Use of gamma spectroscopy based on HPGe detector to assess the radioactivity of NORM in environmental samples

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Abstract

Natural environment radionuclides are responsible for the most of the external exposures of gamma radiation. Radionuclides in the natural environment are responsible for most external exposures to gamma radiation. This contribution is mainly due to the natural chain radionuclides ^{238}U, ^{235}U and ^{232}Th, followed by ^{40}K, which are found globally in Earth. Environmental radioactivity is produced by radioactive materials in the human environment. It is necessary to determine and estimate the activity of various radio nuclides in environmental media such as vegetation, soil, water and fertilizers. In this paper, we will shed some light on how to obtain accurate results of ^{226}Ra, ^{232}Th and ^{40}K measured in environmental samples using a gamma ray spectroscopy system. Using high-resolution γ−ray spectroscopy, the terrestrial gamma radiation in different samples can be determined with high accuracy if correction factors such as dead time, true coincidence, gamma attenuation, absolute full energy peak efficiency, sample geometry and spectroscopic interference are introduced. In this manuscript, these important correction factors will be discussed in more details.

Keywords: gamma spectroscopy, NORM, radioactivity, environmental samples, correction factors

Introduction

Naturally occurring radioactive materials (NORM) due to existence of primordial ^{238}U and ^{232}Th series and ^{40}K radionuclides in the environment constitute about 85% of public exposure to terrestrial gamma radiation (UNSCEAR, 2000). They can be found in soil, rocks, sediments, food chainand other activitiesresulting from human industrial activities.

Natural radiation is the main source of radiation exposure in humans and has led to intensive research works around the world on radiation levels, doses from natural radiation sources and its effects on health.

Furthermore, the knowledge of distribution and classificationof radionuclides in the environment improves our awareness and understanding of radiation damage, and, therefore, is of great

importance as guidance whenstandards and regulatory control actions on radiation protection are established.

Many studies conducted worldwide revealed that ^{238}U and ^{232}Th and their decay products in soil, rocks and sands are the main sources of high natural background radiation [4].Natural environmental radiation and radioactivity in soils have gained considerable research interest because humans are exposed to natural radioactivity at different levels depending on natural radioactive minerals present in each region worldwide [1].

Human are continuously exposed to natural radiation producedmostly from the cosmic rays and the terrestrial natural radionuclides. The source of these radionuclides is the earth's crust. Natural radionuclides are present in rocks, air, water, and fertilizers(El-Taher, 2012). Naturally Occurring Radioactive Materials (NORMs) include all natural radionuclides such as uranium isotopes (^{234}U and ^{238}U), radium isotopes (^{226}Ra, ^{228}Ra), thorium (^{232}Th), radon (^{220}Rn) and potassium-40 (^{40}K). Also, there are a remarkably higher radiation doses to man existing in Naturally Occurring Radioactive Material (NORM), resulting from human industrial activities such as production of building material, cement and concrete and fertilizers (Landsberger et al., 2013).

In addition to the above source of NORM, fertilizers which extensively used in the agriculture sector to boost crop growth, therefor fertilizer industries have speared all overthe world. Since phosphorus is the main constituent of fertilizer, which part of it is extracted from phosphate rocks, contain a relatively high concentration of naturally occurring radioactive materials and their daughters. Therefore, natural radioactivity in soil varies from one location to another due to the extensive use of fertilizer which is the main source of radioactivity in soil other than its natural origin.

Many countries have already conducted intensive studies on NORM and set standards for radio activities exposure to humans. Unfortunately, this kind of studies are not available in Libya, therefore, local people may accumulate large amount of radionuclides from food chain and environmentthat pose a threat to public health.

The objective of this preliminary work is to propose the creation of a database on the amount of radioactivity to which the citizen is exposed throughout the year from different polluting sources (such as food, water, air, etc.) using nuclear techniques.

Gamma-ray Spectroscopy

Gamma ray spectroscopy is considered a powerful tool for determination the radioactivity of ^{238}U, ^{232}Th and their decay products in different environmental samples with high accuracy. By using high-resolution gamma-ray spectrometry, all radioisotopes of ^{238}U and ^{232}Th decay chains, can be investigated simultaneously

The gamma spectroscopy consists of a semiconductor high purity germanium (HPGe) detector, amplifier, analogue to digital convertor and multi-channel analyzer and analytical software for spectrum unfolding.This type of detector is characterized by its high resolution;also, its efficiency ranges from 20 % to over 100 %.

The most significant factors that should be taken in consideration when measuring environmental bulk samples are (spectroscopic interference, self-attenuation, dead time, full energy peak efficiency and true coincidence).

For a given geometric setup, the correction factor is expressed as the ratio of efficiency of standard to that of the sample (Boshkova, 2003)

In practical applications, standard used for constructing the full energy peak efficiencyand the analyzed sample materials might be completely different in their chemical composition. Therefore, the gamma attenuation especially the low energy gamma lines in the sample and standards will suffer significant attenuation. In this case, the self-absorption factor should be determined experimentally. In many studies, the attenuation factor was not givenhigh attention when performing measurements of different environmental samples based on gamma spectroscopy (references thesis of Shariff 2017, 2010القدار). This practice will lead to inaccurate results.

Therefore it's very important when environmental measurement is performed, attenuation correction should be applied for different kind of samples taking into account their differences in density (Cutshall et al, 1983; Bolivar et al, 1997).

The other significant factor, which should be introduced in order to correct for the lost counts rate under the peak of interest,is the dead time. When the measured sample emits high flux of gamma photons (the analogue to digital convertor unit can't handle suchhigh activity. Therefore, many counts under the measured peak of interest will be lost and the obtained values subjected to high uncertainty (Abugassa &Abed Almajed 2018).

The knowledge of absolute full energy peak efficiency is indispensable and crucial parameter, when measuring environmental samples with different matrix and volumes from the available standard.Also, most calibration sources used for efficiency measurements contain multi-energetic radionuclides that suffer from true coincidence phenomena, where two or more gamma lines reach the detector at the same time when measurement carried out on top of the detector cap;therefore, they will beprocessed as one gamma line as shown in the figure (1) below. Unfortunately, many analysts do not pay much attention to this factor; therefore, large discrepancies in the final results were observed (Abugassa etc. 2014,Al-Shariff2017; Jassim 2017).

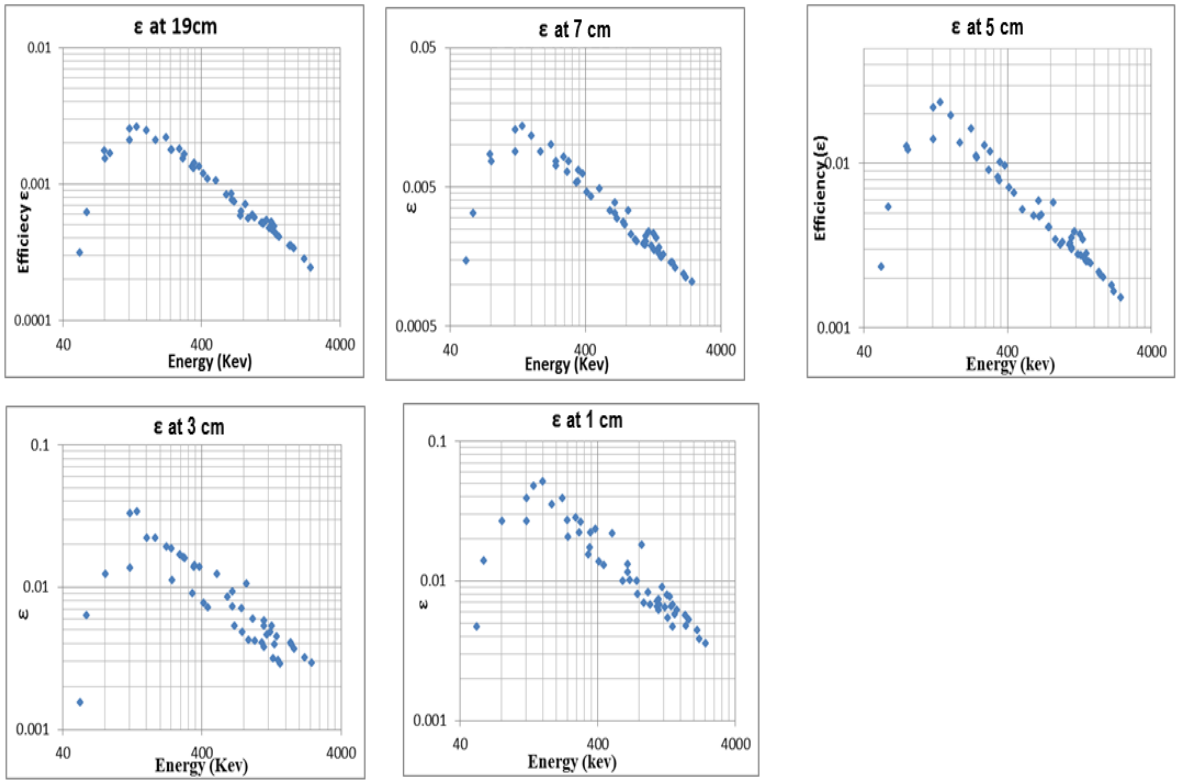

Fig.(1) Absolute efficiency as function of energy measured at different distances from the end cap of the detector

Precise measurements of ^{238}U and its decay products (^{226}Ra, ^{214}Pb and ^{214}Bi) in environmental samples using gamma spectroscopy rely on the assumption of secular equilibrium.In order to achieve that, samples should be kept in their containers for one month before conducting the measurement of the radioactivities in NORM samples. This period of time is enough for radium and its progenies to reach the stage of secular equilibrium.If the sample containers aren't tightly sealed, the radon gas can leak out of the container and, therefore, the secular equilibrium state is disrupted and the values of the mother nuclide ^{226}Ra, and its daughters ^{214}Pb and ^{214}Bi can vary significantly.

Also, ^{238}U is frequently estimated using the energy transitions of 63.3keV (3.6%) and 92.6 (4.9%) of the ^{234}Th (a direct daughter of ^{238}U). Several restrictions draw attention to the use of these two lines to determine ^{238}Udue to interference caused by the 92.6 keV X-ray transition line from Bi, Po, U and Th, interference of the 63.3 keV line from the Th series and the significant self-absorption of both energy transitions due to their considerably low energy.

Conclusion

Since radionuclides are found in land (soil or rocks) or lakes and waters (oceans, seas or lakes), in oil and gas industry, fertilizers and can easily accumulate in the food chain, therefore the use

of HPGe spectroscopy allows determination of NORM radioactivity in various environmental materials. The gamma-spectroscopy method employed the high purity germanium detector to determine the natural and artificial radioactive values, including 226Ra, 232Th, 40K, and 137 Cs.

Using nuclear methods based on gamma spectroscopy, very accurate results can be obtained provided that all correction factors outlined above are performed and introduce in the associated equation of radioactivity calculation.

The importance of knowing the radioactivity in all environmental samples requires the use of such non-destructive methods to know the radioactivity and what radiation doses the person is exposed to and as an indicator of the environmental pollution resulting from human industrial activities, also, such studies can be very useful and the obtained results could be used as a guideline for future investigations and mapping of natural radiation.

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