PHYSICAL REVIEW C VOLUME 7, NUMBER 6 JUNE 1973

Measurement of α-Decay Branching Ratios for ^{150,151}Dy and ^{149}Tb^{m} [†]

C. R. Bingham and D. U. O'Kain*
The University of Tennessee, Knoxville, Tennessee 37916, and Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830

and

K. S. Toth and R. L. Hahn
Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830

(Received 16 March 1973)

With the use of the helium-gas-jet technique and a high-resolution Ge(Li) detector α-decay branching ratios were measured for ^{150}Dy, ^{151}Dy, and ^{149}Tb^{m}. Sources containing these activities were produced by bombarding ^{141}Pr with ^{14}N and ^{12}C ions. They were then assayed for both α-particle and x-ray radioactivity. The branching ratios deduced are as follows: 0.32 ± 0.05 for ^{150}Dy, 0.055 ± 0.008 for ^{151}Dy, and $(2.0 \pm 0.4) \times 10^{-4}$ for ^{149}Tb^{m}. The ^{151}Dy and ^{149}Tb^{m} ratios were found to be in good agreement with previous measurements. While the ratio deduced for ^{150}Dy is almost midway in value between two earlier determinations, it appears to be in real disagreement with the more precise of the two previous measurements. Consequences of this discrepancy are discussed with respect to published cross-section data and α-decay reduced widths for 84-neutron even-even nuclei. In the course of the investigation the half-life of ^{149}Dy was determined to be 4.6 ± 0.4 min from x-ray decay-curve analyses and 5.1 ± 0.9 min from the initial growth period observed in the decay of the 4.1-h, ^{149}Tb 3.967-MeV α group.

I. INTRODUCTION

Experimental values of α-decay branching ratios are useful not only for comparison with α-decay theories, but also because they can often be used to deduce cross sections for complicated reactions in which products are identified by their α decay. Indeed, since the advent of high-energy heavy-ion accelerators, the observation of characteristic α-particle groups has been widely used in the identification of new neutron-deficient isotopes. For many of them, however, the α-decay branching ratios are either lacking or inaccurately determined primarily because of their short half-lives. If these ratios were known then in many instances relative yields could be converted to reaction cross sections. With the recent interest in the investigation of nuclei far from stability the knowledge of these heavy-ion-induced reaction cross sections has taken on a great deal of importance.

In the present work we report on the possibility of measuring α-decay branching ratios by combining the gas-jet technique[1] with the use of high-resolution Ge(Li) x-ray detectors. This combination does away with the necessity of chemical separations for the preparation of thin sources suitable for a α-particle counting and for the elimination of elemental fractions other than the one of interest. The technique could then be extended to isotopes with half-lives down to a few seconds if a capillary is used to transport the gas jet to a shielded position so that α-particle, x-ray, and γ-ray counting can be made simultaneously. Specifically, with this experimental method branching ratios were measured for ^{149}Tb^{m}, ^{150}Dy, and ^{151}Dy and compared with earlier determinations[2-4] for the same three nuclides.

II. EXPERIMENTAL METHOD

The dysprosium and terbium nuclides investigated were produced by bombarding a ^{141}Pr target with ^{14}N and ^{12}C ions accelerated in the Oak Ridge isochronous cyclotron. The ^{141}Pr target consisted of a 300-μg/cm^2 layer of praseodymium oxide deposited onto a 25-μm beryllium backing foil which served as an entrance window to a small chamber filled with helium to a pressure of ~$\frac{1}{2}$ atm. Product recoils ejected from the thin target were stopped in the helium gas and then swept out together with the gas through an orifice by means of a high-speed pump. The recoils were collected on an aluminum foil placed in front of the orifice. After bombardment the foil was automatically moved by means of a rotating wheel in front of a Si(Au) surface-barrier detector for assay of α-particle activity. After a suitable period of counting, the foil was removed from the chamber and assayed for x-ray activity with a Ge(Li) detector. [Recent experiments on gas-flow techniques[5] indicate that essentially no activity ($\leq 1\%$) is lost in such physical transfers.] While the source was being counted with the x-ray detector a new source was made under similar bombardment conditions to obtain an independent measure of the half-lives of the nuclei in question by α-particle counting.

In the case of the ^{14}N irradiations different bombarding energies were obtained by using beryllium-metal foils to degrade the incident energy (119 MeV) of the beam deflected out of the cyclotron. Bombardments were made at ^{14}N energies of 81.7, 87.5, and 109.4 MeV. These energies and respective bombardment times of 1 h, 800 and 200 sec, were chosen to enhance activities of ^{151}Dy and ^{152}Dy, ^{150}Dy and ^{151}Dy, and ^{149}Dy and ^{150}Dy. The ^{12}C bombardments were made at 82.2 MeV to emphasize the production of ^{149}Tb^{m}.

Since different isotopes of a given element have K x rays of the same energy, it was necessary to resolve the decay curves into the various half-life components present. These analyses were made by means of a multicomponent decay computer program.[6] Half-lives for ^{149}Tb^{m}, ^{150}Dy, ^{151}Dy, and ^{152}Dy were taken from the decay curves measured for their characteristic α-particle groups. (See the compilation by Eskola[7] for the α-decay energies of these four nuclides.) The 83-neutron nuclide ^{149}Dy does not exhibit α decay. For this reason, in the decay-curve analysis of the terbium K-x-ray peaks its half-life and the intensity of all components present were allowed to be varied by the program itself in order to produce a least-squares fit.

Electron-capture decay rates were obtained from the number of $K\alpha_1$ x rays emitted. By using the formulas of Martin and Blichert-Toft[8] and data from the same work and from Refs. 9 and 10, corrections were made for other K x rays emitted, for capture from higher shells and for Auger conversion of K-shell vacancies. Some K x rays may result from internal-conversion processes in the final nucleus, but since the decay schemes of ^{149}Tb^{m}, ^{150}Dy, and ^{151}Dy are not known, no estimate could be made for this factor. The α-decay branching ratio was then $R_\alpha/(R_\alpha+R_{ec}+R_{\beta+})$, where the R's represent absolute disintegration rates for α, electron capture, and positron decay. The positron decay rate, $R_{\beta+}$, was deduced from the electron-capture rate, R_{ec}, by using formulas from Ref. 8 and electron-capture decay energies from Ref. 10. The statistical rate function values f were taken from Ref. 11. In making these corrections the transitions were assumed to be allowed.

III. RESULTS AND DISCUSSIONS

Half-lives measured for ^{149}Tb^{m}, ^{149}Dy, ^{150}Dy, and ^{151}Dy are shown in Table I and compared with published data.[2, 12, 13] The agreement with previous results is satisfactory except perhaps in the case of ^{149}Dy. Since ^{149}Dy does not have an α branch the value reported here was obtained in two ways: (1) from the x-ray decay-curve analyses, and (2) from the initial growth period observed in the decay of the 4.1-h 3.967-MeV α group due to the decay of ^{149}Tb. Analyses of the decay curve for ^{149}Tb produced in a number of bombardments at varying incident ^{14}N energies yielded a value of $T_{1/2}=5.1\pm0.9$ min which is consistent with that of 4.6 ± 0.4 min obtained from the x-ray decay-curve analyses. The 8 ± 2-min value[12] was also determined by observing an initial growth period. In that instance, however, the sample counted was thick so that instead of discrete α-particle groups one broad unresolved peak was seen. The growth period was found in the portion of the spectrum which corresponded in energy to that of the 4.1-h ^{149}Tb. It was on that basis that the 8-min activity was assigned[12] to ^{149}Dy.

At that time the existence of the 4.2-min ^{149}Tb^{m} was not known. Subsequently, when Macfarlane[14] reported its discovery he was able to show by means of a "recoil-milking" experiment that it, and not the 4.1-h α emitter, represented the isomeric state. It is, thus, worth considering what effect its presence has on the half-life of ^{149}Dy as determined from the initial growth period in the decay curve of ^{149}Tb. The ground and isomeric states in this terbium nucleus are almost certainly due to the odd 65th proton being in either the $d_{5/2}$ or $h_{11/2}$ orbital; the two orbitals should be close to one another in excitation energy, as the $d_{5/2}$ subshell is filled at $Z=64$. The parent activity, ^{149}Dy, would be expected to have a ground state represented by the 83rd $f_{7/2}$ neutron. The neighboring 83-neutron nucleus, ^{147}Gd, also with a

TABLE I. Half-lives of isotopes studied compared with published values.

Nucleus	Present work (min)	Other (min)	Reference
^{149}Dy	4.6 ± 0.4 [a] 5.1 ± 0.9 [b]	8 ± 2 [b]	12
^{150}Dy	7.17 ± 0.02	7.20 ± 0.10	2
^{151}Dy	16.9 ± 0.5	18.0 ± 0.2 17.7 ± 0.5	2 13
^{149}Tb^{m}	4.16 ± 0.04	4.3 ± 0.2	2

[a] Half-life determined from the decay-curve analyses of $K\alpha_1$ x-ray peaks.

[b] Half-lives determined from the initial growth period observed in the decay of the 4.1-h 3.967-MeV α group of ^{149}Tb.

proposed $f_{7/2}$ ground state, is known[15] to decay primarily to two $\frac{5}{2}^-$ states in ^{147}Eu. If ^{149}Dy should do the same then there is no reason why both the $h_{11/2}$ and $d_{5/2}$ states in ^{149}Tb should not get populated in subsequent γ-ray transitions. The intensity, however, of the isomeric transition seems to be extremely weak *vis-à-vis* the electron-capture and β^+ modes of decay because in our ^{141}Pr + ^{12}C bombardments a noticeable initial growth period was not observed for the ^{149}Tb α-particle group. It would thus appear that the ^{149}Dy half-life deduced from the growing-in portion of the ^{149}Tb decay curve is close to its true half-life. Parenthetically one might add that the ^{149}Tb^m α-group was not observed in our ^{14}N + ^{141}Pr bombardments presumably because of the isomer's extremely low α-decay branching ratio and the presence of the strong α peaks due to ^{149}Tb, ^{150}Dy, and ^{151}Dy.

The α-decay branching ratios deduced for ^{150}Dy, ^{151}Dy, and ^{149}Tb^m are shown in Table II and compared with published results.[2-4] It is seen that our ratios for ^{149}Tb^m and ^{151}Dy are in agreement with the previously determined values. In the case of ^{150}Dy our value of 0.32 is in between those reported in Ref. 2 (0.18) and Ref. 4 (0.75). This latter number has an extremely large error limit set on it; since within these errors it agrees with both our value and that of Ref. 2, we will drop that particular ratio from any further discussion. On the other hand the disagreement between our ratio and the one reported by Macfarlane and Seegmiller seems to be real even if the error limits in both investigations are taken into account. Possible sources of error in the present measurement were carefully considered. First, a correction for $K\alpha$ x rays produced by internal-conversion processes would result in a higher α-decay branching ratio and therefore increase the disagreement. Second, the value, of course, would be lower if the contribution of ^{149}Dy to the x-ray activity were neglected. We feel, however, that the contribution was properly accounted for since the 29% variation in beam energy resulted in large differences in the ratio of the ^{149}Dy and ^{150}Dy activities, but not in the branching ratio for ^{150}Dy. Third, the component of activity due to positron decay was calculated for allowed transitions and ground-state decay energies. If these transitions are not allowed, the positron components are then overestimated and once again the true α-decay branching ratios would be higher than those presently reported. This increase, however, would have essentially no effect on the ^{150}Dy ratio because the amount of positron decay is estimated to be only about 1% of the electron-capture branch based on a value of 1.96 MeV for the ^{150}Dy electron-capture decay energy.[10]

Some support for the higher branching ratio comes from reported[16] cross sections for the formation of ^{150}Dy and ^{151}Dy in (heavy ion, xn) reactions; those for ^{150}Dy are consistently larger than the ones for ^{151}Dy. The cross sections were obtained by detecting the α decay of ^{150}Dy and ^{151}Dy and then correcting the α-disintegration rate by applying the branching ratios of Ref. 2. Figure 1 shows the maximum cross sections obtained for the two isotopes, produced in the interaction of various combinations of even-even targets and projectiles, plotted as a function of the number of neutrons emitted from the compound system. Part (a) shows the peak cross sections as reported in Ref. 16, while part (b) shows the same results if the present branching ratios are used. It is clear that in Fig. 1(b) not only are the two sets of cross sections similar in value but they now show a more systematic variation with the number of emitted neutrons. We would also like to consider two other excitation functions reported by Alexander and Simonoff,[16] i.e., for the reactions ^{141}Pr(^{14}N, 4n)^{151}Dy and ^{141}Pr(^{14}N, 5n)^{150}Dy. Here again the cross section for the formation of ^{150}Dy is about a factor of 2 greater than the one for ^{151}Dy, whereas if the present α-decay branching ratios are utilized the two sets of cross sections become essentially equal. This is shown in Fig. 2 where in addition to the data points we include the same two excitation functions as recently calculated by Zganjar[17] using the computer code developed by Blann and collaborators (see, e.g., Ref. 18). It can be seen that the calculation: (1) predicts correctly the bombarding energies at which the two cross sections peak, although the widths of both calculated excitation functions are

TABLE II. Experimental α-decay branching ratios.

Nucleus	Present data	Other results Ref. 2	Ref. 3	Ref. 4
^{150}Dy	0.32 ± 0.05	0.18 ± 0.02		0.75 ± 0.60
^{151}Dy	0.055 ± 0.008	0.059 ± 0.006	0.055 ± 0.010	0.07 ± 0.05
^{149}Tb^m	$(2.0 \pm 0.4) \times 10^{-4}$	$(2.5 \pm 0.5) \times 10^{-4}$	$<3 \times 10^{-4}$	$<5 \times 10^{-4}$

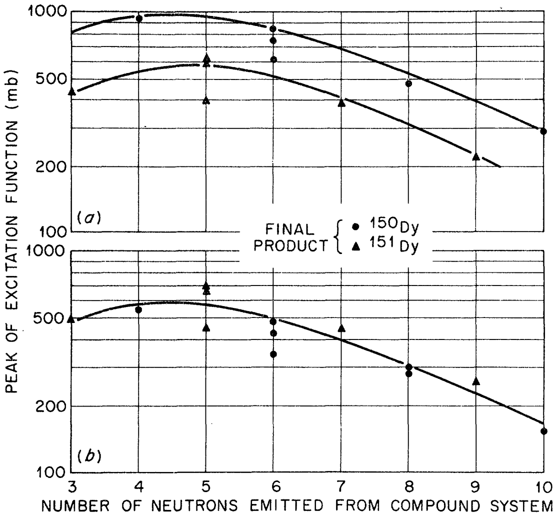

FIG. 1. Maximum cross sections for the formation of ^{150}Dy and ^{151}Dy in various (heavy ion, *xn*) reactions induced by even-even projectiles on even-even targets. Part (a) shows the data as reported in Ref. 16; part (b) shows cross sections deduced when the presently determined branching ratios are used to obtain total disintegration rates.

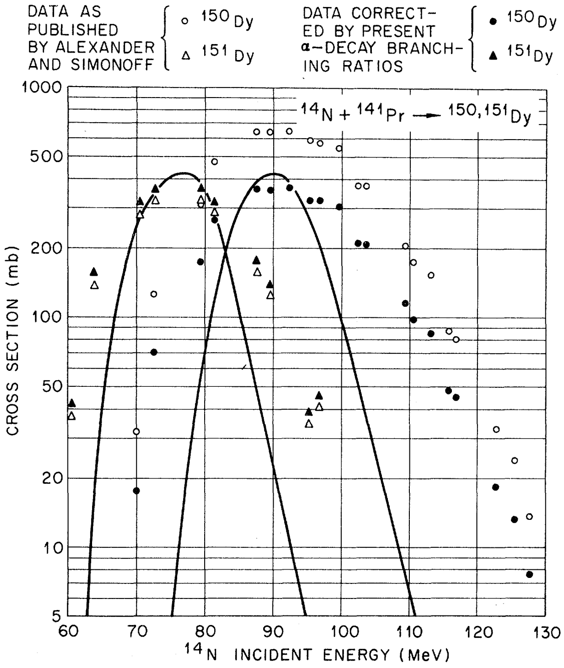

FIG. 2. Excitation functions for the reactions ^{141}Pr-(^{14}N,4*n*)^{151}Dy and ^{141}Pr(^{14}N,5*n*)^{150}Dy. Open points indicate data as reported in Ref. 16; closed points show the same data when these are corrected by the α-decay branching ratios determined in the present investigation. Curves represent excitation functions calculated by using a computer code based on the theory of Ref. 18.

much less than those indicated by the data points, and (2) is in better agreement with the data when these are corrected with the presently determined branching ratios.

From the information summarized in Tables I and II partial α-decay half-lives can be determined. These can then be considered within the framework of some α-decay-rate parametrization so as to obtain relative decay probabilities after the energy dependence has been removed. One such formalism, developed by Rasmussen,[19] defines the α-decay reduced width, δ^2, as

$$\lambda = \delta^2 P/h\,, \tag{1}$$

where λ is the decay constant, P is the barrier-penetrability factor, and h is Planck's constant. In this particular formalism P is calculated from an optical-model potential derived from α-particle scattering.

Macfarlane and Seegmiller[2] determined δ^2 for the three α emitters considered and we would like to discuss these reduced widths further, particularly with respect to the one involving ^{150}Dy. With the 0.18 branching ratio Macfarlane and Seegmiller calculated a δ^2 of 0.052 for ^{150}Dy, a value that is substantially less than those of other even-even nuclei in the rare-earth region. In particular, other 84-neutron even-even α emitters have reduced widths as follows: ^{146}Sm, 0.082; ^{148}Gd, 0.097; ^{152}Er, 0.091; and ^{154}Yb, 0.091. The lower probability for ^{150}Dy was thought to be due to the proton subshell at $Z = 64$ and Macfarlane, Rasmussen, and Rho[20] examined this possibility by using a Gaussian residual force in a BCS treatment for the proton system of 82-neutron nuclei. By varying the residual-force strength they were able to obtain a dip in the reduced width for $Z_{\text{parent}} = 64$; no reasonable adjustment of parameters, however, could reproduce the experimental dip at $Z_{\text{parent}} = 66$. From Eq. (1) it is clear that δ^2 is directly proportional to λ and therefore inversely proportional to the α-decay half-life. Because P had been calculated [2,20] with the correct α-decay energy our branching ratio increases δ^2 for ^{150}Dy to ~0.092. This value is in line with the reduced widths for the remainder of the even-even $N = 84$ nuclides and opens the question as to whether a decrease in α-decay probability does exist as a result of the proton subshell at $Z = 64$.

The ^{151}Dy δ^2 was calculated[2] to be 0.071. Since this value is close to those for even-even nuclei the indication is that the α decay of this odd-*A* nucleus is not hindered. This is probably because the ground-state spin of both the parent and daughter nuclei is represented by the odd $f_{7/2}$ neutron. The spin of ^{151}Dy has been measured [21] to be $\frac{7}{2}$, while ^{147}Gd decay data[15] are consistent with its ground-state spin assignment being $\frac{7}{2}^-$.

The ^{149}Tb^m δ^2 was determined[2] to be 0.0036 indicating that its α decay is considerably hindered. The small reduced width was calculated even assuming that the decay involved an $l=3$ α wave, proceeding from an $h_{11/2}$ state to a $d_{5/2}$ ^{145}Eu ground state. At that time the level structure of ^{145}Eu was not known. Since then ^{144}Sm(^{3}He, d) studies[22] have established the low-lying levels; in particular the first three states in ^{145}Eu are as follows: 0 keV ($d_{5/2}$), 330 keV ($g_{7/2}$), and 716 keV ($h_{11/2}$). Despite the favorable spins of the two higher states the α decay proceeds almost certainly to the ground state as assumed by Macfarlane and Seegmiller. This statement is based on systematics[23] of lifetimes for $E3$ transitions which indicate that these are in general hindered by factors of 100–1000 from the single-particle estimate. If the difference of 40 keV between the two ^{149}Tb^m α groups is taken to be the energy of the $E3$ isomeric transition then its single-particle half-life is ~1 sec, i.e., ~250 times faster then the 4.2-min half-life of ^{149}Tb^m. If the α decay proceeded to the 330-keV state in ^{145}Eu then the isomeric transition would be 370 keV in energy with a single-particle half-life in the vicinity of 10^{-4} sec. Using even the largest hindrance factors for $E3$ transitions, the isomeric state would still have a half-life of about 0.1 sec, i.e., much less than the experimental half-life of ^{149}Tb^m.

We would conclude by saying that despite the disagreement in the case of ^{150}Dy we feel that reasonable ratios can be obtained by the use of the experimental technique described in this paper. Its applicability to shorter-lived α emitters by using a capillary transport system would seem to be quite straightforward.

ACKNOWLEDGMENT

Thanks are due Wolf-Dieter Schmidt-Ott for helpful discussions and for his aid in the analysis of some of the experimental data obtained in this investigation.

---

† Research supported by the U. S. Atomic Energy Commission under contract with Union Carbide Corporation.

*Employee of Oak Ridge Gaseous Diffusion Plant, Oak Ridge, Tennessee. Results reported here were submitted in partial fulfillment of the requirement for a M.S. degree.

[1] R. D. Macfarlane and R. D. Griffioen, Nucl. Instrum. Methods 24, 461 (1963).

[2] R. D. Macfarlane and D. W. Seegmiller, Nucl. Phys. 53, 449 (1964).

[3] N. A. Golovkov, K. Ya. Gromov, N. A. Lebedev, B. Makhmudov, A. S. Rudnev, and V. G. Chumin, Izv. Akad. Nauk SSSR Ser. Fiz. 31, 1618 (1967) [transl.: Bull. Acad. Sci. USSR Phys. Ser. 31, 1657(1968)].

[4] V. G. Chumin, Dubna Report No. 6-3676, 1968 (unpublished).

[5] W.-D. Schmidt-Ott, R. L. Mlekodaj, and C. R. Bingham, Nucl. Instrum. Methods (to be published).

[6] J. B. Cumming, in *Proceedings of the Symposium on Applications of Computers to Nuclear and Radiochemistry, Gatlinburg, 1962,* edited by G. D. O'Kelly (Office of Technical Services, Department of Commerce, Washington, D. C., 1963).

[7] P. Eskola, Ark. Fys. 36, 477 (1966).

[8] M. J. Martin and P. H. Blichert-Toft, Nucl. Data A8, 156 (1970).

[9] A. H. Wapstra, G. J. Nijgh, and R. Van Lieshout, *Nuclear Spectroscopy Tables* (North-Holland, Amsterdam, 1959).

[10] A. H. Wapstra and N. B. Gove, Nucl. Data A9, 276 (1971).

[11] N. B. Gove and M. J. Martin, Nucl. Data A10, 205 (1971).

[12] K. S. Toth and J. O. Rasmussen, Phys. Rev. 109, 121 (1958).

[13] I. Mahunka and T. Fenyes, Izv. Akad. Nauk SSSR Ser. Fiz. 29, 1121 (1965) [transl.: Bull. Acad. Sci. USSR Phys. Ser. 29, 1126(1966)].

[14] R. D. Macfarlane, Phys. Rev. 126, 274 (1962).

[15] M. P. Avotina and A. V. Zolotavin, *Isobaric Nuclei with Mass Number $A=147$,* issue No. 11 of Properties of Atomic Nuclei (Publishing House "Science," Leningrad Section, Leningrad, 1971).

[16] J. M. Alexander and G. N. Simonoff, Phys. Rev. 133, B93 (1964).

[17] E. F. Zganjar, Louisiana State University at Baton Rouge, private communication.

[18] W. G. Winn, H. H. Gutbrod, and M. Blann, Nucl. Phys. A188, 423 (1972).

[19] J. O. Rasmussen, Phys. Rev. 113, 1593 (1959).

[20] R. D. Macfarlane, J. O. Rasmussem, and M. Rho, Phys. Rev. 134, B1196 (1964).

[21] A. Rosen, C. Ekstroem, H. Nyquist, and K. E. Aedelroth, Nucl. Phys. A154, 283 (1970).

[22] E. Newman, K. S. Toth, R. L. Auble, R. M. Gaedke, M. F. Roche, and B. H. Wildenthal, Phys. Rev. C 1, 1118 (1970).

[23] M. Goldhaber and A. W. Sunyar, in *Alpha-, Beta-, and Gamma-Ray Spectroscopy,* edited by K. Siegbahn (North-Holland, Amsterdam, 1965), p. 931.