SHORT COMMUNICATION

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Measurement of absolute intensity of 1001 keV gamma-ray of ^{234m}Pa

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The 1001 keV γ-ray emission from ^{234m}Pa as an analytical peak has ever increasing importance in direct γ-ray spectrometric measurements of ^{238}U content in samples with the development of high pure Ge detectors of larger crystal volumes and higher efficiency. In this study, γ-ray spectrometric measurement for the determination of the absolute intensity, P_γ, of 1001 keV γ-rays of ^{234m}Pa was carried out using powdered uranium samples and a new experimental value of 0.861±0.015% for P_γ of the 1001 keV γ-ray of ^{234m}Pa has been obtained. The present measured value is greater than most of the recent experimental results and the newly recommended value of 0.835±0.004% for P_γ by 2.5 to 4%. The present result differs from the latest experimental value of 0.92±0.02% by 7%.

Introduction

Since the naturally occurring isotope ^{238}U directly emits only very weak γ-rays of 49.55 keV (0.064%) and at 113.5 keV (0.010%), it is possible, however, to measure the γ-ray emissions from a daughter nuclide which is in radioactive equilibrium with the ^{238}U parent. In many cases, the equilibrium state in natural systems for especially daughters such as ^{210}Pb, ^{214}Pb, ^{214}Bi, etc., which are quite far down the decay chain of ^{238}U may not reach. Therefore, the 1001 keV peak of ^{234m}Pa ($T_{1/2}$ = 1.17 m) in equilibrium with ^{234g}Pa ($T_{1/2}$ = 6.70 h), ^{234}Th ($T_{1/2}$ = 24.1 d) and ^{238}U ($T_{1/2} = 4.468 \cdot 10^9$ y) parent can be selected for uranium analysis. Additionally, it does not include any contribution from any other γ-ray emissions and does not have any interference with other peaks in the high energy region. Thus, even if samples contain high amounts of thorium, this "clean" peak gives more accurate indication about the uranium content of samples without requiring any γ-ray self-attenuation correction or with a negligible self-attenuation correction factor.[1]

The absolute and relative γ-ray emission intensities being as one of the nuclear decay parameters for radionuclides have been studied extensively resulting in new and more precise results.[2–7] However, in γ-ray spectrometric measurements of uranium, the problem in quantifying the radionuclide, ^{234m}Pa on the basis of P_γ parameter, there exists the discrepancy among the recent measured values of P_γ for the 1001 keV γ-rays of ^{234m}Pa, which can reach more than 11%.

The value of 0.59±0.01% for P_γ of the 1001 keV γ-ray transition of ^{234m}Pa, first produced by BJORNHOLM and NIELSEN,[8] who measured the disintegration rates from the beta-particles and γ-ray spectra of ^{234m}Pa by radiochemical separation, was adopted in the evaluations carried out in the 1960s. GUNNINK and TINNEY[2] produced the first published evidence that the P_γ value of 0.828±0.008 for the 1001 keV γ-ray was higher than the previously adopted value of 0.59±0.01%. MOMENI[3] has measured 0.92 for P_γ, without estimating the uncertainty. MOSS[4] reported 0.834±0.007% value for P_γ. SCOT and MARLOW[5] have investigated the P_γ of the 1001 keV γ-rays by using uranium solution and they obtained a value of 0.839±0.005%. SIEMON et al.[6] have also obtained a value of 0.845±0.021% for P_γ using one-year old thin depleted uranium metal foils for analysis, adopting a method based on the measurement of absolute activity ^{234m}Pa in equilibrium with ^{238}U. ANILKUMAR et al.[7] have recently obtained a value of 0.92±0.02% for P_γ using samples of various origin and history by fundamental parameter method. Thus, several measurements of P_γ for the 1001 keV γ-rays of ^{234m}Pa have resulted in doubts concerning the earlier recommended decay data as a value of 0.59±0.01%. This old value is now obsolete and a recommended value of 0.837±0.012% was adopted in 1990.[9] So far, all previously known γ-ray intensities (absolute and relative) in the decay of ^{234m}Pa have been examined and evaluated to recommend a new and more precise values for the γ-ray emissions from ^{234m}Pa. According to these latest evaluations,[10,11] the evaluated value of 0.835±0.004% at 1001 keV γ-ray emission from the ^{234m}Pa is more precise than the value of 0.837±0.012%, currently in adoption.

We have observed that there are only seven experimental results in the literature for the P_γ of 1001 keV γ-rays of ^{234m}Pa and the discrepancy among them seems to be high. So, γ-ray spectrometric measurements were carried out for the determination of the P_γ of 1001 keV γ-ray of ^{234m}Pa using uraniumtrioxide (U_3O_8) powder based on the measurement of the absolute activity of ^{234m}Pa in equilibrium with the parent ^{238}U.

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Experimental

The measurements have been performed by using a n-type Ge detector (REGe) with active crystal volume of approx. 100 cm^3, purchased from Canberra Inc. The REGe detector has a relative efficiency of 22.6% and a resolution of 1.80 keV at 1332.5 keV of ^{60}Co. The full energy peak efficiency as a function of gamma-ray energy for REGe detector was determined using a powder radioactive standard source containing a mixture of ^{109}Cd, ^{57}Co, ^{123m}Te, ^{151}Cr, ^{113}Sn, ^{89}Sr, ^{137}Cs, ^{60}Co, and ^{88}Y radionuclides, obtained from the Isotope Products Laboratories, traceable to NIST. The measured efficiency data were fitted to an efficiency function of the form:

$$\varepsilon_p = \exp\left[\sum_{i=1}^{6} a_i \cdot \ln^i\left(E_\gamma\right)\right]$$

where, ε_p is the full energy peak efficiency at a given gamma-ray energy E_γ and a_i (for $i=1$ to 6) are regression constants. Thus, the REGe detector used was calibrated in the range of 100 to 2000 keV gamma-ray energy. Then, a small amount of dried analytical grade-powdered natural uranium trioxide (U_3O_8) of about 1.5 to 8 g, was filled into polystyrene tubes of 1.4 cm inner diameter, with 1 mm wall thickness. Since the U_3O_8 stock has stored more than 15 years, it is expected that radioactive equilibrium with ^{234m}Pa and the parents, ^{234}Th and ^{238}U was achieved. The samples were positioned coaxially with the detector and counted at a distance of 12.5 cm to avoid coincidence summing effects. The predetermined counting time for each run 80,000 to 210,000 seconds was high enough to ensure good statistical quality of data. The measurements were repeated three to five times to improve statistical precision. Thus, a total of forty gamma-ray spectra of uranium were analyzed. The mean values of the measured count rates were used in the calculations. The net sample count for each measurement was obtained after background subtraction of the peak area, and it was corrected for γ-ray self-attenuation. The absolute intensity for gamma-emission, P_γ at a given energy, E_γ can be calculated as:

$$P_\gamma = N_\mathrm{p} / \left(t_\mathrm{c} \cdot A \cdot \varepsilon_\mathrm{p}\right)$$

where N_p is the net counts under the photopeak, t_c is the measuring time, A is the sample activity (Bq) and ε_p is the full energy peak efficiency at a given γ-ray energy, E_γ.

The γ-ray self attenuation correction factor for 1001 keV emission, F_s is negligibly small, however, this correction factor has been taken into account in the determinations. F_s was calculated by:

$$F_s = \mu t / (1 - \exp(-\mu t))$$

where μ (cm^{-1}) is the linear attenuation coefficient for U_3O_8 and t (cm) is the effective sample thickness. The effective thickness, $t = m/(\rho \cdot S)$ was calculated from the area of the extended-source S, the density of the sample, ρ and the weight of the sample, m.[12] The mass attenuation coefficients, μ/ρ (cm^2·g^{-1}) of U_3O_8 were taken from XCOM photon cross sections database of BERGER and HUBBELL.[13] For a given sample thickness of a cylindrical tube geometry, the self attenuation correction factors for 1001 keV γ-ray of U_3O_8 are in the range of 1.015 to 1.12, depending on the amount of the sample.

Results and discussion

From the measurements, a new experimental result of 0.861±0.015% for P_γ of 1001 keV γ-ray of ^{234m}Pa has been obtained. The present value is an average of the measured results obtained by counting of different amounts of natural U_3O_8 by the a REGe detector.

Table 1. Comparison of the present result with other results for the absolute intensity of the 1001 keV gamma-ray of ^{234m}Pa

Year	Data type	Reference	Gamma-ray intensity, P_γ, %	Relative bias, %
2002	Experimental	This work	0.861 ± 0.015	–
1999	Experimental	ANILKUMAR et al.[7]	0.92 ± 0.02	–6.85
1999	Evaluated	NZURUBA[11]	0.835 ± 0.004	+3.02
1998	Evaluated	ADSLEY et al.[10]	0.835 ± 0.011	+3.02
1996	Evaluated	FIRESTONE and SHIRLEY[9]	0.837 ± 0.010	+2.78
1992	Experimental	SIEMON et al.[6]	0.839 ± 0.005	+2.55
1990	Experimental	SCOTT and MARLOW[5]	0.839 ± 0.005	+2.55
1986	Experimental	MOSS[4]	0.834 ± 0.007	+3.13
1986	Evaluated	BROWNE and FIRESTONE[14]	0.65	+24.51
1982	Experimental	MOMENI[3]	0.92	-6.85
1971	Experimental	GUNNINK and TINNEY[2]	0.828 ± 0.008	+3.83
1963	Experimental	BJORNHOLM and NIELSON[8]	0.59 ± 0.10	+31.47

Main sources of uncertainty of the absolute γ-ray intensity measurement are the statistical error on the counting (<0.2–0.3%) with a confidence level of 96%, the γ-ray detection efficiency of REGe detector (1.2–1.7%), the sample activity (0.1%) and the systematic error (<0.5%). The final combined uncertainty of the measured value of P_γ is estimated to be 1.3 to 1.8%. The present P_γ value together with the previous experimental and evaluated values are given Table 1.

The present experimental value agrees within 2.5 to 4% with the measurements of GUNNINK and TINNEY,[2] MOSS,[4] SCOTT and MARLOW,[5] and the recent evaluated values by FIRESTONE and SHIRLEY,[9] ADSLEY[10] and NZURUBA.[10] However, there exists a discrepancy of 7% between the present value and the result of 0.92%, measured by MOMENI[3] and ANILKUMAR et al.[7] The value of 0.92% for P_γ is larger than all the recent experimental and evaluated results given in Table 1. However, our measured result of 0.861±0.015% implies that the newly recommended value of 0.835±0.004% of P_γ for 1001 keV γ-rays is small and the latest experimental results should reevaluate for obtaining a newly recommended value.

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