

it is possible to do so without loss of coincidence efficiency.

ACKNOWLEDGMENTS

The author is greatly indebted to Dr. Paul H. Stelson who first suggested the possibility of carrying

out this research at Oak Ridge National Laboratory. His generous and invaluable assistance was greatly appreciated. Thanks are also due to Mr. James Johnson and other members of the staff at Oak Ridge whose excellent cooperation contributed substantially to this project.

Activation Cross Sections for 14.8-Mev Neutrons and Some New Radioactive Nuclides in the Rare Earth Region*

R. G. WILLE† AND R. W. FINK‡

Department of Chemistry, University of Arkansas, Fayetteville, Arkansas

(Received September 21, 1959)

Activation cross sections on 27 stable nuclides of elements Ba, La, Ce, Pr, Nd, Sm, Eu, Gd, Dy, Er, Yb, and Lu were measured for 14.8 ± 0.8 -Mev neutrons. Highly enriched isotopes were used as targets in most cases, and in a few instances radiochemical separations were performed whenever it was necessary and possible in view of the product half-lives. The measured cross sections for $(n,2n)$ reactions were found to agree within an order of magnitude with predictions from statistical evaporation theory. However, experimental values of (n,p) and (n,α) cross sections generally appear to be larger than calculated from continuum theory of the compound nucleus. The cross sections show no significant effects due to the 82-neutron closed shell and, furthermore, the Levkovskii effect, which is quite striking in the low Z region, appears to be negligible for (n,p) and (n,α) reactions in the rare earth region. The $(n,2n)$ cross sections show little variation with mass number

at constant Z , and they exhibit a decrease with increasing mass number at $N=82$.

Several previously unreported activities were observed; their half-lives, assignment, and gamma radiations are as follows: 12 ± 3 min Pr^{148} from the $\text{Nd}^{148}(n,p)$ reaction; 0.5 ± 0.1 min Sm^{157} from the $\text{Gd}^{160}(n,\alpha)$ reaction, 0.57 ± 0.01 Mev gamma; 7 ± 1 min Tb^{168} from the $\text{Dy}^{168}(n,p)$ reaction, 0.18 ± 0.05 -Mev gamma; 3.3 ± 0.5 min Ho^{168} from the $\text{Er}^{168}(n,p)$ reaction, 0.85 ± 0.05 -Mev gamma; 40 ± 10 sec Ho^{170} from the $\text{Er}^{170}(n,p)$ reaction; 4.4 ± 0.4 min Dy^{167} from the $\text{Er}^{170}(n,\alpha)$ reaction; 2.0 ± 0.5 min activity with gammas at 0.18 ± 0.01 , 0.25 ± 0.01 , and 0.36 ± 0.01 Mev which may be Tm^{176} , Er^{173} , or possibly isomeric Yb^{177m} from enriched Yb^{176} bombardments. Tentative assignment of a 5.5 ± 0.5 -min activity to Tm^{174} is suggested from bombardment of enriched Yb^{174} .

INTRODUCTION

IN a previous paper¹ cross sections for some samarium isotopes were reported for 14.8-Mev neutrons as part of a larger study in the rare earth region to determine whether the relative variation in (n,p) and (n,α) cross sections, as pointed out by Levkovskii² for low Z nuclides, persists in higher Z elements. In the low Z (up to $Z=22$) cases examined by Levkovskii,² the relative variation in (n,p) and (n,α) cross-sections at 14 Mev shows an almost integral decrease by factors of 2, 4, or 8 with increasing mass number at constant Z .

It was also of considerable interest in this work to investigate neutron cross sections in the region of the 82-neutron shell closure in order to determine whether shell effects³ noticed in the region of $Z=20$ and $Z=50$

are present in the vicinity of $N=82$. As may be seen from the present results, both the Levkovskii variation and the shell closure appear to have negligible effect on the 14.8-Mev neutron cross sections in the rare earth region.

EXPERIMENTAL

Activation cross sections on 27 stable nuclides of elements barium, lanthanum, cerium, praseodymium, neodymium, samarium, europium, gadolinium, dysprosium, erbium, ytterbium, and lutetium were measured for 14.8 ± 0.8 -Mev neutrons from the $\text{H}^3(d,n)\text{He}^4$ reaction with total fluxes of 10^{10} to 10^{11} neutrons/second from the University of Arkansas 400 kv Cockcroft-Walton accelerator. Samples of natural and enriched rare earths, either as metal or as oxides, were pressed into flat tablets of known weight and area, weighed thin copper monitor foils were placed in front and back of the tablets, and irradiated for periods ranging from a few minutes to about 9 hours.

By absolute beta counting of 9.9 min Cu^{62} (or 12.8 hour Cu^{64} when the length of the bombardment made Cu^{62} a poor monitor) induced in the copper foils, the

* Supported in part by the U. S. Atomic Energy Commission and based in part on the Ph.D. thesis of R. G. Willie, University of Arkansas, 1959 (unpublished).

† Phillips Petroleum Company predoctoral fellow, 1958-59.

‡ Present address: The Gustaf Werner Institute for Nuclear Chemistry, University of Uppsala, Sweden.

¹ R. G. Wille and R. W. Fink, *Phys. Rev.* **112**, 1950 (1958).

² V. N. Levkovskii, *J. Exptl. Theoret. Phys. (U.S.S.R.)* **31**, 360 (1956); **33**, 1520 (1957) [translations: *Soviet Phys. JETP* **4**, 291 (1957); **6**, 1174 (1958)].

³ D. J. Hughes and D. Sherman, *Phys. Rev.* **78**, 632 (1950); H. P. Eubank, R. A. Peck, Jr., and M. R. Zatzick, *Nuclear Phys.* **10**, 418 (1959) ($Z=50$); R. A. Peck, Jr., *Bull. Am. Phys. Soc.* **4**,

9 (1959) ($Z=50$); M. R. Zatzick and H. P. Eubank, *Bull. Am. Phys. Soc.* **4**, 141 (1959) ($Z=20$); and H. P. Eubank (private communication, 1959) ($Z=20$).

flux through the sample was established. All cross sections reported here are based on mean values^{4,5} of 519 mb for the $\text{Cu}^{63}(n,2n)$ reaction and 1020 mb for the $\text{Cu}^{65}(n,2n)$ reaction. Samples and copper monitor foils were counted under identical geometry in an aluminum-walled, methane-flow beta-proportional counter fitted with a 0.9 mg/cm² aluminized Mylar end-window. The counting rates were small enough so that dead-time losses did not exceed 0.8%. Corrections for the following counting factors were made: efficiency (defined as the ratio of events registered to total events in the sensitive volume of the detector), air and window transmission,⁶ saturation backscattering from aluminum,⁷ self-absorption and self-scattering,⁸ and background. For electron-capture branching, the efficiency was estimated from experimental values of the EC/β^+ ratio whenever possible.⁹ For isomeric transitions, the efficiency was estimated from conversion coefficients and the measured K/L conversion ratio.⁹ The efficiency for particulate radiation is essentially unity, while for electromagnetic radiation it could be estimated from the work of Norling¹⁰ and Sullivan.¹¹ All samples were followed until decay was substantially complete. Chemical separations were made¹² to aid identification for barium, lanthanum, cerium, and praseodymium. Elements above neodymium were studied without chemical separation but with the aid of enriched isotopes.

Cross sections were computed from the corrected counting data, after correction to saturation bombardment time, as described previously¹ on the assumption of constant flux during irradiations. During accelerator runs the neutron flux is monitored continually with a

"long counter" for neutrons and an argon-flow proportional counter for alpha-particles from the $\text{H}^3(d,n)\text{He}^4$ reaction; the observed steadiness of the neutron yield ($\pm 5\%$) during short bombardments justifies the constant flux assumption. In long bombardments (2 hours or more), however, the flux may drop gradually by as much as 40% which would cause a large error if 9.9 min Cu^{62} were being used to monitor the yield of a much longer-lived species; in such cases, 12.8 hour Cu^{64} was taken as the monitor.

In addition to monoenergetic 14.8-Mev neutrons from the DT reaction, neutrons of lesser energy may be present owing to (a) partial slowing down by scattering and diffusion from surrounding target objects, and (b) monoenergetic 2.95 Mev neutrons from the DD reaction, which always accompanies the DT reaction when zirconium-tritium targets are bombarded with deuterium ions. Other experiments in this laboratory have shown that the relative abundance of thermal and epithermal neutrons around our target is less than 0.1%, but in the rare earth region even this small amount could be important owing to the extremely large thermal neutron capture cross sections in this region (e.g., Gd^{157} has a total thermal neutron activation cross section of 160 000 barns; Sm^{152} and Eu^{151} exhibit values of 140 and 1400 barns, respectively. Other rare earth nuclides have similar large cross sections for thermal neutrons).⁴ In order to eliminate thermal neutrons, all samples were wrapped in 0.6 mm thick cadmium foil. The number of DD neutrons (2.95 Mev) produced with a new zirconium-tritium target is a small fraction (less than 1%) of the DT yield. In older targets the buildup of the DD yield can amount to 25% or more of the DT yield if an old target is kept in use too long. In this work DD neutrons do not influence the (n,p) and $(n,2n)$ measurements, because the threshold energies usually are greater than 3 Mev. However, for the (n,α) measurements the DD neutrons conceivably could cause errors if the DD and DT (n,α) cross sections differ greatly and if the DD flux were significantly large.

To check on this question, four bombardments were made using zirconium-deuterium targets, from which the maximum obtainable DD flux (7×10^8 DD neutrons/second total) is down from our normal DT fluxes by factors of 10^2 to 10^3 . Samarium and neodymium samples irradiated with DD neutrons gave no detectable (n,α) products, although these reactions are energetically possible. [The (n,γ) reactions in unwrapped samples were, however, observed in these experiments.] It is concluded that the DD flux present in our usual DT irradiations is too small to influence any of the (n,α) measurements.

Another source of error arises in cases where a single product may be formed by two or more reactions, e.g., by $(n,2n)$ and (n,γ) simultaneously on different isotopes. Use of cadmium wrapping and highly enriched isotopes minimized this error.

⁴ *Neutron Cross Sections*, compiled by D. J. Hughes and J. A. Harvey, Brookhaven National Laboratory Report BNL-325 (Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 1955) and revisions.

⁵ L. A. Rayburn, *Bull. Am. Phys. Soc.* **3**, 337, 365 (1958); S. Yasumi, *J. Phys. Soc. (Japan)* **12**, 443 (1957); and A. Poularikas and R. W. Fink, *Phys. Rev.* **115**, 989 (1959).

⁶ G. I. Gleason, J. D. Taylor, and D. L. Tabern, *Nucleonics* **8**, No. 5, 12 (1951).

⁷ B. P. Burt, *Nucleonics* **5**, No. 2, 28 (1949); J. R. Zumwalt, Atomic Energy Commission Report AECU-567, 1950 (unpublished); and L. Yaffe, *Conference on Absolute Beta Counting*, Preliminary Report No. 8 (National Research Council, Washington 25, D. C., 1950).

⁸ W. E. Nervik and P. C. Stevenson, *Nucleonics* **10**, No. 3, 18 (1952); Walton, Thomson, and Croall, Atomic Energy Research Establishment Report AERE-c/R-1136, 1953 (unpublished); B. T. Bowles and G. N. Walton, Atomic Energy Research Establishment Report AERE-c/R-1463, 1958 (unpublished); Cunningham, Sizeland, and Willis, Atomic Energy Research Establishment Report AERE-c/R-2054, 1957 (unpublished); AERE-c/R-1646, 1955 (unpublished); J. Taylor and W. Parrish, *Rev. Sci. Instr.* **26**, 367 (1955); D. C. Conway and J. O. Rasmussen, University of California Radiation Laboratory Report UCRL-2075, 1953 (unpublished); and R. G. Baker and L. Katz, *Nucleonics* **11**, No. 2, 14 (1958).

⁹ D. Strominger, J. M. Hollander, and G. T. Seaborg, *Revs. Modern Phys.* **30**, 585 (1958); *Nuclear Data Cards* (National Research Council, Washington 25, D. C.).

¹⁰ F. Norling, *Phys. Rev.* **58**, 277 (1940).

¹¹ H. M. Sullivan, *Rev. Sci. Instr.* **11**, 356 (1950).

¹² For details, see the Ph.D. thesis of R. G. Wille, University of Arkansas, September, 1959 (unpublished).

Errors in the measurement of the time interval between end of irradiation and start of counting are especially serious for activities with half-lives less than 1 min. In most cases considered here, half-lives were suitably long and care was exercised to minimize the error in the case of shorter-lived nuclides.

By far the largest source of inaccuracy arises in estimating the correction factors for absolute counting¹²; in particular, the self-absorption—self-scattering factor,⁸ f_{ssa} , is the most serious, e.g., a beta group having a maximum energy of 0.3 Mev has $f_{ssa}=1.12$ in a 2 mg/cm² lead nitrate source, and $f_{ssa}=0.55$ in a 20 mg/cm² lead nitrate source; i.e., a 100% change for 10-fold increase in sample thickness in this case. Most of the limits of error on the cross sections reported here arises from the uncertainty in estimating f_{ssa} .

RESULTS

During the course of this study, several previously unreported activities were observed, and mass assignments were made by the use of highly enriched isotopes¹³ and by eliminating other likely reactions by means of cross bombardments. Table I summarizes the results on these new activities. The measured cross sections and their comparison with the literature appear in Table II. Finally, a comparison with calculations from available nuclear reaction theory is given in Table III.

The new activities observed are: 12 ± 3 min Pr^{148} , 7 ± 1 min Tb^{163} , 3.3 ± 0.5 min Ho^{168} , 40 ± 10 sec Ho^{170} , 4.4 ± 0.4 min Dy^{167} , 5.5 ± 0.5 min $\text{Tm}^{174}(\text{?})$, and 2.0 ± 0.5 min which may belong to Tm^{176} , Er^{163} , or Yb^{177m} . The evidence for these new activities is given individually below:

12 min Pr^{148}

The 12-min activity was not observed in bombardments of natural Nd_2O_3 , since a 30 ± 10 min half-life was produced which is an unresolvable mixture of 17

min Pr^{144} , 24 min Pr^{146} , and 15 min Nd^{151} . All other activities (Table II) could be assigned to known species, including 65 ± 10 sec Ce^{139m} , whose yield decreased by a factor of 3 to 4 in 84.6% enriched Nd^{148} (as oxide) bombardments. The yield of the 12 min species in the enriched sample was 3 times greater than that of the unresolved 30 min unassigned activity in the natural neodymium sample. Since the abundance of all neodymium isotopes in the enriched sample was down by a factor of at least 2 relative to Nd^{148} , which was increased nearly 15-fold, the 12-min activity must arise from a reaction on Nd^{148} , the possibilities being $(n,2n)$, (n,α) , (n,γ) , or (n,p) . The $(n,2n)$ reaction may be ruled out since cross sections for $(n,2n)$ reactions in this region are of the order of hundreds to several thousand millibarns, while the cross section measured for the 12 min nuclide is only 3.5 mb (Table II). Bombardment of natural samarium gave no 12-min activity from an (n,α) reaction on Sm^{150} . Although 17.4 min Nd^{151} is produced when 99% enriched Sm^{154} is bombarded,¹ the 12-min activity cannot be due to this, since Nd^{151} could only have been produced by the (n,γ) reaction on Nd^{150} , and thus would have been in larger yield from natural neodymium than from enriched Nd^{148} in which the abundance of Nd^{150} was reduced 2-fold. Likewise, 93% enriched Nd^{150} exhibited no 12-min species after irradiation, thus ruling out Nd^{151} and any possible isomeric state in Nd^{149} .

Chemical separation¹² of cerium from neodymium and praseodymium showed no 12-min activity in the cerium fraction; thus, the 12-min species must arise from an isotope of praseodymium, i.e., from the $\text{Nd}^{148}(n,p)\text{Pr}^{148}$ reaction. Attempts to study the gamma rays from the 12-min species with an NaI(Tl) scintillation spectrometer failed because of masking from 1.8 hour Nd^{149} . However, a gamma at about 0.31 Mev was observed to decay with a 3-min half-life from irradiations of enriched Nd^{148} , but was not observed from

TABLE I. Summary of previously unreported activities and probable mass assignments.

| Observed half-life | Probable assignment | Mode of production | Observed gamma rays (Mev) | Basis of assignment |
|--------------------|-----------------------------|-----------------------------|---------------------------|---|
| 12 ± 3 min | Pr^{148} | $\text{Nd}^{148}(n,p)$ | | Enriched Nd^{148} Chemical separation to eliminate (n,α) product. ^{a,b} |
| 0.5 ± 0.1 min | Sm^{157} | $\text{Gd}^{160}(n,\alpha)$ | 0.57 ± 0.01 | Enriched ^a Gd^{160} |
| 7 ± 1 min | Tb^{163} | $\text{Dy}^{163}(n,p)$ | 0.18 ± 0.05 | Enriched ^a Dy^{163} |
| 3.3 ± 0.5 min | Ho^{168} | $\text{Er}^{168}(n,p)$ | 0.85 ± 0.05 | Enriched ^a Er^{168} |
| 40 ± 10 sec | Ho^{170} | $\text{Er}^{170}(n,p)$ | | Enriched Er^{170} |
| 4.4 ± 0.4 min | Dy^{167} | $\text{Er}^{170}(n,\alpha)$ | | Enriched Er^{170} |
| 5.5 ± 0.5 min | $\text{Tm}^{174}(\text{?})$ | $\text{Yb}^{174}(n,p)$ | | Enriched Yb^{174} |
| 2.0 ± 0.5 min | Tm^{176} | $\text{Yb}^{176}(n,p)$ | 0.18 ± 0.01 | Enriched Yb^{176} Not from Yb^{174} |
| | Er^{173} , or | $\text{Yb}^{176}(n,\alpha)$ | 0.25 ± 0.01 | |
| | Yb^{177m} | $\text{Yb}^{176}(n,\gamma)$ | 0.36 ± 0.01 | |

^a Other likely assignments eliminated by cross bombardments.

^b From fission studies, a 12-min activity was assigned to Pr^{147} and a 1.95-min activity to Pr^{148} [D. C. Hoffman and W. R. Daniels, Bull. Am. Phys. Soc. 4, 372 (1959)].

¹³ Obtained from Stable Isotopes Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

TABLE II. Summary of observed reactions and measured cross sections.

| Reaction | Product | Measured half life | Observed cross section (mb) | | Reference |
|--------------------------|--------------------|--------------------|-----------------------------|------------|-----------|
| | | | This work | Lit. Value | |
| Ba ¹³⁴ (n,2n) | Ba ^{133m} | 38.9±0.1 hours | 940±80 | | |
| Ba ¹³⁶ (n,2n) | Ba ^{135m} | 28.7±0.2 hours | 700±80 | | |
| Ba ¹³⁸ (n,2n) | Ba ^{137m} | 2.6±0.1 min | 1250±100 | | |
| Ce ¹⁴⁰ (n,2n) | Ce ^{139m} | 65±10 seconds | 1200±400 | | |
| Ce ¹⁴² (n,2n) | Ce ¹⁴¹ | 32±2 days | 1600±300 | | |
| Pr ¹⁴¹ (n,2n) | Pr ¹⁴⁰ | 3.5±0.2 min | 2100±300 | 2060 | (a) |
| Nd ¹⁴² (n,2n) | Nd ¹⁴¹ | 2.5±0.3 hours | 2060±200 | 2480 | (b) |
| Nd ¹⁴⁸ (n,2n) | Nd ¹⁴⁷ | 11.5±0.5 days | 2160±200 | | |
| Nd ¹⁵⁰ (n,2n) | Nd ¹⁴⁹ | 1.8±0.1 hours | 2200±300 | | |
| Sm ¹⁴⁴ (n,2n) | Sm ¹⁴³ | 8.5±0.3 min | 1200±300 | | |
| Sm ¹⁵⁴ (n,2n) | Sm ¹⁵³ | 45±8 hours | 1500±300 | | |
| Eu ¹⁵¹ (n,2n) | Eu ¹⁵⁰ | 15±1 hours | 500±200 | | |
| Eu ¹⁵³ (n,2n) | Eu ^{152m} | 9.3±0.5 hours | 750±200 | | |
| Gd ¹⁶⁰ (n,2n) | Gd ¹⁵⁹ | 18.0±0.3 hours | 1450±300 | 1470 | (a) |
| Er ¹⁶⁶ (n,2n) | Er ¹⁶⁵ | 10±1 hours | 1000±400 | | |
| Er ¹⁷⁰ (n,2n) | Er ¹⁶⁹ | 9.8±0.5 days | 1200±500 | | |
| Yb ¹⁷⁶ (n,2n) | Yb ¹⁷⁵ | 4.2±0.2 days | 430±100 | | |
| Lu ¹⁷⁵ (n,2n) | Lu ¹⁷⁴ | 160±10 days | 1600±300 | | |
| La ¹³⁹ (n,γ) | La ¹⁴⁰ | 40±2 hours | 1.1±0.2 | 1.48 | (c) |
| Pr ¹⁴¹ (n,γ) | Pr ¹⁴² | 19±1 hours | 2.1±1.0 | 3.3 | (c) |
| Gd ¹⁶⁰ (n,γ) | Gd ¹⁶¹ | 3.7±0.5 min | 3.0±1.0 | 18.5 | (c) |
| Dy ¹⁶⁴ (n,γ) | Dy ^{165m} | 1.3±0.2 min | 1.5±0.5 | | |
| Dy ¹⁶⁴ (n,γ) | Dy ^{165g} | 140±5 min | 8±3 | | |
| Lu ¹⁷⁵ (n,γ) | Lu ^{176m} | 3.7±0.5 hours | 2±1 | | |
| Ba ¹³⁶ (n,p) | Cs ¹³⁶ | 13.5±0.5 days | 49±10 | 38.3 | (c) |
| Ba ¹³⁸ (n,p) | Cs ¹³⁸ | 32.5±0.5 min | 2.5±1.0 | 6.3, 2.3 | (a,c) |
| La ¹³⁹ (n,p) | Ba ¹³⁹ | 85±1 min | 5±1 | 2.3, 5.7 | (a,c) |
| Ce ¹⁴⁰ (n,p) | La ¹⁴⁰ | 40±2 hours | 10±2 | 12.1 | (c) |
| Ce ¹⁴² (n,p) | La ¹⁴² | 77±3 min | 5±2 | 9.4 | (c) |
| Pr ¹⁴¹ (n,p) | Ce ¹⁴¹ | 32±2 days | 4.5±1.0 | | |
| Nd ¹⁴⁸ (n,p) | Pr ¹⁴⁸ | 12±3 min | 3.5±0.8 | | |
| Sm ¹⁵² (n,p) | Pm ¹⁵² | 6.5±0.5 min | 3.7±0.2 | | |
| Sm ¹⁵⁴ (n,p) | Pm ¹⁵⁴ | 2.5±0.5 min | 3.5±0.2 | | |
| Dy ¹⁶³ (n,p) | Tb ¹⁶³ | 7±1 min | 3.0±1.0 | | |
| Er ¹⁶⁷ (n,p) | Ho ¹⁶⁷ | 3.1±0.1 hours | 3.0±1.0 | | |
| Er ¹⁶⁸ (n,p) | Ho ¹⁶⁸ | 3.3±0.5 min | 2.5±1.0 | | |
| Er ¹⁷⁰ (n,p) | Ho ¹⁷⁰ | 40±10 seconds | 1.8±0.5 | | |
| La ¹³⁹ (n,α) | Cs ¹³⁶ | 13.5±1.5 days | 1.3 | 1.87 | (c) |
| Ce ¹⁴⁰ (n,α) | Ba ^{137m} | 2.6±0.1 min | 9±2 | 12 | (a) |
| Ce ¹⁴² (n,α) | Ba ¹³⁹ | 85±1 min | 8±2 | 7.04 | (c) |
| Nd ¹⁴² (n,α) | Ce ^{139m} | 65±10 seconds | 10±2 | | |
| Nd ¹⁴² (n,α) | Ce ^{139g} | 140±10 days | 2±1 | | |
| Nd ¹⁴⁶ (n,α) | Ce ¹⁴³ | 34±2 hours | 8.3±2.0 | 2.6 | (c) |
| Nd ¹⁴⁸ (n,α) | Ce ¹⁴⁵ | 3.1±0.2 min | 5±1 | | |
| Sm ¹⁵² (n,α) | Nd ¹⁴⁹ | 1.8±0.1 hours | 10±2 | | |
| Sm ¹⁵⁴ (n,α) | Nd ¹⁵¹ | 17.3±5.0 min | 9±3 | | |
| Gd ¹⁶⁰ (n,α) | Sm ¹⁶⁷ | 0.5±0.1 min | 2±1 | | |
| Dy ¹⁶⁴ (n,α) | Gd ¹⁶¹ | 3.7±0.3 min | 4.5±0.8 | | |
| Er ¹⁶⁸ (n,α) | Dy ^{165m} | 1.3±0.2 min | 1.0±0.2 | | |
| Er ¹⁶⁸ (n,α) | Dy ^{165g} | 140±5 min | 0.5±0.2 | | |
| Er ¹⁷⁰ (n,α) | Dy ¹⁶⁷ | 4.4±0.4 min | 1.0±0.2 | | |

^a See Paul and Clarke, reference 16.^b L. A. Rayburn, Bull. Am. Phys. Soc. 4, 288 (1959).^c See reference 17.

enriched Nd¹⁵⁰. The 0.31-Mev gamma probably belongs to the decay of Ce¹⁴⁶.

Recently, Hoffman and Daniels¹⁴ observed a 12-min isotope of praseodymium formed as the daughter of a new 1.2-min cerium fission product to which they have assigned mass number 147, in disagreement with our suggested assignment. They also report a 300-keV gamma activity in fission-formed praseodymium which decays with a 1.95-min half-life and to which they have assigned mass number 148.

¹⁴ D. C. Hoffman and W. R. Daniels, Bull. Am. Phys. Soc. 4, 372 (1959).

0.5 min Sm¹⁵⁷

A new half-life of 0.5±0.1 min appeared in irradiations of 98% pure natural gadolinium metal with 14.8-Mev neutrons. Further studies with 95.4% enriched Gd¹⁶⁰ (as oxide) showed that the yield ratio of the new 0.5-min species to 3.7 min Gd¹⁶¹ remained constant in both natural and enriched samples, proving that the new period arises from Gd¹⁶⁰. By cross bombarding natural dysprosium, 74% enriched Dy¹⁶³, and 90% enriched Dy¹⁶⁴ (as oxides), the 0.5-min activity was not observed, either in gross beta decay or in gamma-ray decay, thus ruling out isomeric states of

TABLE III. Comparison of measured cross sections and theoretically calculated cross sections at 14.8 Mev.^a

| Reaction | Observed cross section (mb) | a (Mev ⁻¹) | θ | $E_n - S_n$ θ | Compound nucleus cross section Calc. (mb) | $\sigma_{\text{obs}}/\sigma_{\text{calc.}}$ |
|-------------------------------|--------------------------------|-----------------------------|----------|-------------------------|--|---|
| Ba ¹³⁴ ($n, 2n$) | 940±80 | 9.20 | 1.27 | 5.70 | 1890 | 0.50 |
| Ba ¹³⁶ ($n, 2n$) | 700±80 | 9.20 | 1.27 | 5.80 | 1900 | 0.37 |
| Ba ¹³⁸ ($n, 2n$) | 1250±100 | 9.20 | 1.27 | 5.04 | 1760 | 0.71 |
| Ce ¹⁴⁰ ($n, 2n$) | 3000±400 | 9.25 | 1.27 | 5.04 | 1960 | 1.5 |
| Ce ¹⁴² ($n, 2n$) | 1600±300 | 9.25 | 1.27 | 4.80 | 1900 | 0.85 |
| Pr ¹⁴¹ ($n, 2n$) | 2100±300 | 9.20 | 1.27 | 4.2 | 1850 | 1.14 |
| Nd ¹⁴² ($n, 2n$) | 2060±200 | 9.20 | 1.27 | 4.0 | 2060 | 1.0 |
| Nd ¹⁴⁸ ($n, 2n$) | 2160±200 | 9.40 | 1.25 | 4.0 | 2060 | 1.06 |
| Nd ¹⁵⁰ ($n, 2n$) | 2200±300 | 9.50 | 1.25 | 6.3 | 2080 | 1.06 |
| Sm ¹⁴⁴ ($n, 2n$) | 1200±300 | 9.00 | 1.28 | 6.3 | 2280 | 0.53 |
| Sm ¹⁵⁴ ($n, 2n$) | 1500±300 | 9.5 | 1.25 | 5.8 | 2330 | 0.64 |
| Eu ¹⁵¹ ($n, 2n$) | 500±200 | 9.5 | 1.25 | 4.4 | 2000 | 0.25 |
| Eu ¹⁵³ ($n, 2n$) | 750±200 | 9.5 | 1.25 | 4.9 | 2020 | 0.38 |
| Gd ¹⁶⁰ ($n, 2n$) | 1450±300 | 9.6 | 1.24 | 5.9 | 2140 | 0.68 |
| Er ¹⁶⁶ ($n, 2n$) | 1000±400 | 9.8 | 1.23 | 5.6 | 2100 | 0.48 |
| Er ¹⁷⁰ ($n, 2n$) | 1200±500 | 9.8 | 1.23 | 6.1 | 2100 | 0.57 |

| Reaction | Observed cross section (mb) | Calculated comp. nuc. | Cross section (mb) Dir. interact. | Ratio ($\sigma_{\text{obs}}/\sigma_{\text{calc.}}$) Comp. nuc. | Dir. interact. |
|-----------------------------------|--------------------------------|--------------------------|--------------------------------------|---|----------------|
| Ba ¹³⁶ (n, p) | 49±10 | | 9.3 | | 5.3 |
| Ba ¹³⁸ (n, p) | 2.5±1 | 0.24 | 3.0 | 10 | 0.83 |
| La ¹³⁹ (n, p) | 5±1 | 0.33 | 9.3 | 15 | 0.84 |
| Ce ¹⁴⁰ (n, p) | 10±2 | | 4.8 | | 2.1 |
| Ce ¹⁴² (n, p) | 5±2 | | 2.35 | | 2.2 |
| Sm ¹⁵² (n, p) | 3.7±0.2 | 0.13 | | 27 | |
| Sm ¹⁵² (n, p) | 3.5±0.2 | 0.047 | | 74 | |
| Sm ¹⁵² (n, α) | 10±2 | 0.101 | | 99 | |
| Sm ¹⁵⁴ (n, α) | 9±3 | 0.054 | | 165 | |

^a The theoretical calculations were made on the basis of the continuum model of the compound nucleus (see text, references 1 and 12 for details) and the direct interaction cross sections are based on the work of Brown and Muirhead (see discussion in text) for (n, p) reactions. For ($n, 2n$) reactions the calculations were made as described in reference 1 (see text). The following terms were used: a , an empirical constant in the level density formula which varies with mass (see reference 15 in text); θ is the nuclear temperature; E_n is the incident neutron energy and S_n is the neutron separation energy (see references 1, 12, and 15).

Gd¹⁵⁹, Gd¹⁶⁰, and Gd¹⁶¹ as possible assignments. Hence, the 0.5-min nuclide must be either Eu¹⁶⁰ from the (n, p) reaction or Sm¹⁵⁷ from the (n, α) reaction on Gd¹⁶⁰, which is known⁹ to have a level at 75 keV which should be populated in the beta decay of Eu¹⁶⁰. Since no 75-keV photons were detected with a 1×1½-inch NaI(Tl) scintillation spectrometer and manual scaler, we tentatively assign Sm¹⁵⁷ to the 0.5-min activity (although it is possible that the 75-keV transition is very highly converted and thus not observed in the scintillation counting experiment). This assignment requires further verification. A gamma at 0.57±0.01 MeV was observed to follow this decay.

7 min Tb¹⁶³

In bombardment of natural dysprosium metal, a 7±1 min unreported period was observed, whose yield increased 2-fold relative to 140 min Dy¹⁶⁵ when 74% enriched Dy¹⁶³ (as oxide) was irradiated. The isotopic content of all other dysprosium isotopes was down by factors ranging from 2 to 10. Moreover, the 7-min species was not observed from 90% enriched Dy¹⁶⁴ (as oxide). Cross bombardments of natural erbium, natural and enriched gadolinium, and enriched Dy¹⁶⁴ rule out the possibility that the 7-min nuclide is an isomer in

gadolinium. The remaining possibility is the Dy¹⁶³(n, p) reaction which assigns the new 7-min species to Tb¹⁶³. A gamma at 0.18±0.05 MeV was observed to follow this decay. No new activity with half-life between 30 sec and 2 hours was found for Tb¹⁶⁴ from the (n, p) reaction in irradiations of 90% enriched Dy¹⁶⁴.

3.3 min Ho¹⁶⁸

Irradiations of 98% pure natural erbium metal exhibited a new 3.3±0.5-min half-life which could not be immediately assigned. When 76.9% enriched Er¹⁶⁸ (as oxide) was bombarded, the yield of the 3.3-min activity was observed to increase with the enrichment of Er¹⁶⁸; in natural erbium, the ratio of the 3.3-min activity to 9.8 day Er¹⁶⁹ was about 50, whereas in the enriched Er¹⁶⁸ sample, the ratio favored the 3.3-min period nearly 250-fold, which establishes the fact that this new species arises from Er¹⁶⁸. No short-lived activity was found in a cross bombardment of enriched Yb¹⁷⁰ (although a 2.0±1.0-min period was observed from natural ytterbium which was shown to arise from a reaction on Yb¹⁷⁶). Assignment of the 3.3-min period to isomeric Er^{167m} is ruled out on the basis of irradiation of Er¹⁶⁶ without cadmium wrapping (which should increase the thermal (n, γ) reaction on Er¹⁶⁶ and enhance

the yield of Er^{167m} since this failed to increase the yield of the 3.3-min activity. Similarly, the $(n,2n)$ reaction is excluded. With 87% enriched Dy^{164} without cadmium wrapping, an activity of nearly the same half-life, 3.7 min, was formed but this exhibited a quite different gamma spectrum,¹² using a 3×3-inch NaI(Tl) scintillation spectrometer and 50-channel analyzer, showing that the 3.3-min and 3.7-min activities are not identical, the latter in fact being the previously known Gd^{161} . The gamma spectrum of the 3.3-min species, arising from the $\text{Er}^{168}(n,p)$ reaction, shows a prominent line at 0.85 ± 0.05 Mev which is not present in the 3.7-min gamma spectrum from the Dy^{164} bombardments. For these reasons, we assign Ho^{168} to the new 3.3-min activity.

40 sec Ho^{170} and 4.4 min Dy^{167}

From 87% enriched Er^{170} (as oxide) new half-lives of 40 ± 10 sec and 4.4 ± 0.4 min were measured and their yields observed to follow the Er^{170} enrichment (Er^{168} being reduced to only 9.0% in this sample). The cross section for the formation of 3.1 hour Ho^{167} by the $\text{Er}^{170}(n,\alpha)\text{Dy}^{167}(\beta^-)\text{Ho}^{167}$ mechanism was found to agree (within 20%) with the cross section to form 4.4 min Dy^{167} , but if the 40-sec period were assigned to Dy^{167} , then the cross sections would disagree by more than a factor of 2. Therefore, it seems likely that the 40-sec activity is Ho^{170} from the $\text{Er}^{170}(n,p)$ reaction and the 4.4-min period is Dy^{167} from the $\text{Er}^{170}(n,\alpha)$ reaction.

Other Unassigned Activities Observed

From "specpure" natural Lu_2O_3 , new activities of 2.0 ± 0.5 min and 7.5 ± 1.0 min were found, which also were observed with 95.9% enriched Yb^{172} (as oxide), but the yields of both appeared to be more than 10 times too low if Yb^{172} were the responsible isotope. Both periods also were observed again with 92.6% enriched Yb^{173} , but in yields again too small by a factor of 10 to have arisen from Yb^{173} . From 98.4% enriched Yb^{174} , the 2.0-min activity was not found, but a new 5.5 ± 0.5 min half-life was observed, probably due to Tm^{174} from the (n,p) reaction. When 97.5% enriched Yb^{176} was irradiated, however, the 2.0-min activity was found, but the 5.5-min and 7.5-min periods were absent, so that the 2.0-min species arises from an (n,p) , (n,α) , or (n,γ) reaction on Yb^{176} . The (n,p) and (n,γ) reactions, however, would have a cross section of about 90 mb, about 10 times larger than normal for this region (Table II). The (n,γ) reaction would give an unreported isomeric state in Yb^{177} . No further data is at hand with which to make definite assignments of either the 2.0 or the 7.5-min periods. In the bombarded samples of natural ytterbium, enriched Yb^{174} , and enriched Yb^{176} , gammas at 0.18 ± 0.01 , 0.25 ± 0.01 , and 0.36 ± 0.01 Mev were found to decay with the 2.0-min half-life.

An unidentified 9 ± 1 -min activity was found in natural erbium bombardments, but not from enriched

Er^{168} or Er^{170} . Nothing further is known about this species. An unknown 45 ± 10 -min half-life was observed in irradiations of 93% enriched Nd^{150} , but no further data is at hand concerning it.

DISCUSSION

Examination of the cross-section results in Table II indicates that there is no general trend with mass number at a given Z for the $(n,2n)$ reactions, which lie in the range of 100 mb for isomeric states to 3000 mb for total activation cross sections. The $(n,2n)$ reactions on Ce^{140} , Pr^{141} , Nd^{142} , and Sm^{144} exhibit a decrease with increasing mass number at $N=82$, e.g., 3000, 2100, 2060, and 1200 mb, respectively. The observed cross sections for $(n,2n)$ reactions agree within one order of magnitude with the compound nucleus continuum theory calculations,^{1,12} as can be seen in Table III. Values less than unity for the $(\sigma_{\text{obs}}/\sigma_{\text{calc}})$ ratio (Table III) based on the compound nucleus model¹⁵ have been cited as evidence that direct interaction is a competing process.¹⁶ For $\text{Ba}^{134,136,138}$ and Eu^{153} the low ratio can be attributed to the experimental difficulty of measuring the ground-state product.

In the rare earth region (n,p) cross sections usually amount only to 10 mb or less, except for $\text{Ba}^{136}(n,p)$ which has an anomalously high value (49 mb). The influence of the 82-neutron closed shell on 14.8 Mev (n,p) and (n,α) reactions in this region seems to be negligible, e.g., $\text{Ce}^{140}(N=82)$ has an (n,p) cross section of 10 mb while that of Ce^{142} is 5 mb; La^{139} and Pr^{141} ($N=82$ for both) have (n,p) cross sections of 5 and 4.5 mb, respectively; Nd^{148} , Sm^{152} , and Sm^{154} , all nonclosed shell nuclides, have (n,p) cross sections of 3.5, 3.7, and 3.5 mb, respectively. Similarly, the Levkovskii trends² appear to be negligible in this region. The same statements can be made for the (n,α) cross sections as well, e.g., this reaction on Sm^{152} and Sm^{154} exhibits a cross section of 10 and 9 mb, respectively, while with Er^{170} the value is 1.0 mb and 1.5 mb with Er^{168} . Coleman¹⁷ likewise found no apparent trend for (n,α) cross sections in this region at 14 Mev, e.g., the values for Cs^{133} , La^{139} , Ce^{142} , Gd^{156} , Dy^{162} , and Hf^{178} are 1.9, 1.87, 7.04, 3.22, 3.56, and 2.0 mb, respectively.

The deviation between observed cross sections and those calculated on the continuum theory of the compound nucleus¹⁵ for (n,p) and (n,α) reactions (Table III) is larger than unity by factors of 10 to 100, e.g., (n,p) cross sections for Ba^{138} and La^{139} are 10 and 15 times larger than calculated, respectively; those for

¹⁵ J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley & Sons, Inc., New York, 1952).

¹⁶ E. P. Paul and R. L. Clarke, *Can. J. Phys.* **31**, 276 (1953); G. Brown and H. Muirhead, *Phil. Mag.* **2**, 473 (1957); K. Winter, B. Torki, and E. Remy, *Nuovo cimento* **11**, 1 (1959); and E. Remy and K. Winter, *Nuovo cimento* **10**, 664 (1958).

¹⁷ R. F. Coleman, B. E. Hawker, L. P. O'Connor, and J. L. Perkin, *Proc. Phys. Soc. (London)* **73**, 215 (1959); Atomic Energy Research Establishment Report AERE-O-59/57, 1958 (unpublished).

Sm^{152} and Sm^{154} are 27 and 74 times larger,¹ respectively, and the (n,α) cross sections on these same nuclides exhibit the still larger deviations of 99 and 165, respectively. For (n,p) reactions Brown and Muirhead¹⁸ have calculated cross sections from a direct interaction model which assumes that these interactions, in which no intermediate nucleus is formed, occur by collision with nucleons throughout the whole nuclear volume with equal probability for all nucleons. In Table III we have listed for certain (n,p) reactions a comparison with these direct interaction calculations.

Although this direct interaction model gives better agreement with experiment (Table III),¹⁷ it cannot be developed to include the (n,α) process, nor does it take into account that surface reactions are considerably more probable, especially in the heavier mass regions, than are direct interactions with nucleons deep in the nucleus.¹⁹ Wilkinson²⁰ suggests that nucleon clusters,

e.g., alpha-particles and deuterons) tend to exist in the region of low nuclear binding in the diffuse nuclear surface. Such a tendency for preformed alpha-particle clusters to exist in the surface suggests itself as a possible explanation for the large (n,α) cross sections in the heavy mass region. Such a model also might be a possible explanation for the near integral multiple decrease in (n,α) cross sections in the low Z region at 14 Mev pointed out by Levkovskii.² Since the number of such clusters would be much lower in the low Z region, any change in the number of clusters available to the incoming projectile (as for example with increasing A at constant Z) would be much more marked in the cross sections than would be the case at high Z .

ACKNOWLEDGMENTS

We are indebted to Mr. J. Wray for operation of the accelerator during these experiments.

and D. H. Wilkinson, *Phil. Mag.* **3**, 1193 (1958); and P. B. Jones, *Phil. Mag.* **3**, 33 (1958).

²⁰ D. H. Wilkinson, *Phil. Mag.* **4**, 215 (1959).

¹⁸ G. Brown and H. Muirhead, *Phil. Mag.* **2**, 473 (1957).

¹⁹ G. B. Chadwick, S. A. Duranni, P. B. Jones, J. W. G. Wignall,

Decay of Be^{9*} (2.43-Mev State)*†

E. M. HENLEY‡ AND P. D. KUNZ

Department of Physics, University of Washington, Seattle, Washington

(Received July 13, 1959)

The decay of the 2.43-Mev state of Be^9 is treated theoretically. Of the open two-body decay channels all but one involve a nuclear state, the energy of which is not well defined. The usual formalisms have been generalized to take this into account. The estimate of the decay rates is made by means of a variational internal wave function for the Be^{9*} state, based upon the alpha-particle model. It is found that the principal mode of decay is to $\text{He}^3 + \text{He}^4$. Model-dependent arguments are given to show that decay to the ground state of Be^8 should be inhibited. Furthermore, the momentum and angular distributions of alphas emitted in the decay through several two-particle decay modes are computed. These latter calculations do not assume any specific nuclear model, but depend on the weak assumption that the state is excited by a direct reaction. Comparison with recent measurements indicates that in addition to the $\text{He}^3 + \text{He}^4$ decay, approximately 7% of the decay occurs to the ground state of Be^8 , which is consistent with our calculations.

I. INTRODUCTION

THE decay of Be^{9*} excited by various reaction mechanisms has been the source of several studies in the past. For example, early investigations¹ of the decay from the 2.43-Mev excited level have indicated that it proceeds mainly by emission of a neutron to the ground state of Be^8 . However, recent coincidence measurements by Bodansky, Eccles, and

Halpern² (hereafter referred to as BEH) have set an upper limit of 10% on decays through this channel and have concluded that emission occurs primarily by means of other processes. We shall analyze the decay of the 2.43-Mev state theoretically and compare predictions with experiment. Since this decay occurs in part through intermediate states (e.g., He^6), the energy of which is not well defined, we have had to generalize the usual emission width relations to take this into account explicitly.³ This has been done along the lines suggested by Watson.⁴

* Supported in part by the U. S. Atomic Energy Commission.

† Part of this work was done while the authors were guest scientists at Brookhaven National Laboratory during the summer of 1957.

‡ Presently on leave at the Institute for Theoretical Physics, Copenhagen, Denmark.

¹ G. A. Dissanaik and J. O. Newton, *Proc. Phys. Soc. (London)* **A65**, 675 (1952). G. M. Frye and J. H. Gammel, *Phys. Rev.* **103**, 328 (1956).

² D. Bodansky, S. F. Eccles, and I. Halpern, *Phys. Rev.* **108**, 1019 (1957).

³ A. C. Riviere, *Nuclear Phys.* **2**, 81 (1956/57); T. A. Welton, *Phys. Rev.* **95**, 302 (1954).

⁴ K. M. Watson, *Phys. Rev.* **88**, 1136 (1952).