LOW LEVEL RADIOACTIVITY MEASUREMENTS OF $^{238}$U, $^{232}$Th, $^{40}$K AND $^{137}$Cs IN THE ENVIRONMENTAL AND INDUSTRIAL SAMPLES BY HIGH RESOLUTION GAMMA-RAY SPECTROMETRY

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1. Introduction

More than 40 years after its launching, the gamma-ray spectrometry with especially high pure Germanium detectors developed in the last 30 years, is nowadays applied to measure quantitatively gamma emitting radionuclides for various research purposes and environmental problems in numerous laboratories world-wide. Since the gamma-ray spectrometry is a non-destructive and fast analytical technique and it gives the high accuracy and precision in the results, this technique is commonly used in measurements of the low level radioactivity contained various samples, e.g., rock, soil, sediments, sludge, mineral ores, cement, ceramics, clays and petroleum industry. Particularly, in areas of igneous geology, the measurements of concentrations of naturally occuring U, Th series nuclides and $^{40}$K are significant [1]. Additionally, a need has recently arisen to find and produce materials containing especially low in radioactive promordial elements thorium, uranium and potassium [2].

The comparator (i.e, relative) method for radioactivity measurements in samples is straightforward if the standard materials are similar to the samples, especially in view of matrix composition. However, it is difficult to obtain such suitable standard materials. Therefore, the direct (or absolute) gamma spectrometric method for radionuclide analysis in samples is generally applied in a calibrated Ge detector.

In this work, the gamma emitting radionuclides in the spiked soil samples and standard solutions prepared by IAEA for proficiency tests have been analysed by using a gamma-ray spectrometer with a n-type detector within the quality control and quality assurance of the employed technique at ANRTC. The obtained results (i.e., the results reported to IAEA) for proficiency test samples sent by IAEA were compared with the IAEA data by using statistical methods (z-score and u-test score). In this paper, the gamma-ray measuring system and experimental procedure used have been described in detail and the results obtained by absolute gamma-spectrometric method and the target values (IAEA measurements) and their uncertainties have been discussed for the validation of method in view of the proficiency test. Then, the analysis results for various environmental and industrial samples are given.

2. Experimental Procedure

For gamma spectrometric measurements, two p-type and one n-type Ge detectors are being routinely used at ANRTC. The gamma measuring system consisting of a commercially available n-type Ge detector (reverse electrode germanium crystal, REGe)
with its own preamplifier, a spectroscopy amplifier, a 100 MHz- Wilkinson type ADC and a multichannel analyzer (Canberra Model 95+) is described here as an example for me gamma-ray spectrometers used in the measurements. The REGe detector has a total active volume of 100 cm$^3$ and 0.5 mm thick Be window. So, it has high efficiencies for low energy gamma-rays (< 120 keV). The REGe detector has a relative efficiency of 22.6% and its measured resolution is 1.80 keV for 1332.5 keV ($^{60}$Co) and 0.97 keV for 122 keV ($^{57}$Co). The detector was shielded by chevron type lead bricks with 10 cm thickness. The lead gamma shielding of the detector was lined with 1 mm thick Cu sheets to reduce Pb-rays.

The full-energy detection efficiency as a function of gamma-ray energy for the REGe detector was determined using the powder radioactive standard containing a mixture of $^{109}$Cd, $^{57}$Co, $^{123m}$Te, $^{51}$Cr, $^{113}$Sn, $^{85}$Sr, $^{137}$Cs, $^{60}$Co and $^{88}$Y in SiO$_2$ matrix and a 5 ml ampoule containing radionuclides $^{57}$Co, $^{60}$Co, $^{88}$Y, $^{109}$Cd, $^{113}$Sn, $^{137}$Cs from the Isotope Products Laboratories Inc traceable to NIST.

The measured absolute photopeak efficiency data at a desired and reproducible source to detector distance are fitted using the least square fitting method to an efficiency function in the following form:

$$\epsilon_p(E_\gamma) = a + b \exp(-cE_\gamma^d)$$  \hspace{1cm} (1)

where, $\epsilon_p$: absolute photopeak efficiency as a function of gamma-ray energy, $E_\gamma$ and a, b, c and d are constants determined by the fitting method.

For a given source sample geometry, the weighed spiked soil samples coded as RER/2/004/TU-ANK/1 and RER/2/004/TU-ANK/14, and standard solutions coded as RER/2/004/TU-ANK/2 and RER/2/004/TU-ANK/15 were counted in a calibrated REGe detector for different counting times of 100 000 s, 150 000 s and 235 000 s according to the level of activity of the sample to ensure good statistical quality of data. The measurements for each sample were repeated at least three times to improve statistical precision. Thus, the mean values of the measured count rates were used in the calculations.

In peak analysis procedure, as is known, there are many ways of calculating the net gamma ray count. The employed here is valid, common method when there are no interference from photopeaks adjacent to the peak of interest. In case of the interference gamma peaks from the radionuclides that may be contained in sample and/or ambient background, a peak deconvolution method (e.g. Gauss Fitting Deconvolution Method) is applied to separate the adjacent peaks.

Since the series Canberra 95+ MCA used 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 0.1%. Also, the background spectrum was separately collected several times for the different periods. The background peak areas were converted to count rates and then the average value from the background runs were subtracted from photopeak count rates to obtain the net sample count rate for each photopeak of interest. In these measurements the laboratory background values were checked by using control chart of background. They remained within ±1σ (standard deviation) levels of the control chart for this detector. The specific activity of any radionuclide in the sample using net counts of the peak of interest in the measured spectrum was calculated by the following formula: