Alphas Branching Ratio in the Decay of ^{149}Tb†

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4.1-h ^{149}Tb has been found to decay by α emission in (22.6±2.3)% of its disintegrations. This was determined by measurements of the α activity in a chemically and isotopically separated ^{149}Tb sample and by determination of the amount of ^{149}Gd formed in this sample through electron-capture decay. Branching ratios of 0.475±0.055 and 0.104±0.013 were deduced for the 894- and 654-keV γ rays of ^{145}Eu formed in the ^{149}Tb α decay.

ALPHA–PARTICLE emission in neutron-deficient rare-earth nuclides was discovered by Thompson *et al.* in 1949,[1] and among the first of these α emitters reported was a nuclide of 4-h half-life emitting 4.0-MeV α particles, later identified[2] as ^{149}Tb. Among the rare-earth α emitters with appreciable α branches (>1%) this turns out to be the only one with a half-life in the range between 18 min (^{151}Dy) and 84 years (^{148}Gd). For this reason, the α particles emitted by ^{149}Tb can be readily detected even in the presence of complex mixtures of other rare-earth nuclides and from thick samples. The formation of ^{149}Tb in foils of high-Z materials, particularly gold, has therefore often served for the monitoring of high-energy proton beams.[3,4] To put such measurements on an absolute basis, cross sections for production of the α branch of ^{149}Tb from gold by protons of various energies have been carefully determined.[3–6] However, the fraction of ^{149}Tb decays proceeding by α emission is still not accurately known; the literature values[7–10] for this fraction range from 0.1 to 0.2, with large uncertainties for the individual values. We have therefore redetermined the α-branching ratio of 4.1-h ^{149}Tb.

The ^{149}Tb sample was produced from high-energy spallation of tantalum. The tantalum target was irradiated in the circulating beam of the alternating gradient synchrotron at Brookhaven with 28-GeV protons for 30 min. After irradiation the target was dissolved and standard chemical separations were performed to separate the rare-earth group from the tantalum and from other radioactivities produced by the high-energy protons. The gross rare-earth fraction was finally precipitated as oxalate. The sample was dried and ignited to oxides in a furnace at 900°C. The oxides were introduced into the ion source of the Brookhaven electromagnetic isotope separator, where they were converted into chlorides by passing carbon tetrachloride vapor over them. The volatile chlorides were then ionized. The mass-separated samples were collected on a long, thin aluminum foil and a strip containing mass 149 with ≪1% contamination by neighboring masses was used for the subsequent measurement. The α-emission rate of this sample was measured repeatedly with a calibrated surface-barrier silicon detector connected to a multichannel pulse-height analyzer. The α-particle energy (3.95 MeV) and half-life (4.1 h) found were those characteristic of ^{149}Tb. After these α-particle measurements, the mass-149 strip was dissolved in acid solution with Tb, Gd, and Eu carriers. The sample was loaded on a Dowex-50 cation exchange column. The eluent used was 0.4M α-hydroxyisobutyric acid solution adjusted to pH 4.1 with ammonium hydroxide. The column was operated at 80°C. The terbium fraction came off the column about 50 min after the beginning of the elution. For the further measurements, a narrow cut of the terbium elution peak was used so as to minimize the amount of ^{149}Gd present in the ^{149}Tb sample. The terbium was precipitated as oxalate. Later yield determinations showed the cross contamination of gadolinium in this terbium sample to be less than 0.1%, whereas the terbium recovery was about 60%.

In the chemically separated Tb sample, the 4.1-h ^{149}Tb was allowed to decay and then the γ radiations of 9.25-day ^{149}Gd and 5.9-day ^{145}Eu, grown in as a result of the electron capture and α decays of ^{149}Tb, respectively, were determined with a calibrated Ge(Li) detector. From the α-disintegration rate of ^{149}Tb measured in the gross $A=149$ sample, from the chemical yield of Tb in the chemically separated sample, and from the amount of ^{149}Gd formed in this sample by electron-capture decay, the α-branching ratio of ^{149}Tb was calculated. Since the abundances of ^{145}Eu γ rays determined by different authors[11–14] vary widely—from

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0.41 to 0.68 per disintegration for the most abundant γ ray at 894 keV—the present measurements were used to deduce, from the ^{149}Tb α-disintegration rate and from the intensities of the 894- and 654-keV γ rays of ^{145}Eu in the ^{149}Tb sample, the branching ratios of these two γ rays.

The counting geometry of the surface-barrier Si detector used for the α measurements was calibrated with an ^{241}Am standard. The linewidth of the α peaks was 35–40 keV full width at half-maximum (FWHM). The resolution obtained with the ^{149}Tb sample was comparable to that of the ^{241}Am standard. The γ radiations of both ^{149}Gd and ^{145}Eu were measured with a 9-cm^2 $\times$1-cm Ge(Li) detector with a resolution of 3.5 keV FWHM at 661 keV.

The amount of ^{149}Gd formed in the ^{149}Tb sample was determined through measurements of the most abundant γ ray emitted in ^{149}Gd decay, at 150 keV. This γ ray was found to decay with a half-life of 9.25$\pm$0.10 days as compared with literature values[15,16] ranging from 9.0 to 10.3 days for the half-life of ^{149}Gd. The abundance of the 150-keV γ ray was taken from Ref. 15 to be 0.48 per disintegration. The same value results from the data in two recent papers[17,18] under the plausible assumption that no electron-capture decays proceed directly to the ground state of ^{149}Eu. An independent check of this branching ratio was made as follows: The intensities of the 150-keV γ ray were measured in two samples of ^{149}Gd which were then allowed to decay into ^{149}Eu. The disintegration rates of the resulting ^{149}Eu were determined through measurement of its K x-ray emission with a calibrated, 2-mm-thick NaI(Tl) detector and single-channel analyzer. These duplicate determinations both yielded values of 0.47 for the branching ratio of the 150-keV γ ray of ^{149}Gd.

Using the above-mentioned half-lives of ^{149}Tb and ^{149}Gd, the branching ratio of the 150-keV γ ray, and the measured disintegration rates, the α-branching ratio of ^{149}Tb was found to be 0.226.

In estimating the error of this branching ratio, there are several sources of uncertainties. Since the amount of decay of ^{149}Tb into ^{149}Gd during the chemical separation on the cation column was not negligible, the time at which the terbium fraction was, in fact, freed of gadolinium is somewhat uncertain. This factor was estimated to contribute an uncertainty of $\pm$4% to the value of the branching ratio. Other uncertainties were estimated as follows: abundance of the 150-keV γ ray of ^{149}Gd—5%; efficiency calibration of γ detector—7%; efficiency calibration of α detector—2%; chemical yield of ^{149}Tb—2%; counting statistics—1%. The root-mean-square combination of these uncertainties is about 10%. Thus we report a value of 0.226$\pm$0.023 for the α branching of 4.1-h ^{149}Tb.

From this α-branching ratio of ^{149}Tb, from the α-decay rate of the ^{149}Tb sample, and from the amount of 894- and 654-keV γ radiation of ^{145}Eu grown in, the abundances of these two γ rays were found to be 0.475$\pm$0.055 and 0.104$\pm$0.013 per disintegration, respectively.

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