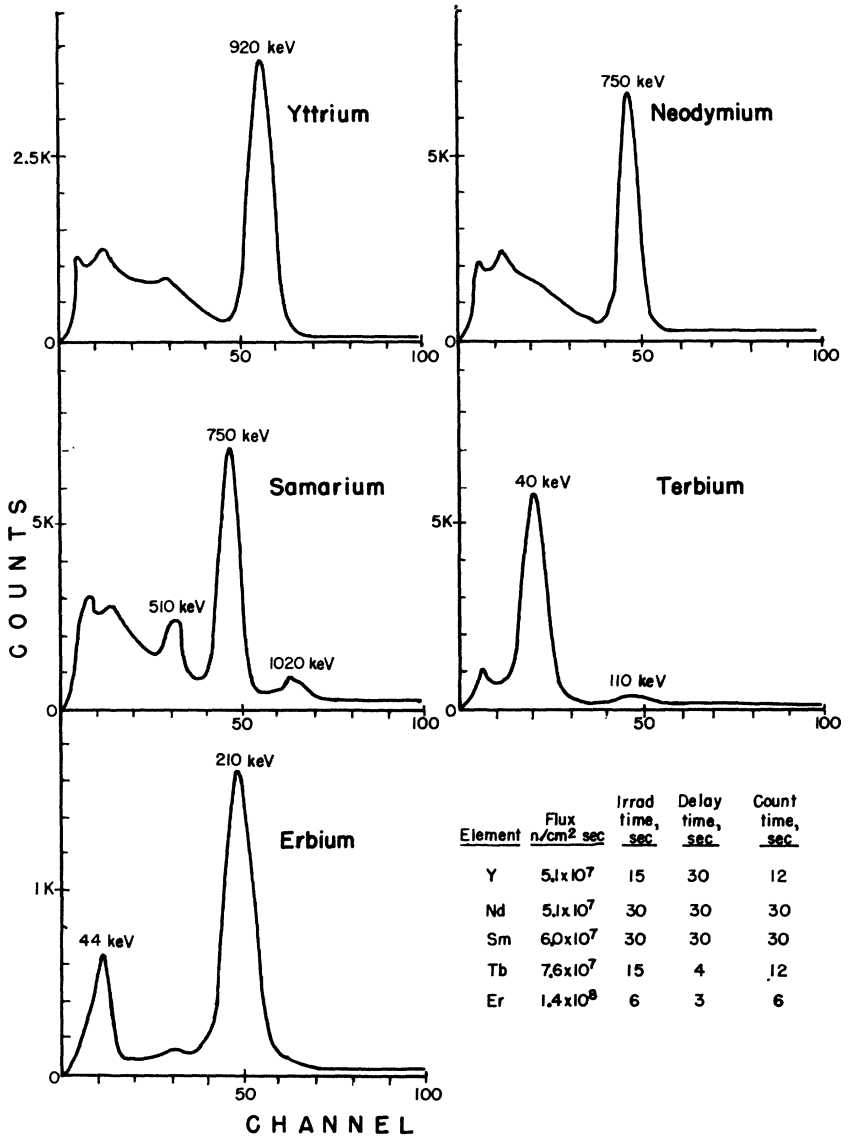

Element	Flux n/cm² sec	Irrad time, sec	Delay time, sec	Count time, sec
Y	5.1×10^7	15	30	12
Nd	5.1×10^7	30	30	30
Sm	6.0×10^7	30	30	30
Tb	7.6×10^7	15	4	12
Er	1.4×10^8	6	3	6

FIG. 1. Gamma spectra for five rare-earth elements.

necessary to keep the dead time of the multichannel analyzer under 3%.

The effect of thermal (n,γ) reactions was minimized by surrounding the sample tube with a gadolinium oxide "paint" and also with a 0.3-mm cadmium sheet. Experiments showed that the thermal flux at sample position was about 10^{-4} of the fast flux.

RESULTS AND DISCUSSION

All five reported reactions produce isomeric transitions, and the associated gamma spectra are quite simple. Figure 1 shows the gamma spectra of the observed reactions along with the irradiation and counting data.

As pointed out by Wille and Fink,[1] neutrons having energies less than 14.9 MeV may be present in the total neutron flux due to both the $H^2(d,n)He^3$ reaction and scattering of the 14.9-MeV neutrons and subsequent energy loss. However, the threshold energies required for $(n,2n)$ reactions in the rare-earth region, and for the $Si^{28}(n,p)Al^{28}$ reaction, are in excess of the 2.95-MeV neutrons carried off by the d-D reaction. A new target produces few of these d-D neutrons, but as a target is used, more and more of these neutrons are generated as the tritium in the target is replaced with deuterium. Although these lower energy neutrons would not interfere through direct interactions with the samples, there is the possibility of their contributing to the neutron monitor count, thereby giving an erroneously high neutron flux value. However, experience has shown that at a constant neutron monitor reading, the specific activity produced in various isotopes is essentially

constant irrespective of the amount of use the target has undergone. Apparently, the probability for the d-D neutrons escaping through the heavy concrete shielding and being detected by the monitor is negligible.

The output spectrum for our particular neutron source-sample geometry was calculated by the method of Ricci,[7] wherein it was assumed that the deuteron beam was monatomic and homogeneous. Over 91% of the neutrons fell within the 14.3- to 14.9-MeV range. Consequently, in calculating the cross sections, it was assumed that the observed activities were produced solely by 14.6±0.3 MeV neutrons.

Table I gives the cross sections determined for the five reactions along with the presently and previously[8–11] observed values for the half-lives and gamma energies. The literature values for the half-lives were used in the cross-section determination as it was assumed that these were more accurate than our observed values.

Sources of errors and their estimated magnitudes include: (1) uncertainty in the silicon cross section, ±9.1%; (2) short term variations in flux, ±2.5%; (3) counting efficiencies, ±3.5%; (4) half-lives of the produced isotopes, ±4.9% for Y^{89m} to ±0.4% for Tb^{158m}; (5) irradiation and delay times, ±1.8 and ±4.6%, respectively, for Er^{167m} to ±0.4 and ±1.0% for the longer lived isotopes; (6) gamma-ray attenuation, as determined experimentally from an attenuation curve, ±5.0% at 40 keV to ±2.0% at 900 keV; and (7) statistical errors, ±2.5% for Sm^{143m} to 0.6% for Tb^{158m}.

Possible interferences caused by other neutron reactions are listed below. The maximum error associated with these interferences is estimated to be about 3%.

$Y^{89}(n,n'\gamma)Y^{89m}$

There is no possibility of interference by other reactions. The gamma ray arises from a pure isomeric transition with no internal conversion.

$Nd^{142}(n,2n)Nd^{141m}$

No internal conversion was noted. The possibility exists for interference from the $Nd^{142}(n,\alpha)Ce^{139m}$ reaction which decays with essentially the same half-life and the same energy gamma ray. However, the magnitude of the observed reaction is far too high since (n,α) reactions do not exceed 10 mb for elements in this region of the periodic table.

$Sm^{144}(n,2n)Sm^{143m}$

The $Sm^{144}(n,\alpha)Nd^{141m}$ reaction could interfere. As mentioned above, the possibility of this type of reaction of this magnitude appears extremely remote in the rare-earth region. The Sm^{143m} decays to 8.8-min Sm^{143g}, a positron emitter. The annihilation radiation from the positron was counted, and the half-life curve thus obtained showed definitely that, initially, the positron activity was increasing. The growth of the daughter Sm^{143g} from the parent Sm^{143m} was thereby indicated.

TABLE I. Cross sections for five rare-earth elements.

Reaction	Cross section, mb	Half-life, sec This work	Half-life, sec Previous	Gamma or x-ray, keV This work	Gamma or x-ray, keV Previous
$Y^{89}(n,n'\gamma)Y^{89m}$	400±47	16.8	16.1±0.3[a]	920	913[a]
$Nd^{141}(n,2n)Nd^{141m}$	545±60	61.3	61±2[b]	750	755[b]
$Sm^{144}(n,2n)Sm^{143m}$	400±44	61.0	64±3[b]	750	748[b]
$Tb^{159}(n,2n)Tb^{158m}$	160±19	10.2	11.0±0.1[c]	110 40	111[d] 44
$Er^{168}(n,2n)Er^{167m}$	190±24	2.1	2.5±0.1[c]	210 44	208[e] [e]

[a] Reference 8.
[b] Reference 9.
[c] Reference 10.
[d] Reference 11.
[e] No energy listed for the Er x ray.

$Tb^{159}(n,2n)Tb^{158m}$

No interferences were noted. However, the Tb^{158m} decays with internal conversion of the 110-keV gamma ray.[11] Consequently, in determining the cross section, both the 110-keV peak and the 44-keV x-ray peak produced by the conversion were counted. It was assumed that there was initially one gamma for each 40-keV x ray.

$Er^{168}(n,2n)Er^{167m}$

$Er^{166}(n,\gamma)Er^{167m}$ could cause interference, but the reaction cross section is far too large to be produced by 14.7-MeV neutrons. In order to produce the observed activity from thermal neutrons, the cross section would have to be in the order of 2000 b. Alexander and Brinckmann[12] gave a value of only 15 b for this reaction; consequently, the thermal flux could not contribute much more than 1% to the production of the isotope. While the activity could be due to (n,γ) reactions produced by epithermal neutrons, with energies greater than the Cd and Gd cutoff energies, this possibility seems extremely remote since no other reactions due to epithermal fluxes of the required magnitude have been observed. The reaction $Er^{167}(n,n'\gamma)Er^{167m}$ could interfere; however, the probability for the $(n,2n)$ reaction is greater and, consequently, any contribution due to the $(n,n'\gamma)$ reaction has been ignored. Er^{167m} does exhibit some internal conversion,[7] and it was assumed that one 44-keV x ray was formed per conversion.

The total error, obtained by taking the square root of the sum of the squares of the individual errors dis-

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cussed above, ranges from $\pm 11.0\%$ for Sm^{143m} to $\pm 12.8\%$ for Er^{167m}.

The described reactions are the predominant ones produced by short-time irradiation of the respective rare-earth elements. Since the gamma spectra are simple, they are thereby well suited for use in activation analyses. Several of these reactions are currently being utilized by the Bureau of Mines at the Reno Metallurgy Research Center for analyzing the respective rare-earth constituents in titanium alloys and other compounds.

PHYSICAL REVIEW VOLUME 139, NUMBER 6B 20 SEPTEMBER 1965

Muon Capture in $(p\mu d)^+$ Molecules*

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(Received 5 April 1965)

The $(p\mu d)^+$ molecules and μ-He^3 atoms formed in liquid hydrogen were used to detect neutrons from muon capture by deuterons and from muon capture by He^3. In the experiment, a purified muon beam was stopped in a target containing ultra-pure liquid hydrogen with 0.32% deuterium added. Neutron–gamma-ray discriminating detectors and oscilloscope photography enabled us to measure the time distribution of neutrons from the following rare muon-capture processes:

$$\mu^- + d \rightarrow \nu + 2n,$$
$$\mu^- + He^3 \rightarrow \nu + n + d$$
$$\rightarrow \nu + 2n + p.$$

Knowledge of the time distributions of the various $(p\mu d)^+$ hyperfine states and of the μ-He^3 atoms was used to unravel the various muon-capture rates of interest. Converted to the μd atom case, the measured rate gives 365 ± 96 sec^{-1} for muon capture from the doublet state of μd. This is to be compared with the theoretical rate of 334 sec^{-1} predicted by the current phenomenological muon-capture theory; and it provides the simplest verification of the Pauli exclusion-principle effect in muon capture. The neutron rate from muon capture by He^3 was determined to be $(1.20 \pm 0.17) \times 10^3$ sec^{-1}.

I. INTRODUCTION

UNTIL recently, the only accurate data concerning the interaction between the muon and the atomic nucleus were obtained from total-capture-rate measurements with complex nuclei.[1,2] The inherent difficulty of the attempt to extract the basic form of the muon-nucleon weak coupling then naturally suggested exploring the possibility of observing muon capture by protons and other light nuclei, such as deuteron and He^3, where an understanding of the nuclear structure may be more accessible.

Recently, measurements of the muon-capture rate in $(p\mu p)^+$ molecular ions were carried out independently by using hydrogen-bubble-chamber[3] and scintillation-counter techniques[4]; and the rate of the μ-He^3 reaction into neutrino and H^3 has since been obtained with rather good accuracies[5] (i.e., of the order of a few percent). Although the hydrogen results are statistically limited and there are still some uncertainties in the detailed nuclear structure of He^3 and H^3, the experimental results can be said to agree remarkably well with the theory. But even with the accuracy of the He^3 measurements, the rather intricate, theoretically predicted pion-induced coupling effects in muon capture are only crudely tested, simply because the He^3 rate is not very

* Work supported in part by U. S. Office of Naval Research under contract No. ONR-266 (72).

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