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ALPHA DECAY PROPERTIES OF SOME TERBIUM AND DYSPROSIUM ISOTOPES NEAR THE 82-NEUTRON CLOSED SHELL

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Abstract: The half-lives and alpha particle energies of Tb^{149m}, Tb^{151}, Dy^{150}, Dy^{151}, Dy^{152}, and Dy^{153} have been measured with greater precision than previously reported. Alpha branching ratios for these nucleides were also obtained using direct counting techniques.

1. Introduction

Previous work by Rasmussen and co-workers on the alpha decay properties of terbium and dysprosium isotopes lying near the 82-neutron closed shell provided estimates of the alpha branching ratios of Tb^{151}, Dy^{152}, and Dy^{153} based on excitation function data [1]. No information was obtained by them on the alpha branching ratios of the 84- and 85-neutron dysprosium alpha emitters, Dy^{150} and Dy^{151}.

As more alpha decay data have become available in this region, the need for accurate alpha partial half-lives and alpha particle energies of these nucleides has become necessary in order to study the systematics in greater detail and provide date of sufficient accuracy to warrant theoretical analysis. The purpose of this paper is to report on some new measurements of the alpha branching ratios, alpha particle energies, and half-lives of the nucleides mentioned above as well as the recently observed [2] short-lived isomer of Tb^{149}.

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2. Experimental Details

2.1. Mode of Production of Nucleides

The high spin isomer of Tb^{149} was produced by bombarding a natural lanthanum oxide target ($\sim$2 mg/cm^2) with 121 MeV O^{16} ions using the Berkeley heavy ion accelerator. This energy is near the peak of the excitation function for the production of this activity by an O^{16}, 6n reaction [2]. The Tb^{151} activity was produced by bombarding a europium oxide sample enriched in Eu^{151} to 98% with 48 MeV α-particles using the Berkeley sixty-inch cyclotron. This bombarding energy gives a favourable cross-section for an (α,4n) reaction. The nucleides Dy^{150} and Dy^{151} were produced together by bombarding an enriched Ce^{140} oxide target with 105 to 120 MeV O^{16} ions. These nucleides are the products of (O^{16},6n) and (O^{16},5n) reactions. The nucleides Dy^{152} and Dy^{153} were produced by (α,4n) and (α,3n) reactions on an enriched Gd^{152} target at bombarding energies of 48 MeV and 37 MeV respectively.

2.2. Sample Preparation

Samples of the particular nucleide to be studied were prepared in carrier-free form suitable for alpha-particle counting with minimum self-absorption. The nucleides produced at the sixty-inch cyclotron were separated from the rare earth target material using cation exchange chromatography and α-hydroxyisobutyric acid as the eluant. This procedure also separated the individual rare earth elements from each other [3].

The shorter-lived nucleides, Dy^{150}, Dy^{151}, and Tb^{149m}, which were produced by heavy ion reactions, were separated from the target material by collecting recoils ejected from the target on to a palladium leaf catcher during the bombardment. After bombardment, the catcher was dissolved in nitric acid containing chloride ion and passed through an anion-exchange column to remove the palladium. The rare earth fraction was then co-precipitated as the oxalate with added strontium carrier. The oxalate was redissolved and

the oxalate ion destroyed with nitric acid. The rare earths were re-precipitated as the hydroxide on added Fe^{+3} carrier and the Fe^{+3} was finally removed by dissolving the precipitate in hydrochloric acid and passing through an anion-exchange column. The eluant contained the pure rare earth fraction in carrier-free form.

Alpha sources were prepared by solution-evaporation (usually ~50λ) on a 0.05 mm-thick platinum plate. Gamma sources were prepared by placing a small quantity of the solution (~10λ) in the bottom of a thin polyethylene test tube which fitted into the well-type scintillation crystal used for measurement of the combined E.C. + β^+ branching ratio.

2.3. Alpha Particle Energy and Half-Life Determinations

Alphas particle energies were measured using a Frisch-grid ionization chamber connected to standard electronics and a 100-channel pulse-height analyzer. The alpha particles from Tb^{149} (3.95 MeV[1]) and U^{234} (4.75 MeV (average))[4] were used as energy standards.

Half-lives were obtained from alpha decay curves which were in most cases single component. Particular care was taken to obtain very accurate half-lives for Dy^{150} and Dy^{151} because of the usefulness of these nucleides for nuclear reaction studies. Single component alpha decay curves of these nucleides were obtained by measuring the decay of the Dy^{150} and Dy^{151} groupes in the alpha particle spectra of a sample containing both activities. The data obtained were then analyzed by the method of least squares with each point weighted according to the statistical uncertainty of the measurement.

2.4. Alpha Branching Ratios

The alpha branching ratio, which is defined in this paper as the ratio of the alpha disintegration rate to the total disintegration rate, was obtained by measuring a simultaneous alpha disintegration rate and combined β^+ + E.C. disintegration rate.

The alpha counter used in the measurements was a standard 2π geometry flow counter whose counting efficiency was periodically checked using alpha standards. The overall detection efficiency was taken as 52%. The alpha counting rate of a measured aliquot of a sample to be measured was obtained at a particular time - zero from an alpha decay curve. These decay curves were in each case single component with a negligible background contribution.

The corresponding β^++ E.C. disintegration rate was obtained indirectly by measuring the absolute photodisintegration rate. The nucleides studied, because of their position on the neutron - deficient side of the beta stability line, decay mainly by K-electron capture and positon emission. A 4π gamma detection system consisting of two shielded back-to-back NaI (Tl) scintillation crystals connected to photomultiplier tubes and standard slow electronics was used to detect radiations associated with positon and K-electron capture events. The output of each of the crystals were mixed and fed into a tube. The resolving time of the electronic system was 2 · sec. To minimise the background, pulses whose amplitude corresponded to less than 30 keV energy were rejected.

One of the detectors was a 7.6 cm x 7.6 cm crystal containing a cylindrical well in the centre which was 1.2 cm in diameter and 4.7 cm deep (Harshaw Type 12AK12-S16). The aluminum wall around the well was 0.8 mm thich and this was coated with a layer of Al_2O_3 reflector (20 mg/cm^2). The other crysatl was a 3.8 cm x 3.8 cm flat type which fitted over the well of the larger crystal in order to complete the 4π geometry.

The efficiency for detecting positons and rare earth K- Xrays was studied using a Na^{22} standard which emits positon and Am^{241} which has a prominent 60 keV gamma ray, and energy close to that of the K- X rays of the rare earth elements. The system was found to be 76% efficient for detecting the Na^{22} activity. The γ/α ratio for Am^{241}, where photons greater than 30 keV were detected, was found to be 0.39 ± 0.02. The γ/α ratio for the 59.6 keV photons emitted in Am^{241} alpha

decay was measured by Magnusson [5] to be 0.359). There are, however, a small fraction of gamma rays above 30 keV in the Am^{241} decay scheme in addition to the main 59.6 keV group which could slightly increase the γ/α ratio for photons above 30 keV. The value of 0.39 that we obtain seems reasonable and is an indication that the detection efficiency for radiations in the range of 60 keV is close to 100% for our system.

The problem of estimating the overall efficiency for detecting the β^++ E.C. branch of the nucleides studied in this work is particularly difficult because the decay schemes of these nucleides are not known. In each case, the energy available for β^++ E.C. decay is sufficiently high that a complex decay scheme would be expected. In our method for detecting the β^++ E.C. branch, the greater the multiplicity of cascade gamma rays per decay, the greater is the efficiency for detecting the decay event. On the basis of our measurements with Na^{22} and Am^{241}, and considering that, for the nucleides studied, the β^++ E.C. decay scheme is probably complex, we estimate that the overall efficiency for detecting the β^++ E.C. branch is 0.90 ± 0.10.

To obtain the absolute photodisintegration rate, a gross photon rate decay curve was obtained using an accurately measured portion of the same solution used in preparing the alpha source. The decay rate was measured until an essentially constant counting rate was obtained. This was then subtracted from the gross decay curve. For the Tb^{149m} measurement, the decay curve after subtracting a constant background was a single component corresponding to a half life of 4.3 min, a value in excellent agreement with that obtained from alpha decay measurements[2]. The decay curves for the other nucleides studied, after background subtraction, showed longer-lived components whose half-lives corresponded to known isotopes of Tb and Dy. In order to obtain the most accurate resolution of the desired component from the gross decay curve the following method was employed. An iterative procedure was used to subtract the background plus the contribution from other activities until a single component decay curve was obtained which was linear (log counting

rate or time) within the statistical uncertainty of all the date points and which yielded the identical half-life obtained from alpha counting results. From this curve, the photodisintegration rate at the time zero used for the alpha decay measurements was obtained.

The possibility of interference from Dy and Tb activities not yet discovered with half-life values near those of the ones measured in this work was considered. For Tb^{151}, Dy^{152}, and Dy^{153}, this possibility seems to be remote because all the nucleides immediately in their vicinity have been studied and none has a half-life which would interfere with measurements on these nucleides. To provide a check on the possibility of interference from unknown activities in measurements of the shorter-lived nucleides, comparisons were made of results obtained using samples produced at different bombarding energies. If an interfering activity is present and its excitation function is different from the nucleide being studied the value of the alpha branching ratio obtained will be different when the nucleide is produced at various bombarding energies. Within the statistical uncertainty of the measurements, the experimental values of the alpha branching ratios of Tb^{149m}, Dy^{150} and Dy^{151} appeared to be unaltered by 10% changes in bombarding energy.

The expression used for the alpha branching ratio $\frac{\Gamma_\alpha}{\Gamma}$ is

$$\frac{\Gamma_\alpha}{\Gamma} = \frac{\frac{C_\alpha}{\ell_\alpha}}{\frac{C_\alpha}{\ell_\alpha} + \frac{C_\gamma}{\ell_\gamma}} \times \frac{V_\gamma}{V_\alpha}$$

where C_α = alpha counting rate at = o

C_γ = photodisintegration counting rate at = o

ℓ_α = alpha detection efficiency (0.52)

ℓ_γ = photodetection efficiency (0.90)

V_γ = volume of solution used for gamma counting

V_α = volume of solution used for alpha counting

It is assumed that C_γ is equivalent to the positon plus K-electron capture counting rate.

For Tb^{151}, Dy^{152}, and Dy^{153}, duplicate samples for alpha and gamma counting were prepared and the initial activities of each measured. If satisfactory agreement was obtained between samples, the measurements of the decay curves were begun using one of them. For these nucleides, only one determination of the alpha branching ratio was made. For the shorter-lived nucleides, Tb^{149m}, Dy^{150}, and Dy^{151}, at least two separate measurements of the alpha branching ratio were made. The average values of the measurements are reported in table 1 which summarizes the results.

3. Results

The alpha decay curves from which half-lives and alpha branching ratios were obtained are shown in figs. 1 and 2. The gamma decay curves which were used to obtain the β^++ E.C. branch are shown in figs. 3 and 4. The final values of the alpha particle energies, half-lives and alpha branching ratios are summarized in table 1.

4. Discussion

Using the alpha branching ratios and half-lives obtained in this work, the corresponding alpha partial half-lives were calculated. These were then used to calculate alpha reduced widths (δ^2) which give an indication of the relative alpha decay probability after the energy dependence has been removed. It is defined by the expression

$$\lambda = \frac{\delta^2 P}{h}$$

where λ is the alpha decay constant, P is the barrier penetrability factor, and h is Planck's constant. Barrier penetrability factors were calculated using the method of Rasmussen [6]. To account for electron screening effects, 0.02 MeV was added to each of the alpha decay energies [7]. The calculated values of δ^2 are listed in table 2. Except for Tb^{149m}, the reduced widths were calculated assuming $\ell = 0$ alpha waves only. For Tb^{149m}, the reduced width was calculated for an $\ell = 3$ alpha wave, which is the lowest possible ℓ-wave if the postulated alpha decay scheme

of this nucleide is correct [2]. This scheme assigns the metastable state to an $h_{11/2}$ level which alpha decays to a $d_{5/2}$ Eu^{145} ground state. The average value of δ^2 for an even-even nucleide in this region is 0.08. The low value of 3.6×10^{-3} for the reduced width of Tb^{149m} indicates that alpha decay from this nucleide is considerably hindered. The reduced width for the Tb^{149} ground state alpha transitions is a factor of three larger than that for the metastable state [1]. It is interesting to note that δ^2 for Tb^{151} has approximately the same value as for Tb^{149m}.

The reduced widths for the dysprosium and terbium isotopes are somewhat smaller than those observed for the corresponding gadolinium, holmium and erbium nucleides. These differences are probably related to structural changes associated with the filling of the $d_{5/2}$ and $h_{11/2}$ proton states. Theoretical calculations are currently in progress in order to determine whether the low values of the alpha reduced widths of the terbium and dysprosium isotopes can be accounted for by shell model or pairing model effects.

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Table I. Summary of results

Nuclide	Results from this work					
	$t_{1/2}$	E_α (MeV)	α-branching ratio	$t_{1/2}$	E_α (meV)	α-branching ratio
Tb^{149m}	4.3 ± 0.2 m	3.99 ± 0.03	$(2.5 \pm 0.5) \times 10^{-4}$	4.3 ± 0.2	3.99 ± 0.03	-
Tb^{151}	17.2 ± 0.5 h	3.42 ± 0.03	$(4.8 \pm 0.5) \times 10^{-6}$	19 ± 1 h	3.44 ± 0.10	3×10^{-6}
Dy^{150}	7.20 ± 0.10 m	4.23 ± 0.02	0.18 ± 0.02	7 ± 2 m	4.21 ± 0.06	-
Dy^{151}	18.0 ± 0.2 m	4.06 ± 0.02	0.059 ± 0.006	19 ± 4 m	4.06 ± 0.04	-
Dy^{152}	2.3 ± 0.1 h	3.65 ± 0.02	$(5 \pm 1) \times 10^{-4}$	2.3 ± 0.2 h	3.66 ± 0.05	2×10^{-4}
Dy^{153}	6.4 ± 0.2 h	3.48 ± 0.02	$(3.0 \pm 0.3) \times 10^{-5}$	5 ± 0.5 h	3.48 ± 0.05	4×10^{-5}

Table 2. Alpha reduced widths for Tb and Dy alpha emitters

Nuclide	Alpha Partial Half-life (sec)	δ^2 (MeV)
Tb^{149m}	1.0×10^6	3.6×10^{-3} ($\ell = 3$)
Tb^{151}	1.29×10^{10}	1.19×10^{-3} ($\ell = 0$)
		4.6×10^{-3} ($\ell = 3$)
Dy^{150}	2.40×10^3	0.0517 ($\ell = 0$)
Dy^{151}	1.83×10^4	0.0710 ($\ell = 0$)
Dy^{152}	1.68×10^7	0.0392 ($\ell = 0$)
Dy^{153}	7.68×10^8	0.0196 ($\ell = 0$)

Figure Captions.

Figure 1 Alpha decay curves for Dy^{150} and Dy^{151} from which the quoted half-lives were obtained.

Figure 2 Alpha decay curves for Dy^{152}, Dy^{153}, and Tb^{151} used in obtaining half-lives and alpha branching ratios.

Figure 3 Photodisintegration rate decay curves for A.) Tb^{149m} and B.) Tb^{151} used in obtaining partial widths for β^+ + E.C. decay of these nucleides.

Figure 4 Photodisintegration rate decay curves for A.) Dy^{150} + Dy^{151} B.) Dy^{152} and C.) Dy^{153} used in obtaining partial widths for β^+ + E.C. decay.

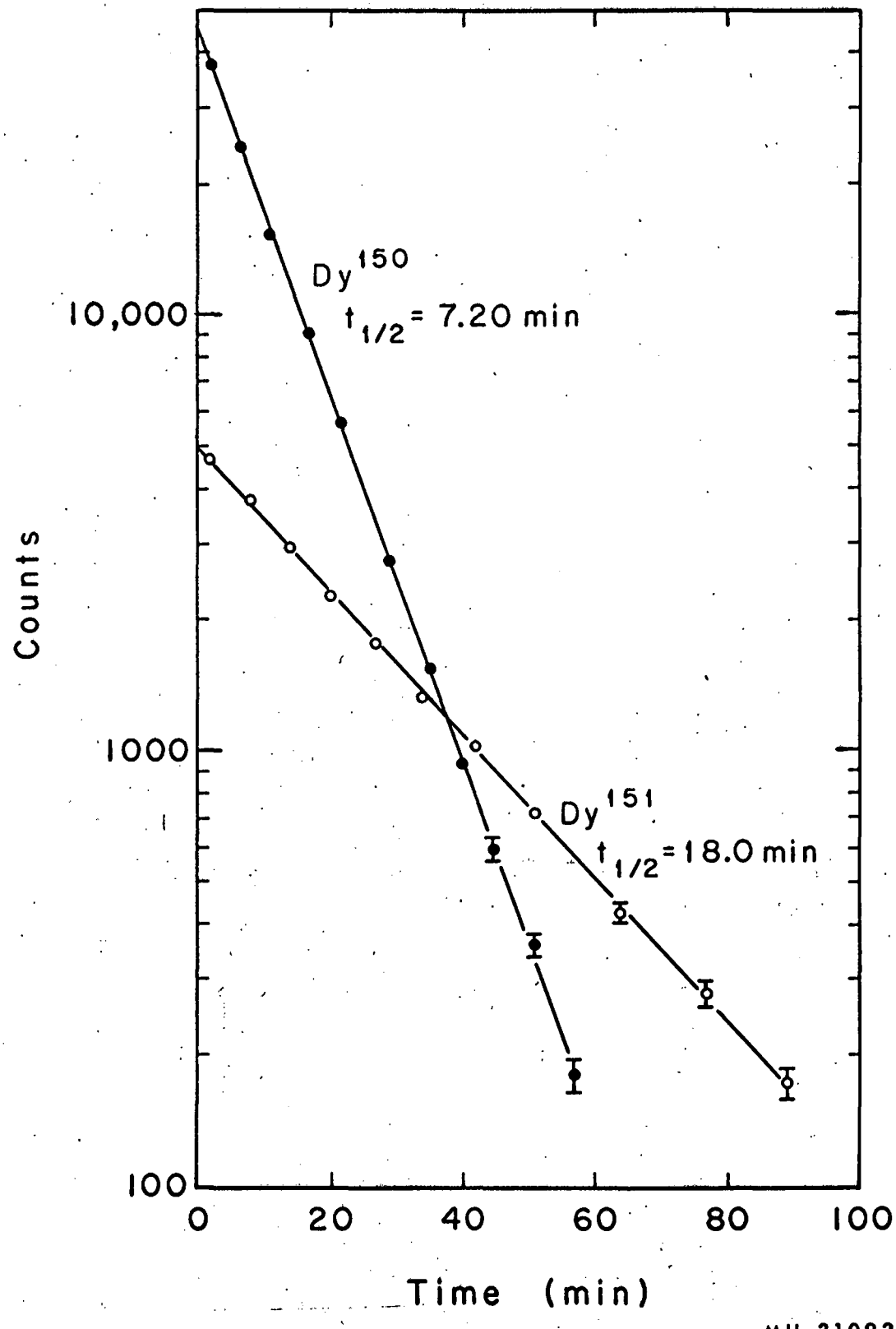

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Fig. 1

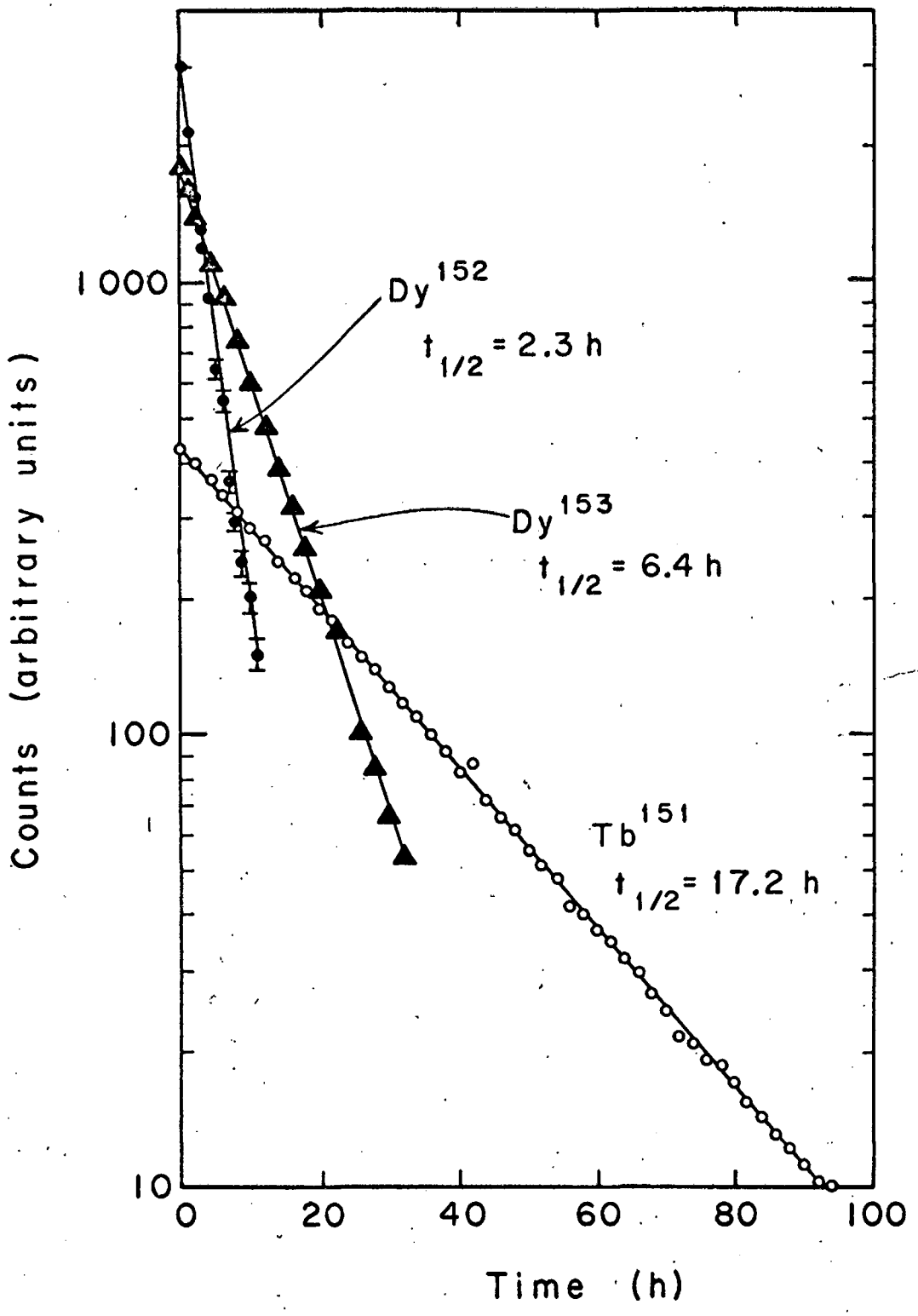

Fig. 2

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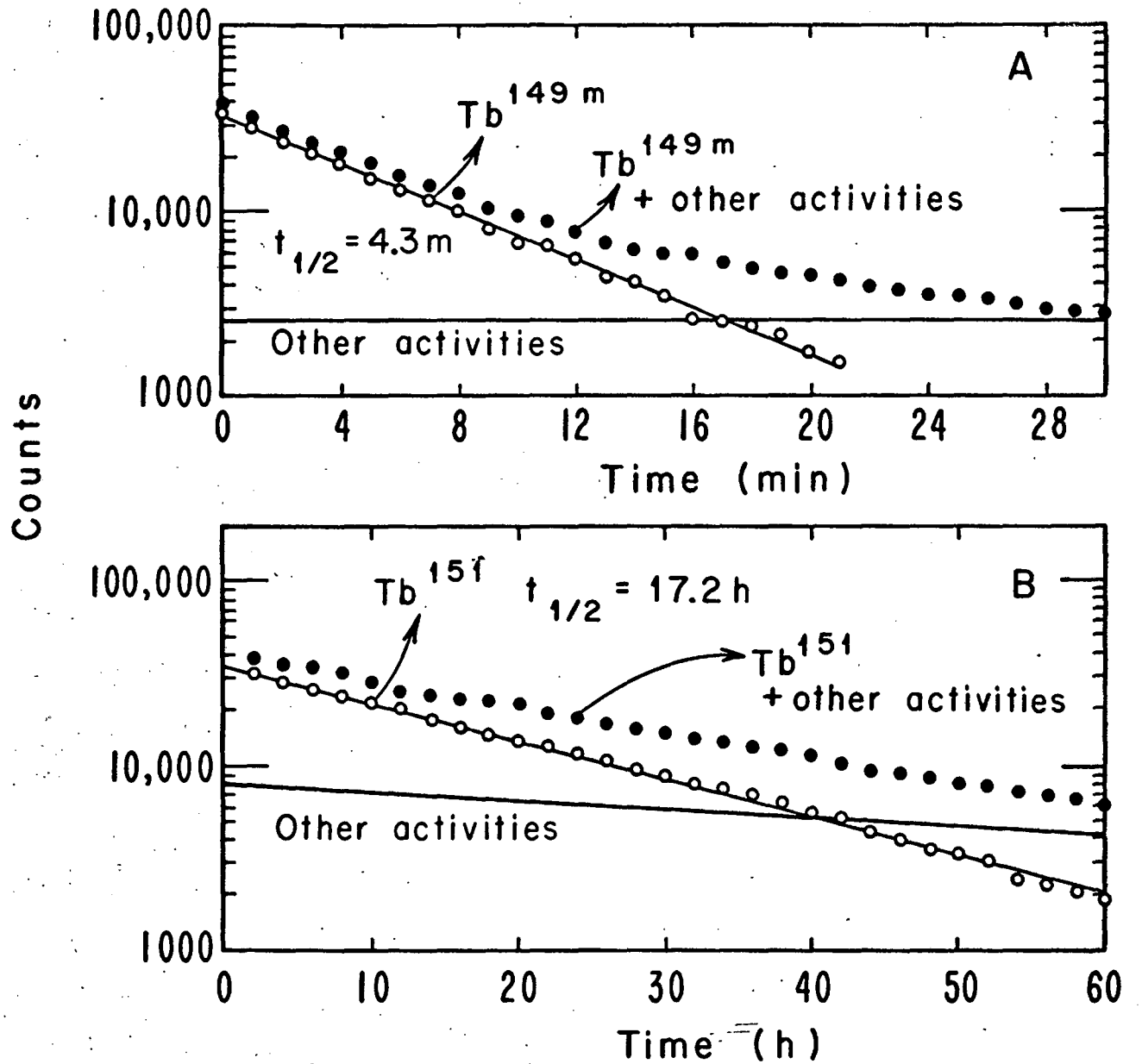

Fig. 3

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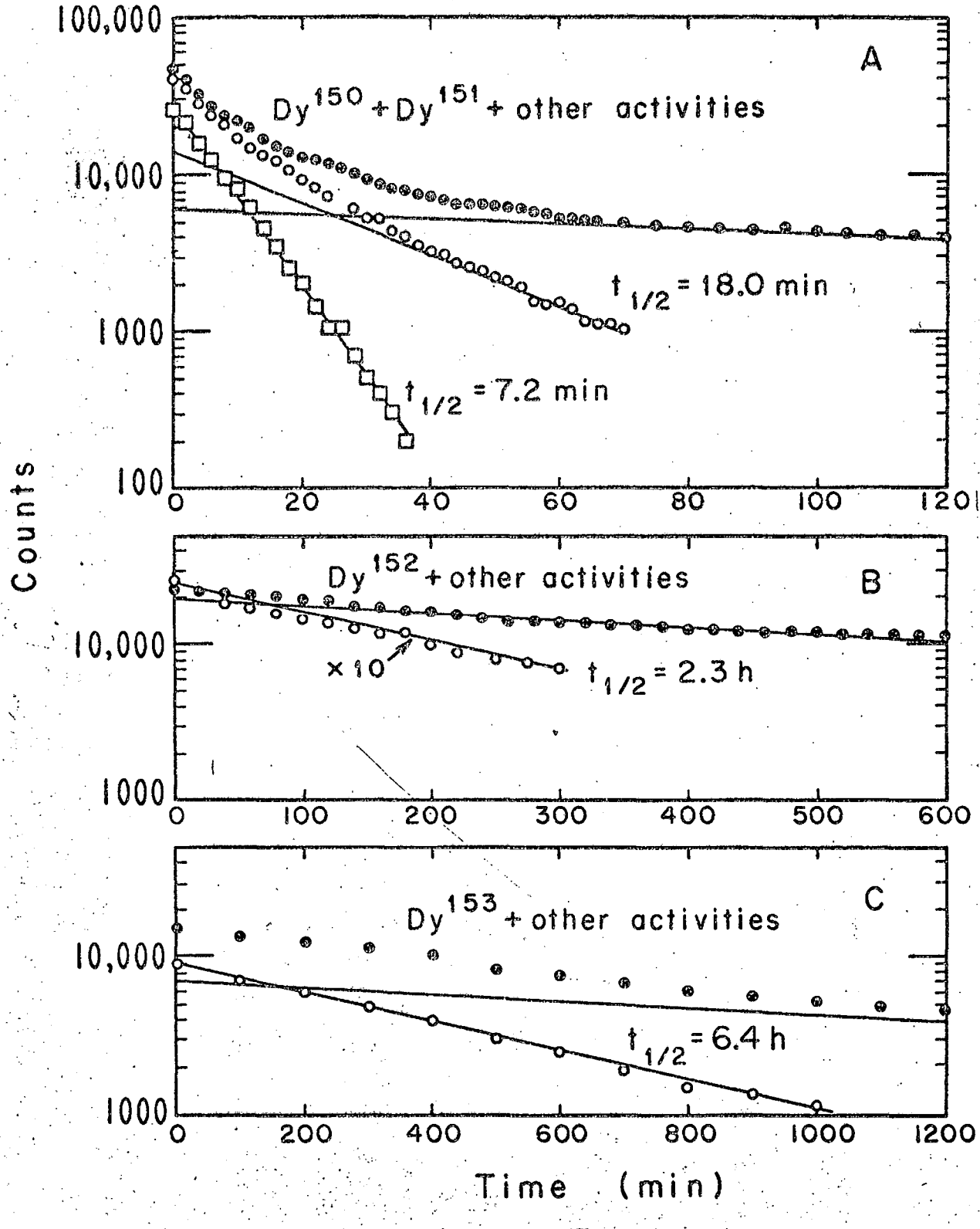

Fig. 4

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