Theoretical Study on the effect of Radon emanation process in soil samples on the absolute efficiency of an HPGe detector

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Abstract- Radium-226 activity is determined by gamma spectrometry, based on the measurement of radon progeny short lived isotopes ([214]Pb and [214]Bi) when Ra-Rn secular equilibrium is reached. Depending on the behaviors of radon emanation from the soil matrix, the homogeneous distribution of these radionuclides in measured soil samples may be lost, which may affect the quality of the obtained results if not taken into consideration. This paper highlights some assumptions regarding the distribution of radon daughters in the soil matrix and their effect on the detection efficiency of an HPGe detector using Mont Carlo simulation code.

Index Terms- Gamma spectrometry, Mont-Carlo simulation, Particle distribution, Radon emanation,

INTRODUCTION

G amma spectrometry is a useful non-destructive method that is commonly applied to determine, indirectly, the concentration of radium-226 in environmental samples through the measurement of the progeny nuclides of radon isotopes [1]. The progeny of the radon isotope that is commonly used to determine its concentration is composed of the two short lived [214]Pb and [214]Bi radionuclides with corresponding gamma energy line groups of 295.0 , 352.0 and 609.0 1120.0 , 1764.0 keV; respectively.

The importance in the measurements of radon isotopes lies in their detrimental effect on human. Since they occur in nature, man has always been exposed, mainly through inhalation of their decay products. Radon is released to the atmosphere through three modes namely; i) emanation: as its atom emanated from radium decay is escaping by recoil energy into the pores of the grains; ii) transport or diffusion: which is the diffusion flow that causes the movement of radon atoms to ground surface and iii) exhalation: where the transported atoms are exhaled to the atmosphere [2]. The most important factor relating to radon concentration is the emanation coefficient which is the fraction of radon that reaches the pore space. It depends on different factors such as crystalline structure, grain size and moisture contents [374]. The values of emanation coefficient vary from 1% to 50% over a wide range of materials, conditions, definitions and measurement methods [5&6]. The radon exhalation from soil surface is affected by the soil's characteristics like radium contents, the internal structure, grain size, porosity, permeability and the emanation coefficient, etc.[7]

For the determination of emanation coefficient of [222]Rn using gamma spectrometry, the sample is sealed in a container and the count rates from [214]Pb or [214]Bi gamma lines are measured at the time of sealing and after the establishment of radioactive equilibrium between [226]Ra and its progeny (28 days) [8].

Several techniques are used to determine the emanation factor and the effective [226]Ra activity from [222]Rn exhalation [9&10]. Two main approaches use a time varying mean [222]Rn concentration approximation in a confined atmosphere [11&12].

The emanation coefficient "*E*" is calculated from the following equation [8]:-

$$E = \frac{N_{eq} - N^\circ}{N_{eq}} \text{--------- (1)}$$

Where Neq is the specific counts selected from peaks of [214]Pb and [214]Bi in equilibrium condition; and *N0* is the number of counts corresponding to *Neq* in initial condition.

Based on the mentioned modes of Rn gas in the soil matrix, there are three probabilities for the existing the radon atoms in the soil where:-

1- Emanation of radon from radium nuclides without transport,
2- Emanation and transport of radon through the grain pores,
3- Emanation, transport and exhalation of radon to the adjacent atmosphere,

The activity concentration of Radium in soil, in the absence of radon transport can be calculated as [13&14] :-

$$C_{Ra-226,228} = \frac{C_{Rn-222,220}}{f * \rho_s * \epsilon^{-1} * (1 - \epsilon)} * (\mu[k_T - 1] + 1) - - - -(2)$$

Where
CRn-222,220 : is the radon, thoron concentration in soil (Bq/m^3)

CRa-226,228: is the activity concn. in dry mass of ^{226}Ra,^{228}Ra in soil (Bq/kg),

f: is soil emanation factor: radon (0.2) and thoron (0.1),

ρ_s :is the density of soil, (kgm^{-3}),

ϵ : is the porosity, (0.25),

µ: is the porosity fraction that is water filled and is zero if the soil is dry

k_T : is the radon partition coefficient between water and air phases and if the soil samples are dried before measurement, then m is zero.

By normal gamma spectrometry where the calculation is fully based on the emanation of radon gas from radium and assuming complete homogeneous distribution of the decayed atoms on the soil matrix of the sample, the activity concentration of radium (Bq/kg) is calculated through the following equation [15]:-

$$A_{Ra} = \frac{\text{Net count}_i}{\epsilon * T * m * B.R.} - - - - (3)$$

Where

Net count$_i$: is the net count under peak area of radon progeny gamma line (i)

ε: is the efficiency of gamma spectrometry

T : is the total time of counting

B.R. : is Branching ratio of the selected gamma line

m: is the mass of the sample (kg)

This work is highlighting the effect of the other two radon modes in the soil matrix (namely, the transport and the exhalation), where the radon atoms transferred to the adjacent atmosphere and the decayed progeny is deposited on the surface of the sample, which leads to a non-uniform distribution of the atoms on the soil matrix of the sample. The absolute efficiency of the gamma detector was estimated for two assumptions i) uniform distribution of radon progeny in the soil matrix and ii) deposition of radon progeny on the surface of the soil sample. The study was carried out using the general Mont Carlo Code MCNP5 for the two assumptions considering the same properties of the soil sample as well as the same setup configuration of the modeled problem.

I. Material and methods

The effect of different distributions of radon daughters in soil samples on the absolute efficiency of an HPGe detector was studied. The study was carried out on different soil samples using the MCNP5 general MC Code [16&17].

- ### *Modeled Samples*

Three different samples were modeled in this study. The sample characteristics were assumed to be identical except for material composition and density. The model was applied to the IAEA reference materials RGU, 375 and soil 6. The activity concentrations of ^{226}Ra in the selected samples as well as their densities are illustrated in table (1) [18&19]. All samples were assumed to have the same volume (70 cm^3), and kept in polyethylene containers with the same shape and dimensions. The radon daughters that were considered in this study are ^{214}Pb and ^{214}Bi with corresponding gamma energy groups of 242.0, 295.0, 352.0 ,768.7keV and 609.0 ,665.4,1120.0, 1764.0 keV, respectively. The study includes two assumptions where; i) there is a homogenous distribution of the decayed isotopes on the whole volume of the sample (referred to volume source) and ii) there is a non-uniform distribution of the decayed isotopes where they will remain on the surface of the sample (referred as surface source).

- ### *Monte Carlo Simulation*

Monte Carlo Calculations were performed using the MCNP5 Code to calculate the absolute full energy peak efficiency for an HPGe detector. The calculations were performed to investigate the effect of radon daughters' distribution on the accuracy of measurements. The samples were modeled according to the above mentioned assumptions. The material compositions were modeled using material cards based on data given in the references [19&20]. The fine details of the sample container and detector type and model as given in figures 1 and 2 are described elsewhere [21]. Source definition cards (SDEF) were specified to describe two cases i) volume source for which the modeled isotopes were assumed to be uniformly distributed over the whole sample volume and ii) surface source for which the isotopes were assumed to be distribute on a very thin layer on the surface of the sample. The particle display feature in the MCNP5 code is used to illustrate the two cases as shown in figure 1. The number of histories was chosen (1E7) to keep the uncertainty in MC calculations always less than 0.5%.

Table (1) : The activity concentration of ^{226}Ra in IAEA reference materials

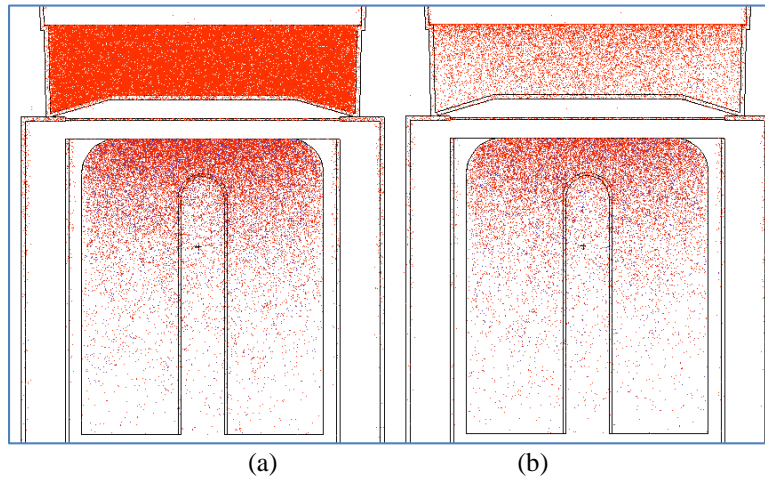| Sample Name	Density (Bq/kg)	Type	^{226}Ra Activity (Bq/kg)
RGU	1.23	Uranium ore	857.0E+3
Soil -6	1.19	Soil	79.92±
IAEA-375	1.28	Sediment	20.0

Figure 1. Particle display illustrating (a) volume source and (b) surface sources

II. RESULTS AND DISCUSSION

The difference in the efficiency of gamma spectrometer based on HPGe detector due to the distribution of radon progenies was carried out. Figures 2,3&4 illustrate the differences in the detector efficiency depending on the volume source and surface source for the selected reference samples .
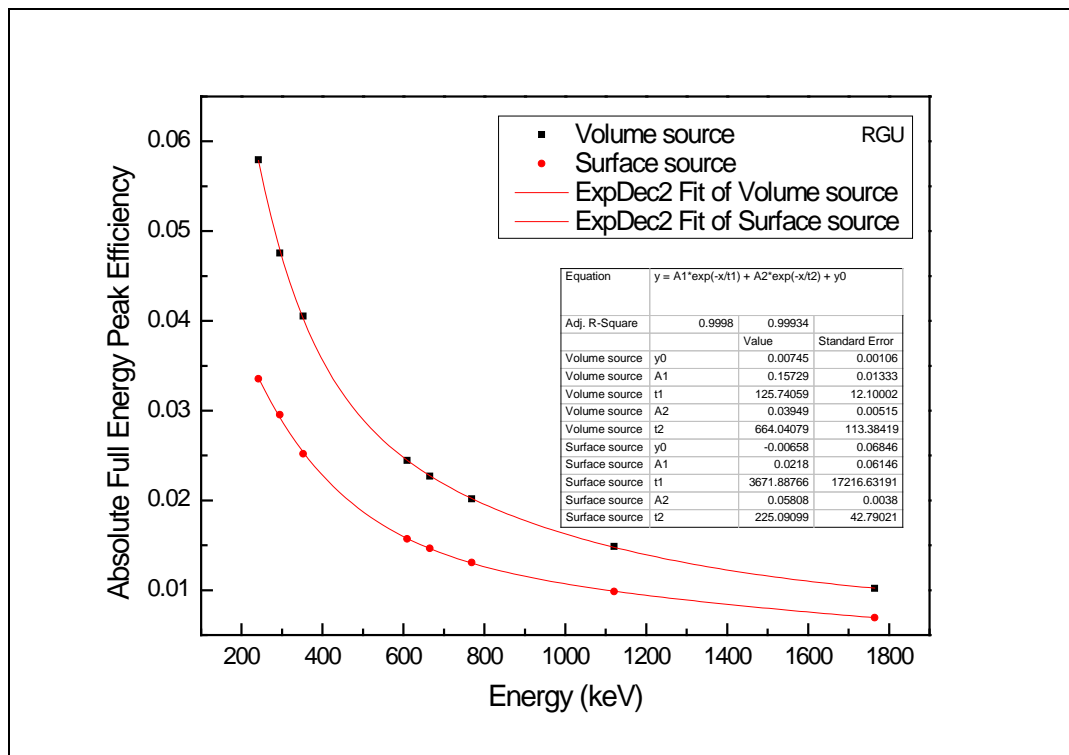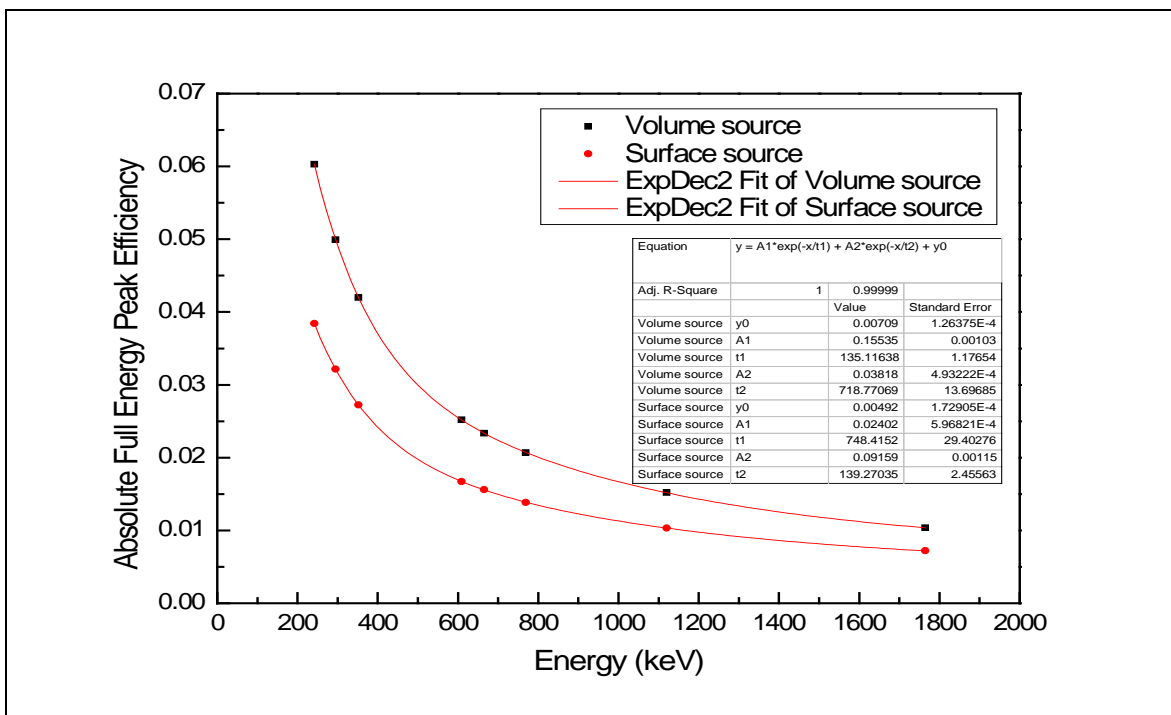

Figure 2. The detector efficiency differences in RGU-1

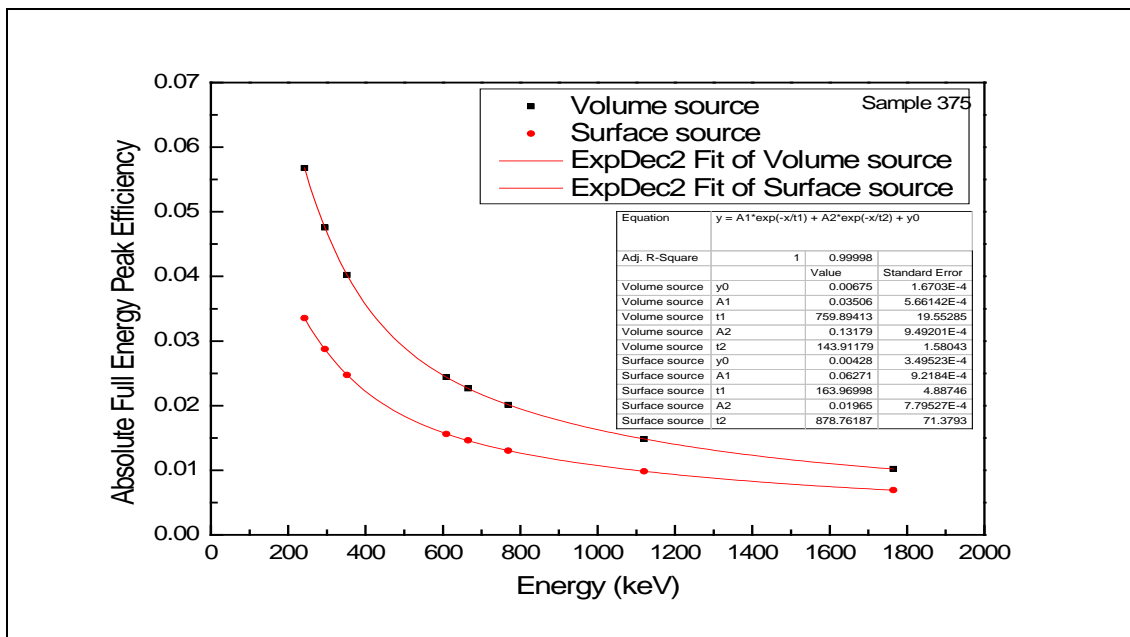**Figure 3. The detector efficiency differences in Soil-6**

Figure 4: The detector efficiency differences in IAEA-375

The average differences in the detector efficiency were found to be 36.23% (32.1%-42.1%), 33.7% (30.6% -36.2%) and 36.4% (32.1%- 40.9%) based on the data of RGU-1, soil-6 and IAEA-375 respectively.

It is clear from the three figures that the same trend is repeated for which the maximum efficiency differences occurs at low gamma energies, while they reduce gradually as energies increase. Three essential factors could effectively contribute to such differences. The fractional solid angle of the source subtended by the detector is the main factor [15&22]. An estimation of the differences in such geometry factor showed a value of about 29%. The other two factors, namely, material composition and density are responsible for the rest of differences.

In our study the effect of material composition and density are constant for each sample while the effect of the distribution of radon daughters on the soil matrix is examined.

For surface source the gamma self-attenuation is expected to be relatively high in comparison with volume source, since the all gamma rays have to cross the whole thickness of the sample before falling on the detector. The lower the energy of gamma rays, the higher the self-attenuation effect as it is clear from the figures.

Only two extreme cases were considered in this investigation, in which all the daughter nuclides of the emanated radon particles are either distributed homogeneously in the soil matrix, or they distributed on the surface of the sample due to complete exhale of emanated radon to the adjacent atmosphere. The results showed the importance of taking the emanation coefficient and exhalation flux calculations into consideration while performing gamma ray measurements on samples containing radon and radon daughters.

III.   CONCLUSION

Modeling of the two assumption related to the behavior of radon gas on the soil matrix was carried out where there is a homogeneous distribution of the radon decay products on the soil matrix based on the emanation process of radon from radium atoms (referred as volume source), and the other is a non-uniform distribution of radon's progeny on the soil matrix based on the exhalation of radon gas from the soil matrix to the sample volume's atmosphere and re-deposition of the radon progeny on the surface of the soil (refereed as surface source). The results of the two assumptions showed that there are big differences in the absolute efficiency of the detector. The study highlighted that during the determination of ^{226}Ra through the short lived daughters of ^{222}Rn gas by gamma spectrometry; it is very important to take into considerations the behavior of radon emanation from the soil sample before measurements. Suggestion for using packed sample without volume space may increase the homogeneity of the nuclides on the soil matrix. Comprehensive study on the calculation of emanation coefficient and radon flux in soil samples and study their effect on the calculation of the detector efficiency is recommended.
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