Applied Radiation and Isotopes 71 (2013) 11–20

Contents lists available at SciVerse ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Self-absorption correction in determining the ^{238}U activity of soil samples via 63.3 keV gamma ray using MCNP5 code

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HIGHLIGHTS

- Determination of the ^{238}U activity via 63.3 keV gamma rays.
- Self-attenuation factors of 63.3 keV gamma rays for cylindrical sample container.
- The density, chemical composition and geometry effects are taken into account.
- Determination of the ^{238}U activity in three soil types: grey, alluvial and red soils.

ARTICLE INFO

Article history:
Received 27 January 2012
Received in revised form
5 September 2012
Accepted 6 September 2012
Available online 23 September 2012

Keywords:
Self-absorption
Self-attenuation factor
^{238}U activity
63.3 keV
MCNP5 code
HPGe detector

ABSTRACT

The essential issue in analyzing the activity of ^{238}U in an HPGe detector based gamma spectrometer via 63.3 keV line is relating to the strong self-absorption of this weak gamma ray in sample material. The present work suggests a method of the self-absorption corrections for 63.3 keV gamma rays by a combination of experimental measurements and Monte Carlo MCNP5 calculations. The effects of sample chemical composition, density and geometry were calculated in terms of self-attenuation factors. The method, developed for a cylindrical sample geometry, accounted for variable sample heights and densities. The analysis of ^{238}U activity was applied for three main soil types in Vietnam, which are grey, alluvial and red soils. The results obtained with the above outlined method were in good agreement with those derived by other methods.

1. Introduction

The activity of ^{238}U is commonly determined in an HPGe detector based gamma spectrometer by indirect methods. The direct analysis of the ^{238}U activity is impossible because it emits two gamma rays with very weak intensities, namely 49.55 keV (0.0697%) and 113.5 keV (0.0174%), which lie in a large Compton scattering background of gamma spectrum.

The indirect analysis of ^{238}U is accomplished by means of the measurement of gamma rays emitted from daughter nuclides in uranium decay chains. The daughter nuclides often used are: ^{234}Th ($T_{1/2}$=24.1 days), ^{234m}Pa ($T_{1/2}$=1.17 min), ^{214}Pb ($T_{1/2}$=26.8 min) and ^{214}Bi ($T_{1/2}$=19.9 min). Among them, ^{214}Pb and ^{214}Bi are widely applied because of large intensities of gamma rays emitted from these nuclides. They are 241.9 keV (7.268%), 295.2 keV (18.5%), 351.9 keV (35.6%) gamma lines emitted from ^{214}Pb and 609.3 keV (45.49%), 768.4 keV (4.891%) and 1120.3 keV (14.909%) gamma lines emitted from ^{214}Bi. The analysis of ^{238}U activity by using ^{214}Pb and ^{214}Bi faces with two disequilibria. The first is a disequilibrium between ^{226}Ra and ^{214}Pb, ^{214}Bi due to a leakage of ^{222}Rn ($T_{1/2}$=3.825 days), which is a noble gas and is the decay product of ^{226}Ra. To overcome this disequilibrium, samples are sealed for a month to attain the equilibrium between ^{226}Ra and ^{222}Rn. Then, the average activity of ^{214}Pb and ^{214}Bi can be assigned to that of ^{226}Ra as reported in Huy and Luyen (2006). This value is not considered to be ^{238}U activity because there is still a second geochemical disequilibrium between ^{226}Ra and ^{238}U. The disequilibrium is caused by the different chemical properties of ^{238}U and ^{226}Ra in soil environment and a long lifetime of 1620 years for ^{226}Ra. So, the ^{238}U activity obtained by indirect analysis via ^{214}Pb and ^{214}Bi is only approximate when neglecting the geochemical disequilibrium.

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0969-8043/$ - see front matter © 2012 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.apradiso.2012.09.004

^{234}Th and ^{234m}Pa are the nearest daughters of ^{238}U and are very short-lived nuclides compared to ^{238}U, so radioactive equilibrium is quickly established. As a result, the geochemical disequilibrium between these nuclides and ^{238}U can be ignored and the activities obtained for these nuclides can be assigned to that of ^{238}U. The gamma rays preferably used for ^{238}U analysis are 63.3 keV (4.8%) and 92.6 keV (5.58%) emitted from ^{234}Th, and 766.4 keV (0.316%) and 1001.0 keV (0.839%) from ^{234m}Pa (Yucel et al., 1998, 2009; Huy and Luyen, 2004; Dowdall et al., 2004; De Corte et al., 2005; Kaste et al., 2006). However, from the point of view of analysis of ^{238}U activity in soil samples with low specific activities, the measurement of 766.4 keV and 1001.0 keV gamma rays can give activities with large uncertainties because of their weak intensities and low efficiencies of germanium detectors at high energies. Gamma ray of 92.6 keV is a doublet, consisting of 92.4 keV (2.81%) and 92.8 keV (2.77%) and is interfered by other K-X- and gamma peaks, including 93.3 keV. So, in practice, the 92.6 keV peak cannot be separated from the multiplet of about 93 keV. The remaining 63.3 keV gamma ray includes contributions from the 63.3 keV gamma ray (4.8%) emitted from ^{234}Th, 63.9 keV gamma ray (0.023%) from ^{231}Th and 63.9 keV gamma ray (0.255%) from ^{232}Th, but the contributions of ^{231}Th and ^{232}Th can be neglected (Kim and Burnett, 1985; Huy and Luyen, 2004). Therefore, the 63.3 keV gamma ray is a preferred candidate for determination of ^{238}U activity in environmental soil samples.

The important problem in the use of 63.3 keV line for analysis of ^{238}U activity is the strong self-absorption of this weak gamma ray in sample material. As reported in Hasan et al. (2002), the values of the self-absorption correction factor for the IAEA reference samples of 1.0 g/cm^3 and 1.4 g/cm^3 densities, packed in 5 cm^3 tube geometry and measured with a well-type HPGe detector, are approximately 13%, 5% and 2% for the energy ranges of 40–160 keV, 200–600 keV and >600 keV, respectively. Therefore, the self-absorption corrections must be taken into account when the 63.3 keV gamma ray is used. For the volumetric sample with a homogeneous distribution of the attenuating material and the radioactive source, the influencing factors on self-absorption are composed of chemical composition, density and geometry effects. The self-absorption correction factor for a sample under study with given chemical composition, density and geometrical dimension was determined by calculating the ratio between its counting efficiency and that of a standard reference sample with known self-absorption property. Some works have made corrections for only density and geometry effects neglecting the composition effect (Kitto, 1991; Melquiades and Appoloni, 2001; Abbas, 2001; Vargas et al., 2002; Huy and Luyen, 2004). The self-absorption corrections for all three effects, including the composition one, were conducted in the works of Hasan et al. (2002), San Miguel et al. (2002), Nachab et al. (2004) and Carrazana Gonzalez et al. (2010). An approach to perform the self-absorption corrections is to calculate the self-attenuation factor suggested by Debertin and Helmer (1988) and developed by Korun (1999, 2000), Korun and Vidmar (2003), Boshkova and Minev (2001) and Boshkova (2003). This factor describes the probability of interaction between the photons and the sample material and is given by the ratio between the counting efficiency for the actual sample $\varepsilon_V(\mu,E)$, where μ, E and V denote the attenuation coefficient, the energy of photon and the sample volume, respectively, and the counting efficiency for the same sample-detector geometry but without self-attenuation $\varepsilon_V(0,E)$ (Korun, 1999):

$$F_V(\mu,E)=\frac{\varepsilon_V(\mu,E)}{\varepsilon_V(0,E)} \quad (1)$$

The advantage of the self-attenuation factor is that the detector properties enter the expression via both efficiencies and cancel out to a large extent in the ratio. Therefore, the self-attenuation factor is given as a function of sample parameters, disregarding the detector properties.

The aim of present work is to develop a method to analyze ^{238}U activity in soil samples via 63.3 keV by a combination of experimental measurement and Monte Carlo MCNP5 calculation. The corrections of all chemical composition, density and geometry effects were carried out by application of the self-attenuation factor. The method, developed for a cylindrical sample geometry, accounted for variable sample heights and densities. The analysis of ^{238}U activity was applied for three main soil types in Vietnam, which are grey, alluvial and red soils.

2. Experimental

The measuring system used in the study was the Canberra HPGe GC1518 p-type detector based gamma spectrometer installed at the Center for Nuclear Techniques Ho Chi Minh City, Vietnam. It has a relative efficiency of 15% and an energy resolution of 1.8 keV at 1332 keV line. The geometry dimensions and material compositions of the lead shield and the GC1518 detector were described in Huy et al. (2007). An important parameter of the HPGe GC1518 p-type detector is its thickness of dead layer, which was 0.35 mm of germanium equivalent as provided by the manufacturer in 1996 and grown over operation time to 0.65 mm in 1999, 1.15 mm in 2005 and 1.46 mm in 2009 (Huy and Ngo Quang, 2010). The sample container used in the measurement had cylindrical form of 7.2 cm diameter and 5 cm height. The height of sample material depended on its mass and density.

3. Reliability of MCNP5 calculation of self-absorption corrections for 63.3 keV gamma rays

The reliability of MCNP5 calculation of self-absorption corrections for 63.3 keV gamma rays was controlled by using a ^{238}U standard sample and a ^{238}U water solution, chemical compositions of which are presented in Table 1. The ^{238}U standard was supplied by Institute of Nuclear Science and Technology in Hanoi (INST sample) with a specific activity of 1296 ± 12 Bq/kg. The ^{238}U water solution was prepared by addition of an amount of [^{238}U]uranyl-acetate to water with an activity of 11939 ± 126 Bq/kg. Different samples of heights between 0.2 cm and 2.4 cm with 0.1 cm increments for INST sample and between 0.2 cm and 1.8 cm with 0.1 cm increments for water solution were measured in the spectrometer. With the model of detector, lead shield and sample cylindrical container as described in Huy et al. (2007) we simulated gamma spectra by the MCNP5 calculation. The experiment of measuring the efficiencies of INST and water samples were carried out in 2003, therefore the dead layer thickness of GC1518 detector was taken to be 0.9 mm. The F8 tally of the MCNP5 code is a pulse height tally, which provides the energy distribution of pulses created in a detector by radiation. It was used to extract the pulse height distribution of gamma spectra, and the 63.3 keV full energy peak efficiency was determined. Simulation relative error was kept under 0.3% with a total of 3,000,000 source gamma-rays. The densities, chemical compositions and geometries of INST and water samples included in the input data of the MCNP5 code are presented in Table 1.

From the gamma spectra measured in the GC1518 detector based gamma spectrometer, the experimental efficiencies of 63.3 keV full energy peaks were determined with relative uncertainties of less than 5% and plotted in Fig. 1 against the sample height. From Fig. 1 it is seen that the curves calculated by the MCNP5 code describe well experimental data. Besides, the ratios