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**NUCLEAR
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Section A

Use of the 1001 keV peak of ^{234m}Pa daughter of ^{238}U in measurement of uranium concentration by HPGe gamma-ray spectrometry

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Abstract

For the direct gamma-ray spectrometric measurements of uranium concentrations in the samples, the use of 1001 keV peak of ^{234m}Pa , second daughter of ^{238}U is emphasized. This “clean” peak is well resolved by HPGe detectors and gives accurate indication of uranium concentration in the samples without any self-absorption correction. The 1001 keV peak of ^{234m}Pa in the ^{238}U chain is selected because 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. The activity of ^{234m}Pa is determined by calibrating a HpGe detector with uranium standards. The measurement of 1001 keV gamma-ray emission from ^{234m}Pa by high-resolution gamma-ray spectrometry provides the basis for a reliable determination of ^{238}U in the samples. The results obtained from the 63.3 keV peak of ^{234}Th and those of the 1001 keV peak from ^{234m}Pa in the ^{238}U chain for the uranium standards are compared with the certified values of the same samples. The results obtained from the measurements of 1001 keV peak from ^{234m}Pa agreed to within 2–5% with the certified activity values of ^{238}U in the samples with uranium content ranging from 0.014 to 1.02 wt%. The results indicate that the uranium concentrations in the samples can be determined to within 5% error by about 14 h counting in a HpGe detector system. © 1998 Elsevier Science B.V. All rights reserved.

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1. Introduction

One of the most obvious sources of radionuclides in the environment is nature itself. The sources of greatest interest in most environmental studies are primordial nuclides in geosphere. Particularly, in

areas of igneous geology, concentrations of uranium and thorium series, nuclides as well as naturally occurring potassium are significant. It is known that the natural analogue studies using uranium series radionuclides are one of the useful ways for predicting the long-term migration behaviour of the actinides in the geosphere [1,2]. Furthermore, in recent years, a need has arisen to find and produce materials especially in low radioactive primordial elements’ thorium, uranium and potassium

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[3]. For the measurement of uranium series nuclide concentrations, radiochemical analysis followed by alpha spectrometry is most commonly used [4,5]. However, this method is time consuming and complicated. On the other hand, the gamma-ray spectrometry is a simple, non-destructive and fast method, and suitable for accumulating data for many radionuclides simultaneously. So, high-resolution gamma-spectrometry has been extensively used for the quantitative analysis of uranium and thorium [6] and for the absolute determination of uranium content in rocks [7]. If one wishes to measure the ^{238}U content of a sample independent of the ^{235}U content, one is faced with the problem that the ^{238}U isotope emits directly only very weak gamma radiation which is at 49.55 keV (0.064%). It is possible, however, to measure the emission of gamma rays from a daughter nuclide which is in equilibrium with the ^{238}U parent. In many cases this equilibrium state in a natural system may not have been reached, especially for a daughter that is quite far down the decay chain.

In the direct gamma-ray spectrometric measurements of the ^{238}U by using NaI(Tl) crystals, all earlier works which used gamma rays of the distant members (lower Z products), like ^{210}Pb , ^{214}Pb , ^{214}Bi of the uranium series, and which had not predetermined the equilibrium status may obtain the wrong results [8,9]. In such studies, the ^{238}U concentrations in samples are derived by assuming secular equilibrium between ^{238}U and daughters ^{226}Ra or ^{214}Bi . This assumption, however, cannot be considered valid in many situations where the disequilibrium conditions between ^{238}U and its daughters can occur due to difference in their geochemical behaviour, and also due to the presence of some very long-lived daughters. Nevertheless, the new methods for the simultaneous analysis ^{226}Ra , ^{228}Ra and other decay products of ^{238}U in water, sludge, sediments, soil, coal, ash and other types of samples are still developed and applied by scientists using NaI(Tl) detector systems [10,11].

In recent studies, although the accuracy has been greatly improved with introduction of the high-resolution gamma spectrometry with the pure Ge detectors, the prominent peaks of the distant members of ^{238}U such as 352 keV from ^{214}Pb and 609 keV from ^{214}Bi are not used for the measure-

ment of ^{226}Ra , as possible leakage of ^{222}Rn , daughter nuclide of ^{226}Ra . Because the ^{238}U decay chain passes through ^{222}Rn progeny, which is a noble gas with a half-life of 3.8 days and is lost from the sample container, and one needs to have the sample air-tight sealed and kept for about 3 weeks to reach the equilibrium in the ^{238}U series. In the absence of any directly measurable gamma-ray from ^{238}U itself and in cases where the assumption of the secular radioactive equilibrium in the ^{238}U is not permissible. To obtain the exact information about the measurement of ^{238}U by gamma-ray spectroscopy, it is essential that any one of the daughters of ^{238}U should exist in equilibrium with ^{238}U . This condition is fulfilled by one of the first three daughters of ^{238}U , namely ^{234}Th (24.1 d), $^{234\text{m}}\text{Pa}$ (1.17 m) and ^{234}Pa (6.78 h). Because these three daughter nuclei are very short-lived compared with ^{238}U so that radioactive equilibrium is quickly achieved, these daughter products are in secular equilibrium. The natural radioactive decay series for the ^{238}U chain contains 19 radionuclides. The decay scheme for the first three daughters of the ^{238}U chain is shown in Fig. 1.

Since ^{234}Th is the first daughter of ^{238}U and has a relatively short-life 24.1 days, it ensures that the

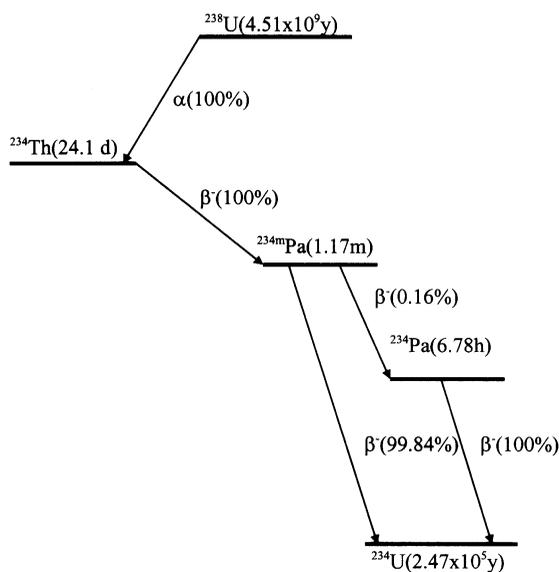


Fig. 1. Decay scheme for ^{238}U – ^{234}Th – $^{234\text{m}}\text{Pa}$ – ^{234}U chain.

radioactive equilibrium with ^{238}U is attained in about 168 days. The gamma peaks of about 63.3 and 93 keV of ^{234}Th have been considered useful for analytical purposes [7,12]. It is known that the conventional NaI(Tl) crystals are incapable of resolving these emissions from other gamma- and X-ray peaks in this low-energy region. In this case, the analyst is faced with another problem, that the 93 keV peak of ^{234}Th is actually a doublet. The 93 keV gamma emission of ^{234}Th consists of 92.370 (2.61%) and 92.793 keV (2.58%) [13]. The 93 keV peak is also thorium (K) X-ray peak. So for samples containing high amounts of thorium, the 93 keV peak does not give accurate indication of the uranium concentration, because thorium (K) X-rays will be produced by self-excitation [7]. The 63.3 keV gamma emission (4.5%) is a cleaner peak of ^{234}Th for uranium analysis especially when using high-purity Germanium Low-Energy Photon Spectrometers (LEPS) that are very capable of resolving gamma-rays in the low-energy region. However, the 63.3 keV peak includes contributions from the 63.9 keV (0.255%) emission from ^{232}Th , the 63.9 keV (0.023%) emission from ^{231}Th and the 62.9 keV (0.018%) emission from ^{234}Th [14]. Of these, the 63.9 keV peak of ^{231}Th , which is a daughter of ^{235}U , is a potential significant contribution only for samples highly enriched in ^{235}U . But the contribution of 63.9 keV (^{231}Th) in the natural uranium sample may be ignored since the concentration of ^{235}U in a natural sample is very low, i.e., 0.71%.

It is purpose of the present work to show that the uranium concentrations in the samples can be measured accurately by use of the 1001 keV gamma emission from $^{234\text{m}}\text{Pa}$ without any self-absorption correction. When the measurements of uranium in the samples are carried out by using a HPGe Well detector instead of LEPS, the 1001 keV peak of $^{234\text{m}}\text{Pa}$ in the high-energy region is considered to be the most suitable “clean” peak. The 1001 keV peak does not include any contribution from any other gamma emissions, and does not have any interference with other peaks in the region of interest. Besides this, in surveying of literature, several recent gamma-ray measurements of the absolute emission probability of the 1001 keV peak of $^{234\text{m}}\text{Pa}$ have resulted in giving a newly recommen-

ded value of 0.835%, and this new value provides a consistent basis for measurement of $^{234\text{m}}\text{Pa}$ in the samples [15]. The results obtained from the 63.3 keV peak of ^{234}Th for the Certified Reference Materials (CRM) are compared with those of 1001 keV peak from $^{234\text{m}}\text{Pa}$ for the same materials.

2. Experimental procedure

The commercially available HPGe Well-type detector was used in these measurements. The detector was a p-type HPGe Well (GWL series) with total active volume of 110 cm³ manufactured by EG&G Ortec, Inc. The crystal size of HPGe Well detector was 5.48 cm in diameter and 6.11 cm in length. The detector has 4.0 cm in active well depth and 1.6 cm in well inside diameter. The measured resolution of the HPGe Well detector was 2.07 keV for the 1332.5 and 1.29 keV for 122 keV. The detector was shielded by lead lined with cadmium and copper on all sides. In the HPGe Well detector system, the preamplified signal is processed through a Spectroscopy Amplifier and an ADCAM Multichannel Buffer (Model 919 SPECTRUM MASTER from EG&G Ortec) interfaced to a Personal Computer for the functions of data acquisition/storage and display/analysis by using a software package named OMNIGAM.

Most of the available CRM samples were stored in our laboratory in 1986. Therefore, the equilibrium between ^{238}U and its near daughters (^{234}Th , $^{234\text{m}}\text{Pm}$) is expected to be attained for 10 years. Only a small amount, about 8 g, of dried and powdered sample is filled into the polystyrene tube of 1.4 cm inside diameter and 1 mm in wall thickness. Then each of the sample is carefully weighed with a balance having a sensitivity down to mg range. The samples were filled to same height to ensure identical counting geometry. The counting geometry for the detector is illustrated in Fig. 2.

The counting time for each measurement predetermined was 50 000 s. The counting times were high enough to ensure good statistical quality of data. The measurements for each sample were repeated three times to improve the statistical precision. Thus, the mean values of the measured count rates were used in the calculations. Also the

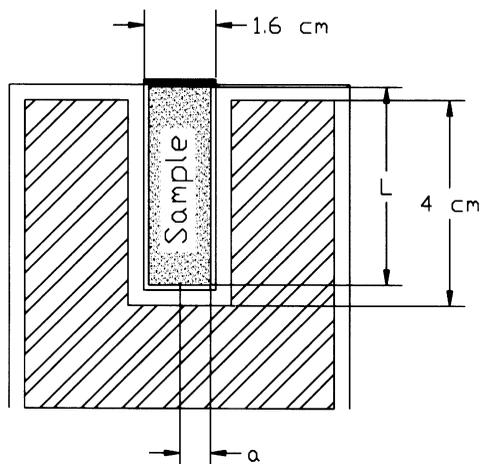


Fig. 2. Counting geometry of the well detector sample holders. In an Ortec HPGe GWL-series, sample height $L = 4.4$ cm, sample radius $a = 0.7$ cm.

background spectrum was collected several times for the same period as that for uranium samples, and the average value from the background runs was used in data analysis. The net sample count rate was obtained after room background subtraction of the peak area. In peak analysis procedure, the variation in the full-energy peak areas by applying automatic computation and manual selection is estimated to be less than 0.5%. Dead times were typically less than 1% due to the low specific activity of uranium standard samples. No effort was made to correct for random summing losses, since for the most active sample the full spectrum count rate was less than 50 counts/s and electronic pulse-pileup and life-time corrections were employed. Also, the counting system was stable since the energy resolution remained the same for counts of 50 000 s.

3. Results and discussion

The densities of powdered CRMs used for evaluation of procedure were in the range of 0.92–1.11 g/cm³. The thicknesses of the samples were the same and chosen as 1.4 cm to minimise the self-absorption effect for especially low-energy gamma-rays.

Table 1

The measured photopeak efficiencies of the standard samples with different uranium contents for the HpGe well detector.

Uranium standard		Photopeak efficiency		
Code	U-content (%)	Gamma-ray energy (keV)		
		46.5	63.3	1001
RG-1	0.040 ± 0.0004	0.4539	0.4621	0.0642
BL-3	1.02 ± 0.01	0.4365	0.4543	0.0624
	Mean	0.4452	0.4582	0.0633

The detection efficiency as a function of energy for the HpGe Well detector was determined using the Canadian BL-3 uranium ore standard of 1.02% ²³⁸U and the IAEA RGU-1 uranium ore standard of 0.04% ²³⁸U. The self-absorption of 1001 keV gamma-ray in the BL-3 and RGU-1 uranium ore samples estimated to within 1.5–3%. However, the magnitudes of self-absorption of 46.5 and 63.3 keV by the samples were determined larger than that of 1001 keV gamma ray. Thus, the self-absorption factors for the 46.5 and 63.3 keV gamma rays by these samples calculated to be approx. 0.78 and 0.82, respectively, using a method described for the GWL series HPGe-well detectors. The measured photopeak efficiency data of the standards with two different uranium content for the HpGe detector used are given in Table 1. In determining photopeak efficiency values, the necessary correction factors for the self-absorption of the gamma-rays of interest has been taken into account. There was no remarkable systematic change on the measured counting efficiencies for the gamma-ray peaks of low intensities such as 46.5 keV (4%) of ²¹⁰Pb, 63.3 keV (4.5%) of ²³⁴Th and 1001 keV (0.835%) of ^{234m}Pa.

The certified and measured ²³⁸U activities in the CRMs are given in Table 2. It has been calculated that 2.966 µg natural uranium (99.2745% ²³⁸U) is equivalent to the activity of approx. 0.037 Bq. As seen in Table 2, the certified activity values in some uranium standards containing U₃O₈ can easily be calculated considering approx. 84.8% of U₃O₈ equals to uranium. The results obtained from the 1001 keV peak of ^{234m}Pa agreed to within 2–5% with the certified activities of CRMs shown Column

Table 2
Results of gamma-ray spectrometric measurements on Certified Reference Materials

No.	Standard code	Uranium	Certified uranium concentration (wt. %)	Certified ^{238}U activity* (Bq/g)	Measured ^{238}U (via ^{234}Th) activity ^{†,‡} (Bq/g)	Measured ^{238}U (via $^{234\text{m}}\text{Pa}$) activity ^{†,‡} (Bq/g)
0	OIEA-5 ^(a)	U_3O_8	0.014	1.466	1.855 ± 0.032	1.763 ± 0.062
1	BL-1 ^(b)	Nat. U	0.022	2.716	2.928 ± 0.097	2.879 ± 0.085
2	S-13 ^(a,i)	U_3O_8	0.039	4.083	4.132 ± 0.125	4.055 ± 0.181
3	RGU-1 ^(c)	Nat. U	0.040	4.939	5.404 ± 0.117	4.917 ± 0.098
4	OIEA-27 ^(a,h)	U_3O_8	0.140	14.658	16.230 ± 0.242	15.617 ± 1.025
5	BL-4 ^(b)	Nat. U	0.173	21.360	24.032 ± 0.597	23.169 ± 0.205
6	S-4 ^(d)	U_3O_8	0.375	39.263	37.691 ± 0.372	39.472 ± 1.284
7	S-3 ^(e)	U_3O_8	0.418	43.765	43.766 ± 0.249	43.413 ± 1.258
9	S-1 ^(f)	U_3O_8	0.471	49.314	50.251 ± 0.276	50.945 ± 0.235
10	OIEA-7 ^(a,g)	U_3O_8	0.514	53.790	57.762 ± 0.324	56.516 ± 1.047
8	BL-2 ^(b)	Nat. U	0.453	55.931	58.795 ± 0.236	57.097 ± 0.933
11	BL-3 ^(b)	Nat. U	1.020	125.938	126.731 ± 0.625	124.159 ± 1.118

*: Activity values calculated from the certified concentrations of CRMs (shown in Col. 3).

†: Corrected for self-absorption effects.

‡: Errors are based on counting statistics of $\pm 1\sigma$.

^(a)Prepared by Junta De Energia Nuclear (JDEN), Madrid, Spain.

^(b)Prepared by Canadian Centre for Mineral and Energy Technology, Ottawa, Ontario, Canada.

^(c)Uranium ore diluted with Silica prepared by IAEA Analytical Quality Control Service, Vienna, Austria.

^(d)Low Grade Uranium Ore: URANINITE (Origin: Australia).

^(e)Low Grade Uranium Ore: CARNOTITE (Origin: USA).

^(f)Low Grade Uranium Ore: TORBERNITE (Origin: Australia).

^(g)Mean value is calculated from the four certified values: 0.527%, 0.527%, 0.526% and 0.475%.

^(h)Mean value is calculated from the five certified values: 0.142%, 0.136%, 0.149%, 0.137% and 0.136%.

⁽ⁱ⁾IAEA Uranium Ore Standard (Pitchblende).

5 in Table 2. Also Fig. 3 shows that the measured and certified activities of CRMs agrees well. But the activities determined from the count rates of 63.3 keV peak of ^{234}Th are somewhat deviated from the certified activities of CRMs used. Thus, the ^{238}U (via ^{234}Th) activities have been determined to the average 8% error by about 14 h counting time in the present HpGe detector system. Although the results obtained from the 63.3 keV emission of ^{234}Th are corrected for the self-absorption effects, the activities determined from the count rates of the 63.3 keV (^{234}Th) are still found to be different from the certified activities of CRMs. It implies that most part of error for the 63.3 keV (^{234}Th) peak results arises from the contributions due to 63.9 keV (^{232}Th), 63.9 keV (^{231}Th) and 62.9 keV (^{234}Th) emissions. The certified uranium values for the standard (No. 4) are 0.142%, 0.149%,

0.137% and 0.136%, and so the mean value is calculated from the five certified uranium concentrations. Similarly, the certified uranium values for the standard (No. 10) are 0.527%, 0.527%, 0.526% and 0.475%, and so the mean value is calculated from the four certified values. It is possible that the measured activities for these samples may be around the mean value of the three high certified values.

The equilibrium of states of CRMs were also determined, assuming equilibrium activity between the ^{234}Th first daughter and the ^{238}U parent, by the ratio of activity between ^{234}Th and ^{210}Pb . As seen in Table 3, the equilibration between near and far product activity of ^{238}U decay chain is attained.

Two factors are important to the successful application of the gamma-ray spectrometric technique in measuring uranium concentrations in the

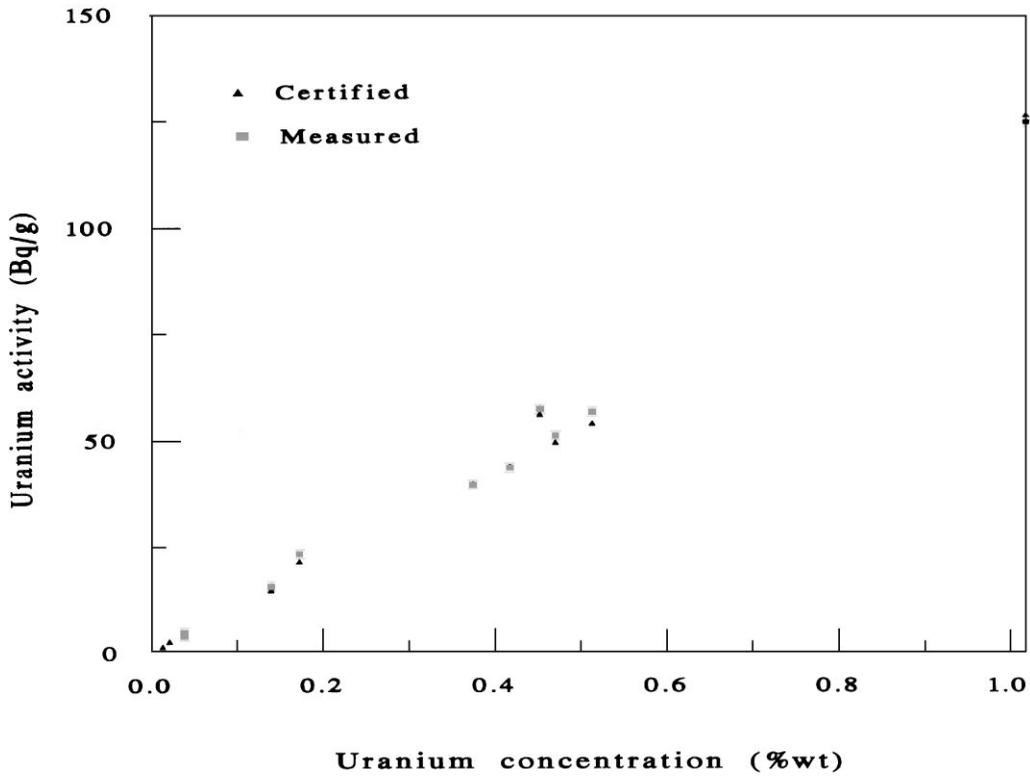


Fig. 3. Measured and certified uranium activities versus uranium concentrations of CRMs.

Table 3
Equilibrium states between near and far daughters of ^{238}U decay chain for CRMs

No.	Standard code	Certified uranium concentration (wt. %)	Measured ^{234}Th activity (Bq/g)	Measured ^{210}Pb activity (Bq/g)	Equilibrium $^{234}\text{Th}/^{210}\text{Pb}$
0	OIEA	0.014	1.855	2.117	0.876
1	BL-1	0.022	2.928	2.862	1.023
2	S-13	0.039	4.132	4.631	0.892
3	RGU-1	0.040	5.404	5.917	0.913
4	OIEA-27	0.140	16.230	16.465	0.985
5	BL-4	0.173	24.032	26.815	0.896
6	S-4	0.375	37.691	38.155	0.987
7	S-3	0.418	43.766	44.088	0.992
9	S-1	0.471	50.251	60.334	0.833
10	OIEA-7	0.514	57.762	58.443	0.988
8	BL-2	0.453	58.795	57.948	1.014
11	BL-3	1.020	126.731	127.058	0.997

samples. First, the gamma-counting statistics principally define the overall detection limit of the method and the accuracy of final values. Second, in the low-energy gamma-ray spectrometric measure-

ments such as using 63.3 and 93 keV, the self-absorption of gamma-rays by the sample matrix causes a fluctuation of counting efficiency. Therefore, there is a need to correct on the measurements

for the self-absorption. A simple mathematical model for estimating sample self-absorption factors for well-type germanium detectors is described in detail [16]. In this model, self-absorption factors are expressed in terms mass attenuation coefficient of the sample and a parameter characterising the well geometry. If L is the length of the sample and $m = \pi\rho a^2 L$ is the sample mass, the emission ratio I/I_0 is given by the formula

$$I/I_0 = f(m) = 2e^{-k\mu m} \left\{ \frac{1}{k\mu m} \sinh(k\mu m) - \frac{1}{(k\mu m)^2} (\cosh(k\mu m) - 1) \right\}, \quad (1)$$

where

$$k = 1/\pi a L \quad (2)$$

is a geometric parameter characterising the dimensions of the sample. μ is mass attenuation coefficient (cm^2/g) of the sample, a is the radius of sample, I_0 is the emission (neglecting absorption) from the whole of the sample.

Once k has been determined, for a given material attenuation coefficient μ (cm^2/g) the emission ratio is then just a function $f(m)$ of the sample mass. Thus, self-absorption factors for the CRMs are calculated using Eq. (1) with aid of a calculator or a computer. In our case, the radius of sample (a) is 0.7 cm, the sample height (L) is 4.4 cm, and the weight of each of CRMs used is known. Further, the value of the total mass attenuation coefficient for each sample must be determined. For this, two examples in Table 4 are presented for the calculations of the attenuation coefficients of gamma-rays of interest in components of CRMs by using literature data [17]. Then, in the case of samples of known composition it is possible to calculate the mass attenuation coefficient μ (cm^2/g) of the sample at a fixed gamma-ray energy as

$$\mu = \sum_i w_i \mu_i, \quad (3)$$

where w_i is the weight fraction of i th component. After inserting the value of the $k\mu m$ for a particular gamma-energy in Eq. (1), the self absorption factor is calculated with the help of a microcomputer. The calculated self-absorption factors for the 46.5, 63.3

Table 4

Mass attenuation coefficients for 46.5, 63.3 and 1001 keV gamma-rays in components of two CRMs with weight percents

Compo- sition	Weight (%)		Mass attenuation coefficient ^a ($\text{cm}^2 \text{g}^{-1}$)		
	S-1	S-3	46.5 keV	63.3 keV	1001 keV
Al ₂ O ₃	6.400	6.000	0.3373	0.2192	0.0625
B ₂ O ₃	0.040	0.003	0.2102	0.1741	0.0622
BaO	0.030	0.040	15.7541	6.2977	0.0586
CaO	0.060	0.550	0.9884	0.4507	0.0639
Cr ₂ O ₃	0.010	0.004	1.4366	0.6124	0.0606
CuO	0.040	0.020	2.7433	1.0841	0.0599
Fe ₂ O ₃	3.600	0.500	1.8828	0.7345	0.0611
Ga ₂ O ₃	0.002	–	3.0161	1.1873	0.0592
K ₂ O	1.100	2.500	0.9469	0.4333	0.0624
Li ₂ O	0.006	–	0.1922	0.1634	0.0597
MgO	0.390	1.200	0.3207	0.2147	0.0633
MnO	0.030	0.090	1.7599	0.7220	0.0597
Na ₂ O	0.100	1.050	0.2954	0.2033	0.0617
NiO	0.002	–	2.6066	1.0269	0.0620
P ₂ O ₅	0.700	0.100	0.3893	0.2379	0.0629
PbO	0.020	–	9.5341	3.8765	0.0705
SiO ₂	85.000	80.000	0.3672	0.2315	0.0637
SrO	0.040	0.040	5.0840	1.9674	0.0624
ThO ₂	10 ppm	4 ppm	12.2523	5.0368	0.0756
TiO ₂	0.500	0.330	1.0209	0.4585	0.0608
U ₃ O ₈	0.471	0.418	12.0992	4.9743	0.0766
V ₂ O ₅	0.030	0.580	1.0632	0.4712	0.0612
ZnO	–	0.040	3.0484	1.1980	0.0603

^aAbsorption of gamma-rays of interest in components of the CRMs estimated by using literature data [17].

and 1001 keV gamma-rays by the two CRMs having different sample matrix compositions are given in Table 5. The self-absorption of the 1001 keV emission did not change strongly by the sample matrix compositions. The magnitudes of the necessary corrections for self-absorption on the 1001 keV peak data are very small and they are in order of 5%. In other words, in gamma-ray spectrometric uranium measurements, only about 5% of uranium concentration in any sample in HPGe well detector might be ignored by use of the 1001 keV peak of ^{234m}Pa without any self-absorption correction. In case of the using 63.3 keV peak of ²³⁴Th, the necessary self-absorption correction in CRMs is in order of 20–23%. The self-absorption factors for the 63.3 keV peak of ²³⁴Th affected from the matrix compositions of the CRMs. The

Table 5

Self-absorption factors for 46.5, 63.3 and 1001 keV gamma-rays by the two CRMs

Factors	S-1			S-3		
	46.5 keV	63.3 keV	1001 keV	46.5 keV	63.3 keV	1001 keV
k	0.103	0.103	0.103	0.103	0.103	0.103
m	7.919	7.919	7.919	8.379	8.379	8.379
μ	0.490	0.275	0.063	0.431	0.249	0.059
$k\mu m$	0.400	0.224	0.051	0.372	0.215	0.051
f	0.697	0.809	0.951	0.713	0.815	0.951

Note: k , geometric parameter (cm^{-2});

m , sample weight (g);

μ , mass attenuation coefficient of sample ($\text{cm}^2 \text{g}^{-1}$);

$k\mu m$, dimensionless parameter inserted in Eq. (1);

f , self-absorption factor for a given gamma-ray energy.

variations in self-absorption factors for the 63.3 keV peak of ^{234}Th estimated to within 2–4%. Consequently, the self-absorption correction is satisfactorily applied to the measurements in HPGe well detector using the above procedure but the main difficulty is that mass attenuation coefficient of any sample must be known. Clearly, the gamma-ray spectrometry using these corrections for the low-energy peaks such as 63.3 keV (^{234}Th) might be more complicated than alpha spectrometry. The problem of self absorption can be minimised by making the sample as thin as possible [18]. However, the detection limit becomes worse as the sample becomes thinner.

The results of the gamma-ray spectrometric measurements (corrected for self absorption effects due to the 1001 keV peak occurring in the high-energy region) on CRMs, given in Table 2, show that high-energy gamma-ray spectrometry can give more reliable ^{238}U measurements on the sample of different thorium and uranium contents. However, the samples containing low uranium contents should be counted longer periods in order to improve counting statistics. When the measurements of uranium in the samples are carried out by using HPGe detectors with high crystal volume, the accurate measurements of uranium in the samples by use of 1001 keV peak of $^{234\text{m}}\text{Pa}$ can be achieved even if the sample size is small. Thus the present measurements were done by using small thicknesses of samples to minimize the self absorption

effects. It is worth noting that the 1001 keV peak of $^{234\text{m}}\text{Pa}$ daughter of ^{238}U is the most clean and suitable gamma emission for the analytical purposes because approx. 0.9987 of the $^{234\text{m}}\text{Pa}$ beta-decay feeds the ground state of ^{234}U directly, the remaining beta branches, together with any gamma-rays and conversion lines are weak [15]. In addition, the 766 keV (0.207%) gamma-ray of $^{234\text{m}}\text{Pa}$ cannot be considered useful for the measurement of ^{238}U (via $^{234\text{m}}\text{Pa}$). Because the 766 keV emission of $^{234\text{m}}\text{Pa}$ interferes with the 768 keV (4.76%) emission from the ^{214}Bi of distant member of ^{238}U . In case of the utility of this peak data, the analyst is faced with the problem of the deconvolution of the 766 keV peak area from the 768 keV peak area.

4. Conclusions

High-energy gamma-ray spectrometry by using 1001 keV peak of $^{234\text{m}}\text{Pa}$ second daughter with very short half-life (1.17 m) of ^{238}U has been shown to be capable of providing reliable measurements for samples containing different uranium contents. The measurement of 1001 keV emission from $^{234\text{m}}\text{Pa}$ does not require any self-absorption corrections on the results since it is high-energy gamma-ray. Although the emission probability of the 1001 keV gamma-ray is low, this peak does not have any interference with other peaks in the

uranium spectra even if the samples contain high amounts of thorium. Consequently, this “clean” peak is useful to determine the ^{238}U concentrations in the samples by HpGe gamma-ray spectrometry.

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