MEASUREMENT OF ABSOLUTE GAMMA RAY EMISSION PROBABILITY OF 1001 keV FROM THE DECAY OF ^{234m}Pa

Çetiner B.[*], *Karadeniz H.*[*], *Yücel H.*[*], *Özgür İ.*[**], *Çetiner M.A.*[*] *and Özmen A*[***]

* Ankara Nuclear and Research Training Center, Beşevler, Ankara, Turkey
** Osmangazi University, Faculty of Science and Art, Physics Dept., Eskişehir, Turkey
*** Gazi University, Faculty of Science and Art, Physics Department, Ankara, Turkey

Abstract

In recent years, the 1001 keV gamma ray emission from ^{234m}Pa as an analytical peak is commonly used in the direct gamma ray spectrometric measurements of ^{238}U content. ^{234m}Pa is the second daughter of ^{238}U and rapidly reaches secular equilibrium with parent nucleus. This "clean" peak is well resolved by high purity Ge detectors and gives more accurate indication of uranium content without requiring any self attenuation correction.

Several experimental results for the absolute emission probability of the 1001 keV gamma ray of ^{234m}Pa have resulted in doubts concerning the old recommended value of 0.59±0.01% obtained by following a radiochemical separation method. Therefore, this old value is now obsolete and a newly value of 0.835±0.004% is recommended. In this study the gamma-ray spectrometric measurements for the determination of the absolute emission probability of the 1001 keV gamma ray of ^{234m}Pa were carried out using the powdered uranium samples. A new experimental value of 0.861±0.015% for the absolute gamma ray emission probability for the 1001 keV gamma ray of ^{234m}Pa has been obtained. The present measured value agrees within 2.5 to 4% with the most recent experimental results appeared in the literature and the newly recommended value of the absolute emission probability for the 1001 keV gamma-ray of ^{234m}Pa. However, it is different from the latest experimental value of 0.92±0.02%, obtained by Anilkumar et al. (1999).

1. Introduction

With the introduction of the high-resolution gamma ray spectrometry with the high purity Ge detectors having the larger crystal volumes(i.e. with the higher detection efficiency), the gamma-ray emission probabilities (absolute and relative) as one of nuclear parameters for various radionuclides have been studied extensively resulting in a new and more precise results[1,2]. The decay data obtained accurately from these studies are necessary for the quantitative assessments of

radionuclides, for example, for the determination of concentrations of those contained in the samples by employing the gamma ray spectrometric method.

The naturally occurring isotope, ^{238}U, a radioactive primordial elements such as thorium and potassium is one of the obvious and most important sources of radioactivity in the environment. The measurements of primordial nuclides in areas of igneous geology, in nuclear wastes and in environmental samples are often carried out by high-resolution gamma-ray detectors. Since ^{238}U isotope directly emits only very weak gamma rays which are at 49.55 keV (0.064%) and at 113.5 (0.010%) shown in Fig.1, it is possible, however, to measure the emission of gamma rays from of a daughter nuclide which is in radioactive equilibrium with the ^{238}U parent.

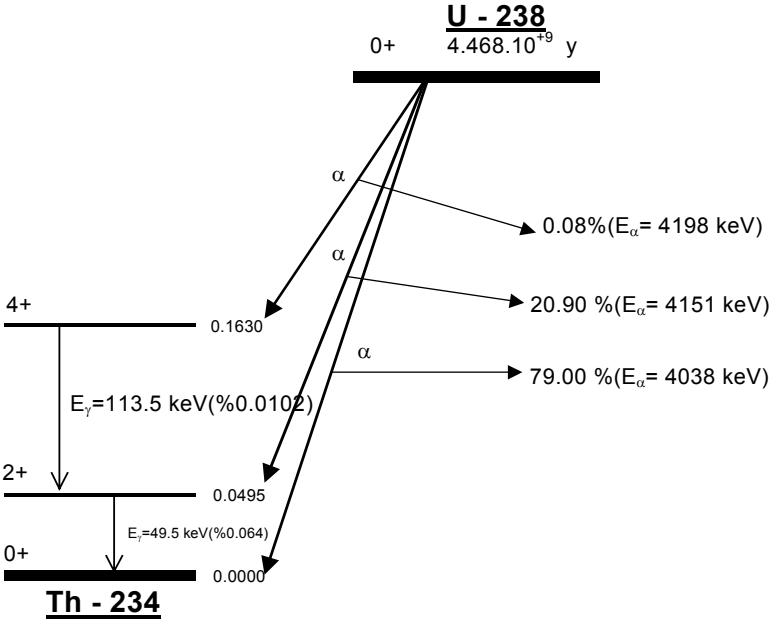

Fig 1. Decay scheme of ^{238}U

This condition is fulfilled by one of the first three daughters of ^{238}U, viz. ^{234}Th (24.1 d), ^{234m}Pa(1.17 m) and ^{234g}Pa(6.70 h) because of very short-lived compared with ^{238}U(4.468x10^9 y). For example, the necessary time for the radioactive equilibrium for the ^{234Th} with ^{238}U parent is attained about 168 days. The partial decay scheme for the first three daughters of ^{238}U chain is shown in Fig.2.

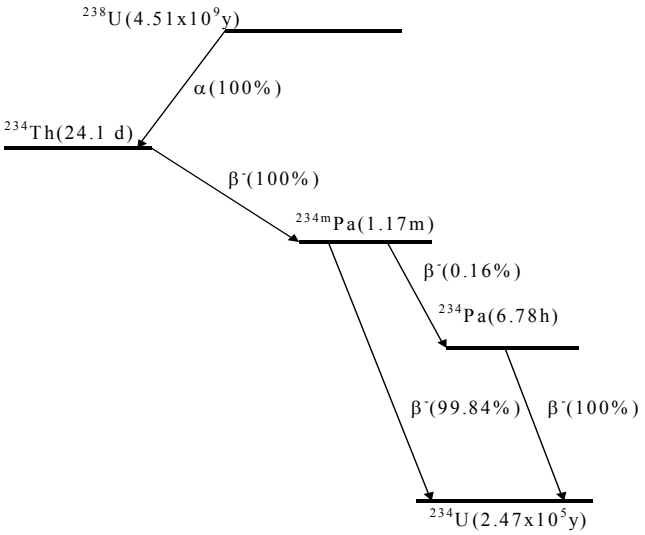

Fig. 2. Decay scheme for ^{238}U-^{234}Th-$^{234, 234m}$Pa-^{234}U chain

On the other hand, in many cases, this equilibrium state in natural system may not reach, especially for a daughter such as ^{210}Pb, ^{214}Pb, ^{214}Bi etc. from the uranium decay series that is quite far down the decay chain. Therefore, recent gamma-ray spectrometry measurements have focused on determining the ^{234Th} and ^{234m}Pa in which they are secular equilibrium with ^{238}U in the samples. However, since the 63.3 keV peak of ^{234Th} includes contributions from the 63.9 keV(0.023%) emission from ^{231}Th , the 63.9 keV(0.255%) emission from ^{232}Th and the 62.9 keV(0.018%) emission from ^{234Th}, a recent study concerning the measurements of uranium concentrations in the samples emphasizes the use of 1001 keV peak of ^{234m}Pa, which is a "clean" peak well resolved by HpGe detectors[3]. The 1001 keV peak of ^{234m} Pa in equilibrium with ^{234g}Pa, ^{234}Th and ^{238}U can also be selected for the uranium analysis since it does not include any contribution from any other gamma emissions and does not have any interference with other peaks in the high energy region even if the samples contain high amounts of thorium. Thus, this analytical peak gives more accurate indication of uranium concentration in the samples without requiring any gamma ray self-attenuation correction or with a negligible self attenuation correction factor because the 1001 keV gamma-ray is in high-energy region in the observed gamma-ray the spectrum. However, the problem in quantifying this radionuclide on the basis of its decay parameters in the gamma-ray spectrometric measurements is that the discrepancy among the recent absolute

emission probabilities (P_γ) measured for the 1001 keV line of ^{234m} Pa can reach more than 15%.

The P_γ value for the 1001 keV line of ^{234m}Pa first produced by Bijornholm and Nielsen [4] who measured the disintegration rates from the beta particles and gamma-ray spectra of ^{234m}Pa in equilibrium with ^{234g}Pa(6.70 h) and the parent, ^{234} Th (24.1 d) . This single datum, which was a value of 0.59±0.01%, generated in the 1960s was extremely important for the P_γ of the 1001 keV gamma-ray transition of ^{234m}Pa following internal conversion, and it was adopted in the evaluations carried out in the past.

Scot and Marlow[1] have investigated the P_γ of the 1001 keV gamma line of ^{234m}Pa by using uranium solution and they obtained a value of 0.839±0.005%. Siemon et al.[5] have also obtained a value of 0.845±0.021% for the P_γ of this gamma-ray using one-year old thin depleted uranium metal foils for analysis, adopting a method based on measurement of absolute activity ^{234m}Pa in equilibrium with ^{238}U. Anilkumar et al.[6] have recently obtained a value of 0.92±0.02% for the P_γ of 1001 keV gamma ray of ^{234m}Pa using samples of different origin and history by fundamental parameter method. This last value is different from the other previously quoted experimental values.

Several measurements of the P_γ of the 1001 keV gamma-ray of ^{234m}Pa have resulted in doubts concerning earlier recommended decay data as a value of 0.59±0.01%. Therefore all previously known gamma-ray probabilities (absolute and relative) in the decay of ^{234m}Pa have been examined and evaluated to obtain a new and more precise values for the gamma-ray emissions from ^{234m}Pa by Nzuruba [7]. He suggested that the evaluated value of 0.835±0.004% at 1001.025 keV from the ^{234m}Pa is more precise than the value of 0.837±0.012% currently in adoption.

In the present study we have observed that there are only six experimental results for the P_γ of 1001 keV line of ^{234m}Pa in the literature survey and the discrepancy among the few experimental results for the 1001 keV gamma line seems to be high. So, the gamma-ray spectrometric measurements for determination of the P_γ of 1001 keV line of ^{234m}Pa were carried out using powder uraniumtrioxide (U_3O_8) samples by the method based on measurement of absolute activity of ^{234m}Pa in equilibrium with the parent ^{238}U.

2. Experimental

Gamma-ray spectrometric measurements have been performed, in turn, using two different high pure Ge detectors manufactured by Canberra Inc. One of the HPGe detectors used is a p-type crystal with active volume of approx. 57 cm^3 with a relative efficiency of 12.4%. The other is a n-type crystal with active volume of approx. 100 cm^3 with a relative efficiency of 22.6%. The full energy peak

efficiency as a function of gamma-ray energy for each Ge detector was determined using the 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 Isotope Products Laboratories, traceable to NIST. The measured efficiency data were fitted to an efficiency function of the form:

$$\varepsilon_P = \exp\left[a_0 + \sum_{i=1}^{6} a_i \cdot \ln^i\left(E_\gamma\right)\right] \quad (1)$$

where, ε_p is the full energy peak efficiency at a given gamma ray energy E_γ and a_0, a_i (for i= 1 to 6) are regression constants . The uncertainties on the detector efficiencies are about 2% in the range of 50 to 2000 keV gamma ray energy. Then, only a small amount, about 1.5 to 8 g, of dried and powdered samples are filled into the polystyrene tubes with 1 mm wall thickness. The samples were filled to same height to ensure identical counting geometry. The counting time for each run predetermined was high enough to ensure good statistical quality of data, and the counting times chosen for runs varied between 80,000 s and 210,000 s. A schematic diagram of the counting system used is shown in Fig 3.

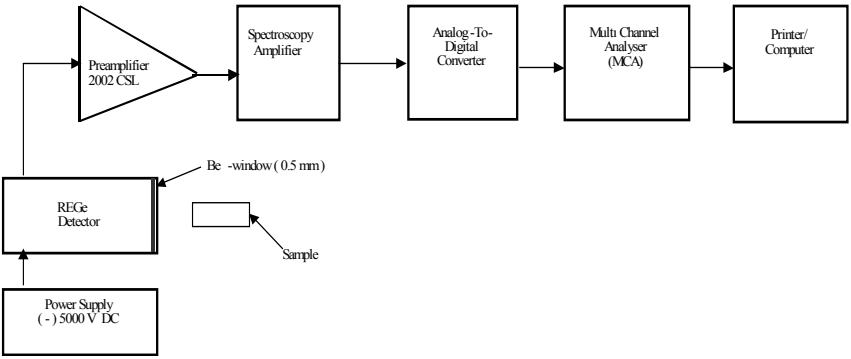

Fig. 3. Block diagram of gamma counting system with a REGe detector

The samples were coaxially positioned with the detector in a given distance. The measurements were repeated three to ten times to improve statistical precision. Thus, the mean values of the measured count rates were used in the calculations. The net sample count rate for each measurement was obtained after background subtraction of the peak area. The emission probability, P_γ is calculated as follows:

$$P_\gamma = \frac{\lfloor N_p / t_c \rfloor}{A \cdot \varepsilon_p} \cdot F_g \tag{2}$$

where, N_p: the net counts under photopeak, t_c: the measuring time, A: sample activity (Bq) and F_g: gamma ray attenuation factor and ε_p is the full energy peak efficiency at a given gamma ray energy E_γ.
The correction factor for gamma-ray self attenuation, F_g has been taken into account by the following equation:

$$F_g = \frac{\mu x}{1 - e^{-\mu x}} \tag{3}$$

where, μ (cm^{-1}) is the linear attenuation coefficient of U_3O_8 and x (cm) is total sample thickness. Mass attenuation coefficients, μ/ρ ($cm^2.g^{-1}$) were taken from XCOM database of Berger and Hubbell[8]. The estimated values of F_g for 1001 keV gamm ray in U_3O_8 samples are small however, they found in the range of 1.0015 to 1.0032 for 1001 keV gamma ray, depending on sample size.

3. Results and Discussion

The measured absolute gamma-ray emission probability, P_γ for the 1001 keV gamma-ray emission from ^{234}Pa, together with previous experimental results and evaluated values are given Table 1.
The present value is the weighted average of the results obtained by using two Ge detectors. Main sources of uncertainty of the absolute emission probability measurement are statistical error (<0.2-0.3%), efficiency of REGe or HPGe (1.5-2.0%), sample activity (0.1%), systematic error (0.5%). Final combined uncertainty of the measured value of the P_γ for 1001 keV from ^{234m}Pa is estimated within 1.2 to2.1%.

The present experimental value agrees within 2.5 to 4 % with the measurements of Scott and Marlow[1], Moss[2], Gunnink and Tinney[10], and the most recent evaluated value of 1001 keV from ^{234m}Pa by Nzuruba[7]. However, there existed a discrepancy between the present value and the result of Anilkumar et al.[6] by 7%. The measurement of Anilkumar et al.[6] seems to be unrealistically larger than our measurement and other recent experimental results given in Table 1, and is different from the newly recommended value of 0.835±0.004% by 9.2%.

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

Year	Data type	Reference	Gamma-ray emission probability, P_γ(%)	Relative Bias (%)
2002	Experimental	This work	0.861±0.015	-
1999	Experimental	Anilkumar et al. [6]	0.92±0.02	-6.85
1999	Evaluated	Nzuruba [7]	0.835±0.004	+3.02
1990	Experimental	Scott and Marlow [1]	0.839±0.005	2.55
1986	Experimental	Moss [2]	0.834±0.007	+3.13
1986	Evaluated	Browne and Firestone's TRI [9]	0.65	+24.51
1971	Experimental	Gunnink and Tinney[10]	0.828±0.008	+3.83
1963	Experimental	Bjornholm and Nielson[4]	0.59±0.10	+31.47

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