

# A GAMMA SPECTROMETRIC METHOD FOR DIRECT MEASUREMENT OF URANIUM CONTENTS IN SOLID SAMPLES

H. YÜCEL\*, I. ERGIN\*\*, M. A.ÇETINER\*, H. DEMIREL\* ,  
M. TAN\*\* and A. ÖZMEN\*\*\*

\* Ankara Nuclear Research and Training Center, 06100 Besevler-Ankara

\*\* Gazi University, Education Faculty, Physics Dept., Teknikokullar-Ankara

\*\*\* Gazi University, Arts and Science Faculty, Physics Dept., Teknikokullar- Ankara

## ABSTRACT

A gamma spectrometric method is described for the direct “absolute” determination of uranium contents in solid samples. To obtain the exact information about the measurement of  $^{238}\text{U}$  in samples by gamma-ray spectroscopy, it is essential that any one of the daughters of  $^{238}\text{U}$  should exist in radioactive equilibrium with  $^{238}\text{U}$ . This condition is mainly fulfilled by one of the first two daughters, namely,  $^{234}\text{Th}$  (24.1 d) and  $^{234\text{m}}\text{Pa}$  (1.17 min) because these daughters are very short-lived compared with  $^{238}\text{U}$ . Therefore, the present analytical method employs direct determination of  $^{238}\text{U}$  via the 63.3 keV gamma emission from  $^{234}\text{Th}$  and via the 1001 keV gamma emission from  $^{234\text{m}}\text{Pa}$ .

The absolute photopeak efficiency data in the range of 40 keV to 2000 keV for the reverse-electrode Germanium(REFe) detector used is measured using a powder radioactive standard source. The certified uranium samples were prepared in thin thicknesses to minimize the self-absorption of gamma rays. They are counted for the time predetermined between 75,000 s and 125,000 s. The measured activities of  $^{238}\text{U}$  of the samples via the 63.3 keV ( $^{234}\text{Th}$ ) and via the 1001 keV ( $^{234\text{m}}\text{Pa}$ ) are corrected for self-absorption effects due to changes in composition of standard reference materials in a given sample-detector geometry. The self-absorption factors applied to the 46.5 keV ( $^{210}\text{Pb}$ ) peak and the 63.3 keV ( $^{234}\text{Th}$ ) peak results ranged from 10.5% to 33.1% and from 4.9% to 18.4%, respectively. For the 1001 keV ( $^{234\text{m}}\text{Pa}$ ) gamma emission, the necessary self-absorption factors varied between 0.1% and 3.9%.

The results obtained from the 46.5 keV of  $^{210}\text{Pb}$  being far distant member of  $^{238}\text{U}$  and those of the 63.3 keV from  $^{234}\text{Th}$  are used to check whether the equilibrium exist in the uranium samples. The results of  $^{238}\text{U}$  via the 63.3 keV peak of  $^{234}\text{Th}$  for the uranium standards agreed to within 0.1 to 6.9% with the certified values of  $^{238}\text{U}$  in the samples with uranium concentration ranging from  $0.022\pm 0.001$  to  $1.02\pm 0.01$  wt%. The results of  $^{238}\text{U}$  via the 1001 keV peak of  $^{234\text{m}}\text{Pa}$  agreed well to within 0.01 to 4.7% with the certified activity values of  $^{238}\text{U}$  of the same samples. When the measurements of the uranium are carried out in a REFe detector system, the results indicate that the uranium concentrations in the samples can be determined to the average 7% error by using the 63.3 keV ( $^{234}\text{Th}$ ) gamma emission with appropriate corrections for self-

absorption effects. The 1001 keV peak of  $^{234m}\text{Pa}$  gives also accurate results for the determination of  $^{238}\text{U}$  in the same samples and the need for self absorption correction for the 1001 keV emission seems to be negligible, except in case of thicker samples than that of 1.5 cm thick.

## Introduction

The major source of radiation exposure to man is the naturally occurring radionuclides in the environment (terrestrial radiation) and direct cosmic(extra-terrestrial) radiation. It is known that one of the most obvious sources of radionuclides in the environment is nature itself. In addition, some sources of natural radiation have been enhanced by human technological activities and include wastes from mineral ores and petroleum industry, sludge, and the articles made from naturally occurring radioactive materials such as thorium in lantern mantels. Therefore, the sources of greatest interest in most environmental studies are primordial nuclides in geosphere. Geological formations and soils may contain isotopes of uranium, thorium, radium, radon and other radioactive elements. Particularly, in areas of igneous geology, concentrations of uranium and thorium series nuclides as well as naturally occurring potassium are significant. In recent years, a need has arisen to find and produce materials especially low in radioactive primordial elements' thorium, uranium and potassium[1].

For the measurement of uranium series nuclide concentrations, radiochemical analysis followed by alpha spectrometry is most commonly used[2,3]. 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. Therefore, the high-resolution gamma spectrometry has been extensively used for the quantitative analysis of the radionuclides such as uranium and thorium in samples[4, 5]. If the analyst wishes to measure the  $^{238}\text{U}$  content of a sample independent of the  $^{235}\text{U}$  content, one is faced with the problem that the  $^{238}\text{U}$  isotope emits directly only very weak gamma radiation which is at 49.55 keV(0.064%). As is seen in decay scheme of  $^{238}\text{U}$  in Fig.1, since other direct gamma ray of 113.5 keV from  $^{238}\text{U}$  has also very low emission probability(0.0102%) , these direct emissions can not be considered the measurable gamma lines in the present detectors.

However, more strong gamma rays emitted from a daughter nuclide of the  $^{238}\text{U}$  parent should be used to obtain the accurate measurement of uranium contained in samples. In the previous work, authors investigated the possible gamma rays from the daughter nuclides in secular equilibrium with the  $^{238}\text{U}$  and the possible interference emissions to the gamma ray used in the uranium analysis by gamma-ray spectrometry[7].

In the absolute gamma-ray spectrometric measurements of the  $^{238}\text{U}$  by using NaI(Tl) crystals, all earlier works which used the gamma rays of 352 keV from  $^{214}\text{Pb}$  and of 609 keV from  $^{214}\text{Bi}$  of the distant members by assuming secular equilibrium between  $^{238}\text{U}$  and  $^{226}\text{Ra}$  or  $^{214}\text{Bi}$ . This assumption, however, cannot be considered valid in many situations where the disequilibrium

conditions between  $^{238}\text{U}$  and its daughters can occur due to difference in their geochemical behaviour. Nevertheless, the new methods for the simultaneous analysis  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{210}\text{Pb}$  and other decay products of  $^{238}\text{U}$  in water, sludge, sediments, soil, coal, ash and other types of samples are still developed and applied by scientists using scintillation spectrometry with NaI(Tl) detector[8,9]. Alternatively, since the accuracy has been greatly improved with introduction of the high-resolution gamma spectrometry with the high pure Ge detectors, it is possible to the  $^{238}\text{U}$  contents in samples by analysing simultaneously both the low and the high energy rays of the uranium decay products. In the absence of any directly measurable gamma ray from  $^{238}\text{U}$  itself and in cases where the assumption of the secular radioactive equilibrium in the  $^{238}\text{U}$  is not permissible. To obtain the exact information about the measurement of  $^{238}\text{U}$  by gamma-ray spectroscopy, it is essential that any one of daughters of  $^{238}\text{U}$  should exist in equilibrium with  $^{238}\text{U}$ . This condition is fulfilled by one of the first three daughters of  $^{238}\text{U}$ , namely  $^{234}\text{Th}$  (24.1d),  $^{234\text{m}}\text{Pa}$  (1.17m) and  $^{234}\text{Pa}$  (6.78h). These first three daughters have very short lives compared with the life of  $^{238}\text{U}$  parent. Thus, the radioactive equilibrium is quickly established. For example,  $^{234}\text{Th}$  is the first daughter of  $^{238}\text{U}$  and has a relatively short half-life of 24.1 d, it ensures that the radioactive equilibrium with  $^{238}\text{U}$  is attained about 160 d. The gamma peaks of 63.3 keV and about 93 keV of  $^{234}\text{Th}$  have been considered useful for analytical purposes in earlier works [5,10]. In fact that the conventional NaI(Tl) crystals are incapable of resolving these emissions from other gamma and X-ray peaks in the low energy region, however, the low energy photon spectrometers(LEPS) with high pure Germanium detectors have high efficiencies and very capable of resolving the peaks in the low energy region(< 120 keV). In this case, the analyst is faced with another problem, that the 93 keV peak of  $^{234}\text{Th}$  is actually a doublet. The 93 keV gamma emission of  $^{234}\text{Th}$  consists of 92.370 keV(2.61%) and 92.793 keV(2.58%) [11]. 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]. Although the 63.3 keV peak also includes contributions from the 63.9 keV (0.255%) emission from  $^{232}\text{Th}$ , the 63.9 keV (0.023%) emission from  $^{231}\text{Th}$ ( $^{235}\text{U}$  series) and the 62.9 keV (0.018%) emission from  $^{234}\text{Th}$ [12], the 63.3 keV(4.5%) of  $^{234}\text{Th}$  gamma emission is often a preferable photopeak for uranium analysis, especially when samples are counted by using LEPS with Ge detectors.

The first aim of the study is to represent that an analytical method for direct determination of  $^{238}\text{U}$  in the solid samples can be employed via the 63.3 keV gamma emission from  $^{234}\text{Th}$  and via the 1001 keV gamma emission from  $^{234\text{m}}\text{Pa}$ , and the second aim is to calculate the self-absorption correction factors to be applied for these gamma-emissions.

## Experimental Procedure

The commercially available REGe detector with its own preamplifier was used in the measurements of uranium concentrations in samples. The detector was a n-type high pure Ge with 0.5 mm thick beryllium window and it has high efficiencies for low energy gamma rays (<120 keV). The active volume of REGe crystal is about 100 cm<sup>3</sup>. The REGe detector has a relative efficiency of 20 %. The measured resolution of the REGe detector was 1.80 keV for the 1332.5 keV and 0.97 keV for 122 keV. The detector was shielded by 10 cm lead lined with 1 mm thick copper. In the REGe detector system, the preamplified signal is processed through a spectroscopy amplifier and a multichannel analyser (Model 95+ CANBERRA).

Most of the certified uranium samples were stored in our laboratory for about 15 years and it is therefore expected to be attained the equilibrium between <sup>238</sup>U and its near daughters such as <sup>234</sup>Th and <sup>234m</sup>Pm. Only a small amount, about 8 g of powdered, the uranium samples filled into the polystyrene tubes 1.4 cm inside diameter and 1 mm in wall thickness. The samples were chosen as thin as possible to minimise the self-absorption effect for especially low energy gamma rays. The samples were prepared to almost same height to ensure identical counting geometry. The densities of powdered CRMs used were in the range of 0.92 – 1.11g/cm<sup>3</sup>. Then each the sample is weighed with a balance having a sensitivity down to mg range. Finally, each of samples is counted for the time predetermined between 75,000 s and 125,000 s.

The counting time for each measurement is high enough to ensure good statistical quality of data. The measurements for each sample were repeated at least three times to improve the statistical precision. Thus the mean values of the measured count rates were used in the calculations. In peak analysis procedure, since the series 95 MCA has advanced functions, the full-energy peak areas are determined by applying automatic computation. Nevertheless, some full-energy peak areas are often checked by manual selection. The areas determined by two ways did not differ more than 0.5%.

Also the 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. After laboratory background subtraction of the peak area of interest, the net sample count rate was obtained.

## Results and Discussion

The detection efficiency as a function of energy for the REGe detector was determined using powder multinuclide standard source obtained from Isotope Product Labs Inc. The measured absolute photopeak efficiency data given in Table 1 are fitted best to a mathematical function in the the following form:

$$\epsilon(E_{\gamma}) = a - b \cdot \exp(-c \cdot E_{\gamma}^d) \quad (1)$$

where,

$\epsilon$  is the absolute photopeak efficiency for a particular energy,  $E_\gamma$ . and a,b,c and d are constants. In the low energy region(defined as about 40 to 120 keV), the four experimental efficiency values were used to determine the efficiency curve of REGe. Two of them are that the efficiencies of 46.5 keV and 63.3 keV photopeak are determined from the count rates of the certified reference materials coded as S-1 and BL-3. As well, the other two efficiency values which are 88.03 keV( $^{109}\text{Cd}$ ) and the 122.06 keV( $^{57}\text{Co}$ ) peaks were determined from the multinuclide reference calibration source. In whole energy range of about 40 to 2000 keV, there was no remarkable systematic change on the measured counting efficiencies for the gamma-ray peaks with low intensities such as 46.5 keV(4.2%) of  $^{210}\text{Pb}$ , 63.3 keV(4.5%) of  $^{234}\text{Th}$  and 1001 keV(0.992%) of  $^{234\text{m}}\text{Pa}$ .

The certified and measured  $^{238}\text{U}$  activities in the CRMs are given in Table 2. It has been calculated that approx. 81  $\mu\text{g}$  natural uranium(99.2745%  $^{238}\text{U}$ ) is equivalent to the activity of 1 Bq. As is seen in Table 2, the certified activity values in some uranium standards containing  $\text{U}_3\text{O}_8$  can be calculated considering approx. 84.8% of  $\text{U}_3\text{O}_8$  equals to uranium.

The measurements of uranium in the samples carried out by using a REGe detector indicate that the counting statistics of the 63.3 keV gamma emission from  $^{234}\text{Th}$  is very good for the analysis because of high detection efficiency in the low-energy region. However, the activity results measured via 63.3 keV peak in Table 2 are always lower than the certified activities of CRMs. Therefore, the order of the necessary self-absorption correction for the 63.3 keV peak from  $^{234}\text{Th}$  must be taken into account. Whereas, the 1001 keV peak of  $^{234\text{m}}\text{Pa}$  in the high-energy region is the most suitable peak for the determination of uranium in samples. 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 and the necessary correction for self-absorption effects applied to this peak is very small and may be negligible[7]. But, the main disadvantage of the 1001 keV peak of  $^{234\text{m}}\text{Pa}$  is that the absolute gamma emission probability is very low, for example, the newly recommended value of 0.992%[13]. Therefore the data acquisition for the samples containing low uranium requires longer times in order to obtain good counting statistics by using the 1001 keV emission.

On condition that the sample (or source) shape and detector distance shown in Fig.2 remain constant, the necessary self absorption due to the changes in composition of the certified uranium sample is corrected for the measured activity as follows:

$$A_c = A_0 \cdot \frac{\mu km}{1 - e^{-\mu km}} \quad (2)$$

where,  $A_c$ : the corrected activity for a self absorption for a particular gamma ray,  $A_0$ : the measured activity,  $\mu$ : mass attenuation coefficient( $\text{cm}^2 \cdot \text{g}$ ),  $k$ : a geometrical parameter( $\text{cm}^{-2}$ ) at a

given counting geometry and  $m$ : sample mass(g). The procedure for the calculation of a self absorption factor is described in the sub-section

The measured activities with a suitable correction for self absorption(according to Eq. 2) and the certified activities of CRMs are shown in Table 3. Then, the results obtained from the 63.3 keV peak of  $^{234}\text{Th}$  are compared with those of 1001 keV peak from  $^{234\text{m}}\text{Pa}$  using the same certified reference materials (CRM). The results of  $^{238}\text{U}$  via the 63.3 keV peak of  $^{234}\text{Th}$  for the uranium standards agreed to within 0.1 to 6.9% with the certified values of  $^{238}\text{U}$  in the samples with uranium concentration ranging from  $0.022\pm 0.01$  to  $1.02\pm 0.01$  wt%. The results of  $^{238}\text{U}$  via the 1001 keV peak of  $^{234\text{m}}\text{Pa}$  agreed well to within 0.01 to 4.7% with the certified activity values of  $^{238}\text{U}$  of the same samples. That is, the results indicate that the uranium concentrations in the samples can be determined to the maximum 7% error by using the 63.3 keV( $^{234}\text{Th}$ ) gamma emission with appropriate corrections for self-absorption effects in a REGe detector system. The self-absorption factors applied to the 63.3 keV( $^{234}\text{Th}$ ) peak results ranged from 4.9% to 18.4%. In addition, the 1001 keV peak of  $^{234\text{m}}\text{Pa}$  gives also accurate results for the determination of  $^{238}\text{U}$  in the same samples. For the gamma emission of 1001 keV, the necessary correction for self absorption due to changes in sample matrix varied between 0.1% and 3.9%. Thus, the measured(corrected for self absorption) and the certified uranium activities versus uranium concentrations of CRM samples are plotted in Fig. 3 and Fig. 4 for 63.3 keV and 1001 keV peak, respectively. These calibration curves show that the measured (corrected for self absorption) and certified activities of CRMs agree well. Nevertheless, the activities determined from the count rates of 63.3 keV peak of  $^{234}\text{Th}$  are somewhat deviated from the certified activities of CRM samples. It implies that most of error for the 63.3 keV( $^{234}\text{Th}$ ) peak results may arise from the contributions due to 63.9 keV( $^{232}\text{Th}$ ), 63.9 keV( $^{231}\text{Th}$ ) and 62.9 keV( $^{234}\text{Th}$ ) emissions. Thus, the  $^{238}\text{U}$ (via  $^{234}\text{Th}$ ) activities have been determined to the maximum 7% error.

The certified uranium values for the standard sample(No.4) are 0.142%, 0.136% 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 sample (No.10) are 0.527%, 0.527%, 0.526% and 0.475%, and so the mean value is calculated from the four certified values. Therefore, the measured activities for these samples(No.4 and No.10) have found to 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  $^{234}\text{Th}$  first daughter and the  $^{238}\text{U}$  parent, by the ratio of activity between  $^{234}\text{Th}$  and  $^{210}\text{Pb}$ . The activities determined from the count rates of the 46.5 keV( $^{210}\text{Pb}$ ) peak were also corrected by the self-absorption factors which are order of 10.5% to 33.1%. As is seen in Table 3, the equilibration between near and far product activity of  $^{238}\text{U}$  decay chain is attained.

Two factors are important to the successful application of the gamma-ray spectrometric technique in measuring uranium concentrations in the samples. First, the gamma counting

statistics principally define the overall detection limit of the method and the accuracy of final values. Second, the self absorption of gamma rays by the sample matrix causes a fluctuation of counting efficiency. An effort was made to correct the measurement results for self-absorption of gamma emissions by the following procedure.

### Self Absorption Correction Procedure

The self absorption of radiation by the sample is relatively difficult to correct because the attenuation coefficients for gamma rays are highly dependent on sample composition. Especially, in the low-energy gamma ray spectrometric measurements such as using 46.5 keV , 63.3 keV or 93 keV, there is a need to correct on the measurements for the self-absorption. A theoretical consideration is used to estimate the self-absorption effect of a sample matrix in the present gamma spectrometric measurements. Referring to Fig.2, the attenuation gamma rays emitted from a sample volume element and travelling a perpendicular element path length  $h$  to the detector window face is simply defined as follows[14]

$$\frac{I(h)}{I_0(h)} = e^{-\mu \cdot h} \quad (3)$$

Total attenuation for a homogeneous sample of thickness  $x$  is then just

$$\frac{I}{I_0} = \frac{1}{x} \int_0^x e^{-\mu \cdot h} \cdot dh \quad (4)$$

and thus the integration of Eq. 4 yields the familiar self-absorption equation

$$\frac{I}{I_0} = \frac{1 - e^{-\mu x}}{\mu x} \quad (5)$$

where,  $I_0$  is the emission intensity(neglecting absorption) from the whole of the sample and  $I$  is the intensity of the emergent rays from the sample (or attenuated beam).  $\mu(\text{cm}^{-1})$  is linear attenuation coefficient for sample matrix and  $x(\text{cm})$  is the mean path length of a gamma ray for a particular energy. The value of  $\mu$  can be calculated from the product of the density and the mass attenuation coefficient ( $\mu/\rho$ ), which can be obtained through a knowledge of the sample's elemental composition.

For a sample that is not more than a few cm thick, the mean path length,  $x$  is taken to be 3/4 times the sample thickness by assuming that 50% of gamma emissions travel a path length being equal to the half of sample thickness( $r$ ) and the 50% of emissions travel the sample thickness( $2r$ ). That is,  $x = \frac{(r + 2r)}{2}$ . Thus, the mean path length( $x$ ) equals to approximately 1.5

times the mean chord length of a sample tube in cylindrical shape. The mean chord which can usually be defined as  $2V/S$ , where  $V$  is cylinder's volume and  $S$  is surface area, is equal to the

radius  $r$  of the cylinder when the radius is much less than the length of the sample ( $L$ ). Then, the self-absorption factor for a particular energy can be expressed in terms mass attenuation coefficient of the sample and a parameter characterising the sample-detector geometry. If  $L$  is the length of the sample cylindrical in shape and the sample mass is  $m = (\pi r^2 L)\rho$ , the gamma emission ratio  $I/I_0$  given in Eq. 5 can be rewritten depending on the sample mass:

$$I/I_0 = \frac{1 - e^{-\mu \rho x}}{\mu \rho x}$$

or

$$f(m) = \frac{1 - e^{-\mu k m}}{\mu k m} \tag{6}$$

where

$$k = \frac{3}{2\pi r L} \tag{7}$$

is a geometric parameter characterising the dimensions of the sample.  $\mu$  is mass absorption coefficient ( $\text{cm}^2/\text{g}$ ) of the sample,  $r$  is the radius of sample. Once  $k$  has been determined, for a given material attenuation coefficient  $\mu$  ( $\text{cm}^2/\text{g}$ ) 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.(6). The inner radius of sample tube ( $r$ ) is 0.7 cm, the sample height ( $L$ ) is about 4.4 cm, and the weight of each of CRMs used is known. The value of the total mass attenuation coefficient for each sample should be determined. Four examples in Table 4 through Table 7 are presented for the calculations of the attenuation coefficients of gamma rays of interest in components of CRMs by using literature data[15]. For the samples of known composition it is possible to calculate the mass attenuation coefficient  $\mu$  ( $\text{cm}^2/\text{g}$ ) of the sample at a given gamma-ray energy as:

$$\mu = \sum_i w_i \cdot \mu_i \tag{8}$$

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. 6, the self absorption factor has been calculated with the help of a microcomputer. For example, the calculated self-absorption factors for the 46.5 keV, 63.3 keV and 1001 keV gamma rays by the four CRMs having different sample matrix compositions are given in Table 5. Finally, the self-absorption factors are used to correct the measured activities of the samples to determine accurately the sample uranium contents when using especially low energy rays such as 46.5 and 63.3 keV photopeaks are used in the analysis.

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 maximum 4%. In other words, in gamma-ray

spectrometric uranium measurements, only about 4% of uranium concentration in any sample in REGe detector might be ignored by use of the 1001 keV peak of  $^{234m}\text{Pa}$  without any self-absorption correction. Whereas, in the 63.3 keV peak of  $^{234}\text{Th}$  results, the necessary self-absorption correction in CRMs is in order of 4.9% to 18.4%. Consequently, the self-absorption correction is satisfactorily applied to the measurements in REGe 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 energies peaks such as 63.3 keV ( $^{234}\text{Th}$ ) might be more complicated than alpha spectrometry. The problem of self absorption can be minimised by making the sample as thin as possible. However, the detection limit becomes worse as the sample becomes thinner. The detection limit of gamma spectrometry is also important for samples containing very low uranium contents. The detection limit can be calculated by using the efficiency data and the background counts of the 63.3 peak region from the following equation [16].

$$D = \frac{A}{\epsilon \cdot P \cdot t \cdot m} \left( A + \sqrt{2B + \frac{A^2}{4}} \right) \quad (9)$$

where, D is the detection limit(Bq/g),  $\epsilon$  is photopeak efficiency, P is the gamma emission rate, t is counting time in seconds, m is sample weight(g), B is background counts and A is the reciprocal of relative counting error. If A, m and t are set to be 3, 6.5 g and 125000 s( about 35 h), the detection limit is calculated to be around 0.0037 Bq/g for  $^{238}\text{U}$ (via  $^{234}\text{Th}$ ).

The results of the gamma-ray spectrometric measurements via the 1001 keV peak (occurring in the high energy region) on CRMs that are given in Table 2 (without correction for self absorption ) and given in Table 3(corrected for self absorption) show that this gamma peak can also give reliable  $^{238}\text{U}$  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. In this case, the analyst may choose the 63.3 keV peak to avoid the longer counting times but it must be taken into account the self-absorption effects when the measurements of uranium in the samples are carried out by using LEPS or a REGe detector with high efficiencies in the low energy region. Thus the present measurements were done by using samples in thin thickness to minimise the self absorption effects. It is worth noting that the 63.3 keV peak of  $^{234}\text{Th}$  and 1001 keV peak of  $^{234m}\text{Pa}$  daughter of  $^{238}\text{U}$  is the most suitable gamma emissions for the analytical purposes.

## Conclusions

Since REGe detectors with Be-window have high efficiencies in the low energy region, the 63.3 keV gamma emission from  $^{234}\text{Th}$  gives an useful peak to obtain the reliable uranium measurements of samples. But, the self-absorption caused by sample matrix for the 63.3 keV peak must be taken into account. The problem of self absorption can be minimised by making the sample as thin as possible. However, the detection limit becomes worse as the sample becomes thinner. In high energy region, the 1001 keV peak of  $^{234\text{m}}\text{Pa}$  of  $^{238}\text{U}$  allows the analyst provide reliable measurements for samples containing especially high uranium contents. The measurement of 1001 keV emission from  $^{234\text{m}}\text{Pa}$  requires usually longer counting times because of its low emission probability. Nevertheless, it is still an useful peak especially samples containing high amounts of thorium with uranium. In addition the self absorption corrections on the results of 1001 keV peak is negligible. For the absolute or direct gamma-ray spectrometric measurements of uranium concentrations in the samples, the present method suggest that the 63.3 keV emission from  $^{234}\text{Th}$  and the 1001 keV emission from  $^{234\text{m}}\text{Pa}$  can be simultaneously used for  $^{238}\text{U}$  analysis, especially when the measurements are carried out by a REGe detector with being capable of well resolving low energy emissions. The results obtained from two peaks for the same sample is finally compared with each other in order to check the analysis results. This method can easily employ the soil samples and the samples collected from mine ores or other samples with very low uranium contents. Consequently, the method can apply successfully to samples varying from about 0.3 ppm  $^{238}\text{U}$  content (equivalent activity of 0.0037 Bq/g) upward.

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**Table 1.** The measured photopeak efficiencies for the REGe detector using multinuclide standard powder source and the standard uranium samples coded as S-1(0.471%U<sub>3</sub>O<sub>8</sub>) and BL-3(1.020% nat.U).

Radionuclide	Gamma ray Energy (keV)	Photopeak Efficiency	
		Measured	Calculated
<sup>210</sup> Pb	46,50	0,174320 ± 0,00141	0,173982
<sup>234</sup> Th	63,30	0,167688 ± 0,00673	0,168898
<sup>109</sup> Cd	88,03	0,161254 ± 0,00105	0,160354
<sup>57</sup> Co	122,06	0,147355 ± 0,00115	0,144796
<sup>57</sup> Co	136,48	0,139805 ± 0,00292	0,136417
<sup>123m</sup> Te	159,00	0,119938 ± 0,00063	0,123699
<sup>113</sup> Sn	391,69	0,050898 ± 0,00027	0,052293
<sup>85</sup> Sr	513,99	0,037863 ± 0,00027	0,038347
<sup>137</sup> Cs	661,66	0,029950 ± 0,00005	0,028653
<sup>88</sup> Y	898,02	0,020472 ± 0,00008	0,020292
<sup>60</sup> Co	1173,24	0,015888 ± 0,00002	0,015248
<sup>60</sup> Co	1332,50	0,014092 ± 0,00001	0,013412
<sup>88</sup> Y	1836,01	0,010420 ± 0,00006	0,009988
Whole energy range (40-2000 keV)		For <u>Low Energy Region</u> ( <u>&lt; 120 keV</u> )	
R=0,999455		r = 0,999999	
A=0,171667		a = 0,179637	
B=0,167139		b = 0,516837	
C=1952,3389		c = 30,40561	
D= -1,449356		d = -0,496732	

**Table 2.** Results of gamma-ray spectrometric measurements on certified reference materials (Not corrected for self absorption)

No.	Standard Code	Uranium	Certified Uranium Concentration (wt. %)	Certified $^{238}\text{U}$ activity* (Bq/g)	Measured $^{238}\text{U}$ (via 63.3 keV from $^{234}\text{Th}$ ) activity* (Bq/g)	Measured $^{238}\text{U}$ (via 1001 keV from $^{234\text{m}}\text{Pa}$ ) activity* (Bq/g)	Measured $^{210}\text{Pb}$ (via 46.5 keV peak) activity* (Bq/g)
1	BL-1	Nat. U	0.022	2.716	2.564±0.250	2.741±0.563	2.587±0.272
2	S-13 <sup>a)</sup>	U <sub>3</sub> O <sub>8</sub>	0.039	4.083	3.565±0.321	4.006±0.639	4.053±0.331
3	RGU-1	Nat. U	0.040	4.939	4.614±0.053	4.969±0.660	4.880±0.074
4	OIEA-27 <sup>b)</sup>	U <sub>3</sub> O <sub>8</sub>	0.140	14.658	12.601±2.109	14.353±2.588	12.601±2.109
5	BL-4	Nat. U	0.173	21.360	18.686±0.886	21.143±1.378	17.926±0.199
6	S-4	U <sub>3</sub> O <sub>8</sub>	0.375	39.263	33.563±4.178	38.234±0.494	28.414±0.179
7	S-3	U <sub>3</sub> O <sub>8</sub>	0.418	43.765	35.739±0.249	39.313±1.241	30.424±0.382
8	BL-2	Nat. U	0.453	55.931	45.573±0.251	55.851±1.734	44.450±0.267
9	S-1	U <sub>3</sub> O <sub>8</sub>	0.471	49.314	40.739±0.388	49.818±1.601	40.735±0.334
10	OIEA-7 <sup>c)</sup>	U <sub>3</sub> O <sub>8</sub>	0.514	53.790	45.399±1.761	50.415±2.515	45.016±2.798
11	BL-3	Nat. U	1.020	125.938	104.718±1.267	124.716±3.153	95.960±6.313

**1, 5, 8 and 11:** Prepared by Canadian Centre for Mineral and Energy Technology, Ottawa, Ontario, Canada

**2, 4 and 10:** Prepared by Junta De Energia Nuclear(JDEN), Madrid, Spain

**3:** Uranium ore diluted with Silica prepared by IAEA Analytical Quality Control Service, Vienna, Austria

**6:** Low Grade Uranium Ore:URANINITE (Origin: Australia)

**7:** Low Grade Uranium Ore:CARNOTITE(Origin: USA)

**9:** Low Grade Uranium Ore:TORBERNITE (Origin: Australia)

a) IAEA Uranium Ore Standard(Pitchblende)

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

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

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

♣:Errors are based on counting statistics of  $\pm 1.65\sigma$

**Table 3.** The corrected activities for self absorption of the CRMs and equilibrium states between near and distant members of  $^{238}\text{U}$  decay chain.

No.	Standard Code	Uranium	Certified Uranium Concentration (wt. %)	Certified $^{238}\text{U}$ activity* (Bq/g)	Corrected $^{238}\text{U}$ (via 63.3 keV from $^{234}\text{Th}$ ) activity (Bq/g)	Corrected $^{238}\text{U}$ (via 1001 keV from $^{234\text{m}}\text{Pa}$ ) activity (Bq/g)	Equilibrium Condition $^{234}\text{Th}/^{210}\text{Pb}$
1	BL-1	Nat. U	0.022	2.716	2.692	2.745	0.942
2	S-13	$\text{U}_3\text{O}_8$	0.039	4.083	4.075	4.049	0.859
3	RGU-1	Nat. U	0.040	4.939	4.849	4.979	0.901
4	OIEA-27	$\text{U}_3\text{O}_8$	0.140 <sup>♦</sup> )	14.658	12.643	14.625	0.929
5	BL-4	Nat. U	0.173	21.360	20.981	21.661	0.942
6	S-4	$\text{U}_3\text{O}_8$	0.375	39.263	39.074	39.205	1.030
7	S-3	$\text{U}_3\text{O}_8$	0.418	43.765	41.850	40.877	1.053
8	BL-2	Nat. U	0.453	55.931	52.276	57.358	0.906
9	S-1	$\text{U}_3\text{O}_8$	0.471	49.314	47.989	51.760	0.885
10	OIEA-7	$\text{U}_3\text{O}_8$	0.514 <sup>♦♦</sup> )	53.790	53.775	52.331	0.916
11	BL-3	Nat. U	1.020	125.938	124.562	126.586	0.996

<sup>♦</sup> Mean value is calculated from the five certified values: 0.142%, 0.136%, 0.149%, 0.137% and 0.136%.

<sup>♦♦</sup> Mean value is calculated from the four certified values: 0.527%, 0.527%, 0.526% and 0.475%.

**Table 4.** Mass attenuation coefficients for 46.5 keV, 63.3 keV and 1001 keV gamma-rays in components of S-1 with weight percents

Component	Weight (%)	Mass Absorption Coefficient* ( $\text{cm}^2 \text{g}^{-1}$ )		
	S – 1	46.5 keV	63.3 keV	1001 keV
$\text{Al}_2\text{O}_3$	6.400	$2.159 \times 10^{-2}$	$1.403 \times 10^{-2}$	$4.001 \times 10^{-3}$
$\text{B}_2\text{O}_3$	0.040	$8.410 \times 10^{-5}$	$6.960 \times 10^{-5}$	$2.490 \times 10^{-5}$
$\text{BaO}$	0.030	$4.726 \times 10^{-3}$	$1.889 \times 10^{-3}$	$1.760 \times 10^{-5}$
$\text{CaO}$	0.060	$5.931 \times 10^{-4}$	$2.704 \times 10^{-4}$	$3.830 \times 10^{-5}$
$\text{Cr}_2\text{O}_3$	0.010	$1.437 \times 10^{-4}$	$6.120 \times 10^{-5}$	$6.100 \times 10^{-6}$
$\text{CuO}$	0.040	$1.097 \times 10^{-3}$	$4.336 \times 10^{-4}$	$2.400 \times 10^{-5}$
$\text{Fe}_2\text{O}_3$	3.600	$6.778 \times 10^{-2}$	$2.644 \times 10^{-2}$	$2.199 \times 10^{-3}$
$\text{Ga}_2\text{O}_3$	0.002	$6.030 \times 10^{-5}$	$2.380 \times 10^{-5}$	$1.200 \times 10^{-6}$
$\text{K}_2\text{O}$	1.100	$1.042 \times 10^{-2}$	$4.767 \times 10^{-3}$	$6.867 \times 10^{-4}$
$\text{Li}_2\text{O}$	0.006	$1.150 \times 10^{-5}$	$9.800 \times 10^{-6}$	$3.600 \times 10^{-6}$
$\text{MgO}$	0.390	$1.251 \times 10^{-3}$	$8.372 \times 10^{-4}$	$2.467 \times 10^{-4}$
$\text{MnO}$	0.030	$5.280 \times 10^{-4}$	$2.166 \times 10^{-4}$	$1.790 \times 10^{-5}$
$\text{Na}_2\text{O}$	0.100	$2.954 \times 10^{-4}$	$2.033 \times 10^{-4}$	$6.170 \times 10^{-5}$
$\text{NiO}$	0.002	$5.210 \times 10^{-5}$	$2.050 \times 10^{-5}$	$1.200 \times 10^{-6}$
$\text{P}_2\text{O}_5$	0.700	$2.725 \times 10^{-3}$	$1.666 \times 10^{-3}$	$4.402 \times 10^{-4}$
$\text{PbO}$	0.020	$1.907 \times 10^{-3}$	$7.753 \times 10^{-4}$	$1.410 \times 10^{-5}$
$\text{SiO}_2$	85.000	$3.121 \times 10^{-1}$	$1.968 \times 10^{-1}$	$5.411 \times 10^{-2}$
$\text{SrO}$	0.040	$2.034 \times 10^{-3}$	$7.870 \times 10^{-4}$	$2.500 \times 10^{-5}$
$\text{ThO}_2$	0.001	$1.225 \times 10^{-4}$	$5.040 \times 10^{-5}$	$8.000 \times 10^{-7}$
$\text{TiO}_2$	0.500	$5.105 \times 10^{-3}$	$2.293 \times 10^{-3}$	$3.041 \times 10^{-4}$
$\text{U}_3\text{O}_8$	0.471	$5.699 \times 10^{-2}$	$2.343 \times 10^{-2}$	$3.609 \times 10^{-4}$
$\text{V}_2\text{O}_5$	0.030	$3.190 \times 10^{-4}$	$1.414 \times 10^{-4}$	$1.840 \times 10^{-5}$

\*Mass attenuation coefficients for gamma rays estimated by linear interpolation using literature data[15].

**Table 5.** Mass attenuation coefficients for 46.5 keV, 63.3 keV and 1001 keV gamma-rays in components of S-3 with weight percents

Component	Weight (%)	Mass Absorption Coefficient* ( $\text{cm}^2 \text{g}^{-1}$ )		
	S – 3	46.5 keV	63.3 keV	1001 keV
Al <sub>2</sub> O <sub>3</sub>	6,000	$2.024 \times 10^{-2}$	$1.315 \times 10^{-2}$	$3.751 \times 10^{-3}$
B <sub>2</sub> O <sub>3</sub>	0,003	$6.300 \times 10^{-6}$	$5.200 \times 10^{-6}$	$1.900 \times 10^{-6}$
BaO	0,040	$6.302 \times 10^{-3}$	$2.519 \times 10^{-3}$	$2.340 \times 10^{-5}$
CaO	0,550	$5.436 \times 10^{-3}$	$2.479 \times 10^{-3}$	$3.513 \times 10^{-4}$
Cr <sub>2</sub> O <sub>3</sub>	0,004	$5.750 \times 10^{-5}$	$2.450 \times 10^{-5}$	$2.400 \times 10^{-6}$
CuO	0,020	$5.487 \times 10^{-4}$	$2.168 \times 10^{-4}$	$1.200 \times 10^{-5}$
Fe <sub>2</sub> O <sub>3</sub>	0,500	$9.414 \times 10^{-3}$	$3.672 \times 10^{-3}$	$3.054 \times 10^{-4}$
K <sub>2</sub> O	2,500	$2.367 \times 10^{-2}$	$1.083 \times 10^{-2}$	$1.561 \times 10^{-3}$
MgO	1,200	$3.848 \times 10^{-3}$	$2.576 \times 10^{-3}$	$7.590 \times 10^{-4}$
MnO	0,090	$1.584 \times 10^{-3}$	$6.498 \times 10^{-4}$	$5.370 \times 10^{-5}$
Na <sub>2</sub> O	1,050	$3.102 \times 10^{-3}$	$2.135 \times 10^{-3}$	$6.478 \times 10^{-4}$
P <sub>2</sub> O <sub>5</sub>	0,100	$3.893 \times 10^{-4}$	$2.379 \times 10^{-4}$	$6.290 \times 10^{-5}$
SiO <sub>2</sub>	80,000	$2.937 \times 10^{-1}$	$1.852 \times 10^{-1}$	$5.093 \times 10^{-2}$
SrO	0,040	$2.034 \times 10^{-3}$	$7.870 \times 10^{-4}$	$2.500 \times 10^{-5}$
ThO <sub>2</sub>	0,0004	$4.900 \times 10^{-5}$	$2.020 \times 10^{-5}$	$3.000 \times 10^{-7}$
TiO <sub>2</sub>	0,330	$3.369 \times 10^{-3}$	$1.513 \times 10^{-3}$	$2.007 \times 10^{-4}$
U <sub>3</sub> O <sub>8</sub>	0,418	$5.057 \times 10^{-2}$	$2.079 \times 10^{-2}$	$3.203 \times 10^{-4}$
V <sub>2</sub> O <sub>5</sub>	0,580	$6.166 \times 10^{-3}$	$2.733 \times 10^{-3}$	$3.548 \times 10^{-4}$
ZnO	0,040	$1.219 \times 10^{-3}$	$4.792 \times 10^{-4}$	$2.410 \times 10^{-5}$

\*Mass attenuation coefficients for gamma rays estimated by linear interpolation using literature data[15]

**Table 6.** Mass attenuation coefficients for 46.5 keV, 63.3 keV and 1001 keV gamma-rays in components of BL-2 with weight percents

Component	Weight (%)	Mass Absorption Coefficient* (cm <sup>2</sup> g <sup>-1</sup> )		
	BL – 2	46.5 keV	63.3 keV	1001 keV
Al	6,620	2.877 x 10 <sup>-2</sup>	1.665 x 10 <sup>-2</sup>	4.069 x 10 <sup>-3</sup>
Ca	4,060	5.246 x 10 <sup>-2</sup>	2.264 x 10 <sup>-2</sup>	2.594 x 10 <sup>-3</sup>
Fe	4,750	1.232 x 10 <sup>-1</sup>	4.615 x 10 <sup>-2</sup>	2.847 x 10 <sup>-3</sup>
K	0,330	3.610 x 10 <sup>-3</sup>	1.599 x 10 <sup>-3</sup>	2.052 x 10 <sup>-4</sup>
Mg	1,500	5.727 x 10 <sup>-3</sup>	3.534 x 10 <sup>-3</sup>	9.444 x 10 <sup>-4</sup>
Na	3,420	1.091 x 10 <sup>-2</sup>	7.198 x 10 <sup>-3</sup>	2.086 x 10 <sup>-3</sup>
SiO <sub>2</sub>	59,120	2.171 x 10 <sup>-1</sup>	1.369 x 10 <sup>-1</sup>	3.764 x 10 <sup>-2</sup>
H <sub>2</sub> O	0,190	5.060 x 10 <sup>-5</sup>	3.770 x 10 <sup>-5</sup>	1.340 x 10 <sup>-5</sup>
U	0,453	6.445 x 10 <sup>-2</sup>	2.642 x 10 <sup>-2</sup>	3.576 x 10 <sup>-4</sup>
Pb	0,090	9.227 x 10 <sup>-3</sup>	3.746 x 10 <sup>-3</sup>	6.390 x 10 <sup>-5</sup>
S	0,036	2.590 x 10 <sup>-5</sup>	1.280 x 10 <sup>-5</sup>	2.300 x 10 <sup>-6</sup>

\*Mass attenuation coefficients for gamma rays estimated by linear interpolation using literature data[15]

**Table 7.** Mass attenuation coefficients for 46.5 keV, 63.3 keV and 1001 keV gamma-rays in components of BL-4 with weight percents

Component	Weight (%)	Mass Absorption Coefficient* (cm <sup>2</sup> g <sup>-1</sup> )		
	BL – 4	46.5 keV	63.3 keV	1001 keV
Al	6,750	2.934 x 10 <sup>-2</sup>	1.698 x 10 <sup>-2</sup>	4.149 x 10 <sup>-3</sup>
Ca	3,270	4.225 x 10 <sup>-2</sup>	1.823 x 10 <sup>-2</sup>	2.089 x 10 <sup>-3</sup>
Fe	5,260	1.364 x 10 <sup>-1</sup>	5.110 x 10 <sup>-2</sup>	3.153 x 10 <sup>-3</sup>
K	0,360	3.939 x 10 <sup>-3</sup>	1.744 x 10 <sup>-3</sup>	2.238 x 10 <sup>-4</sup>
Mg	1,380	5.268 x 10 <sup>-3</sup>	3.251 x 10 <sup>-3</sup>	8.688 x 10 <sup>-4</sup>
Na	3,240	1.033 x 10 <sup>-2</sup>	6.819 x 10 <sup>-3</sup>	1.976 x 10 <sup>-3</sup>
SiO <sub>2</sub>	61,220	2.248 x 10 <sup>-1</sup>	1.417 x 10 <sup>-1</sup>	3.897 x 10 <sup>-2</sup>
H <sub>2</sub> O	0,160	4.260 x 10 <sup>-5</sup>	3.170 x 10 <sup>-5</sup>	1.130 x 10 <sup>-5</sup>
U	0,173	2.461 x 10 <sup>-2</sup>	1.009 x 10 <sup>-2</sup>	1.366 x 10 <sup>-4</sup>
Pb	0,031	3.178 x 10 <sup>-3</sup>	1.290 x 10 <sup>-3</sup>	2.200 x 10 <sup>-5</sup>
S	0,031	2.230 x 10 <sup>-5</sup>	1.100 x 10 <sup>-5</sup>	2.000 x 10 <sup>-6</sup>

\*Mass attenuation coefficients for gamma rays estimated by linear interpolation using literature data[15]

**Table 8.** Self-absorption factors for 46.5 keV, 63.3 keV and 1001 keV gamma rays by the four CRMs

Factors	S-1			S-3		
	46.5 keV	63.3 keV	1001 keV	46.5keV	63.3 keV	1001 keV
k	0.155	0.155	0.155	0.155	0.155	0.155
m	7.919	7.919	7.919	8.3786	8.3786	8.3786
$\mu$	0.4899	0.2752	0.06260	0.4318	0.2500	0.05938
$k\mu m$	0.601	0.338	0.077	0.561	0.325	0.077
f	1.331	1.178	1.039	1.306	1.171	1.039

Factors	BL – 2			BL – 4		
	46.5 keV	63.3 keV	1001 keV	46.5 keV	63.3keV	1001 keV
k	0.155	0.155	0.155	0.155	0.155	0.155
m	6.844	6.844	6.844	6.071	6.071	6.071
$\mu$	0.5155	0.2648	0.05082	0.4802	0.2513	0.05160
$k\mu m$	0.5470	0.2810	0.0539	0.4519	0.2365	0.0486
f	1.2983	1.1471	1.0272	1.306	1.171	1.039

$$k = \frac{3}{2} \left( \frac{1}{\pi r L} \right) \text{geometric parameter (cm}^{-2}\text{)},$$

m: sample weight(g),

$\mu$ : mass attenuation coefficient of sample (cm<sup>2</sup>.g<sup>-1</sup>),

$k\mu m$ : dimensionless parameter inserted in Eq.(4),

f: self-absorption factor at a given gamma-ray energy.

## Figure Captions

Fig. 1. Decay scheme of  $^{238}\text{U}$

Fig. 2. Schematic diagram of sample - detector counting geometry

Fig. 3. Measured and certified uranium activities versus uranium concentrations of CRMs via 63.3 keV peak of  $^{234}\text{Th}$ .

Fig. 4. Measured and certified uranium activities versus uranium concentrations of CRMs via 1001 keV peak of  $^{234\text{m}}\text{Pa}$ .

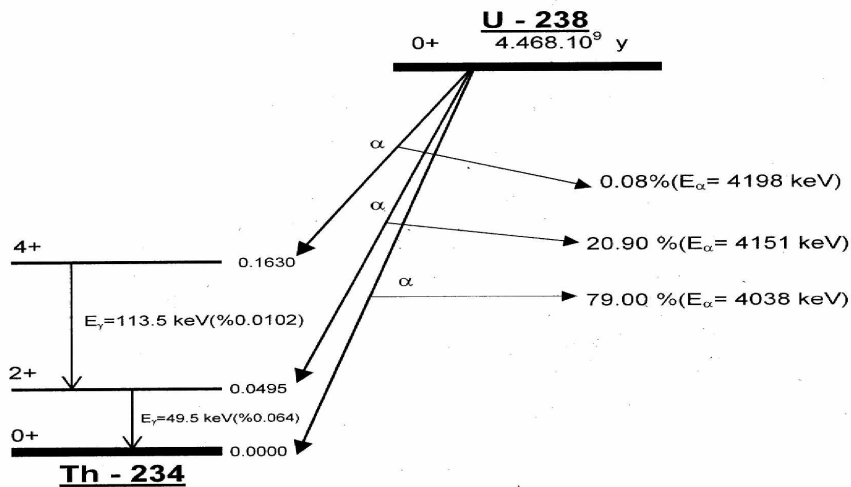


Fig. 1. Decay scheme of  $^{238}\text{U}$

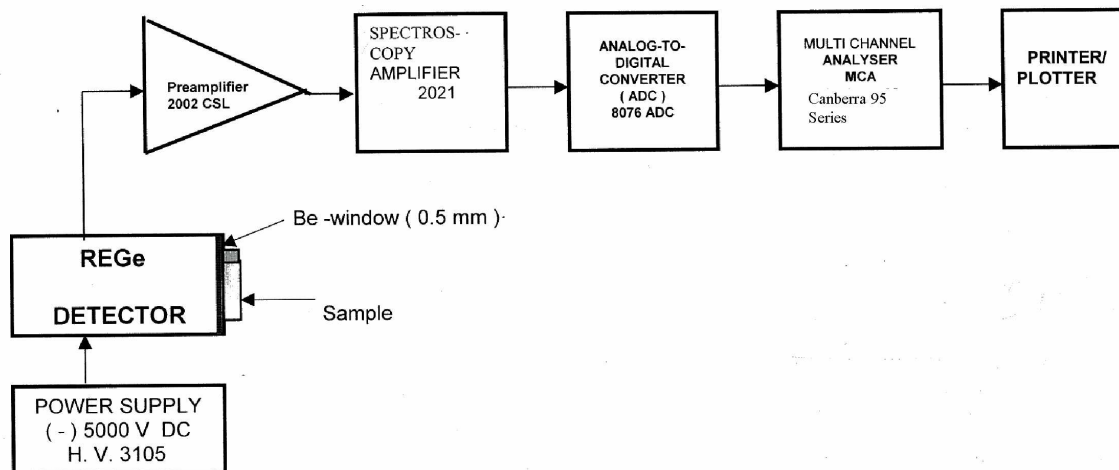
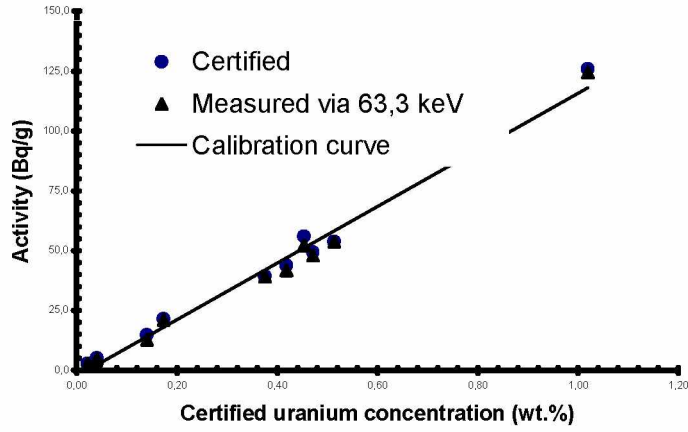
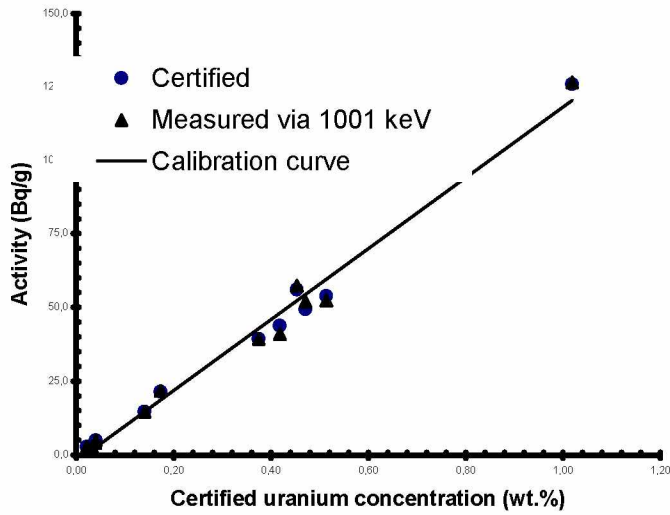


Fig. 2. Block Diagram of Gamma Counting System



**Fig. 3.** Measured and certified uranium activities versus uranium concentrations of CRMs via 63.3 keV peak of  $^{234}\text{Th}$ .



**Fig. 4.** Measured and certified uranium activities versus uranium concentrations of CRMs via 1001 keV peak of  $^{234m}\text{Pa}$ .