Correction for radon distribution in solid/liquid and air phases in gamma-ray spectrometry

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ABSTRACT

The effect of radon diffusion and distribution between a ^{226}Ra matrix and the top air gap inside sample containers for gamma-ray spectrometry was studied. Containers filled at almost 100% or just 70% of total capacity yielded correction factors of about 7% and 20% respectively. Applying these correction factors allowed activity values calculated from ^{226}Ra or radon decay products to agree within 2%.

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1. Introduction

In 2009, the National Institute for Ionizing Radiation Metrology of the National Agency for New Technology, Energy and the Environment (ENEA-INMRI) participated in the IAEA-CU-2009-01 interlaboratory study for characterization of the IAEA-448 "^{226}Ra in soil from oil field" reference material that was further used for an IAEA Proficiency Test among the ALMERA (Analytical Laboratories for the Measurement of Environmental Radioactivity) network laboratories. The objective of the characterization was the determination of ^{226}Ra activity concentration. Measurements at ENEA-INMRI were based on direct gamma-ray spectrometry of both ^{226}Ra and ^{222}Rn short lived decay products (^{214}Pb and ^{214}Bi).

The soil sample was dried and sealed in a cylindrical Teflon container. A reference source was prepared in the same geometry using a standard ^{226}Ra liquid solution. In both cases 1 month time (30 d) was allowed for secular equilibrium to be reached and care was taken to fill the containers as much as possible. Despite this, a 1.4 mm air gap was left between the top surface of each material and the inner lid of the container. The sources were measured on a HPGe gamma-ray spectrometer and the ^{226}Ra, ^{214}Pb, and ^{214}Bi activities in the soil were obtained using mainly the gamma-ray emissions at 186.2, 351.9, and 609.3 keV respectively, with efficiency values obtained directly from the ^{226}Ra liquid reference source (nuclide efficiency calibration). ^{235}U peaks were not detected in the soil spectrum; hence no correction was applied to the 186.2 keV peak for ^{226}Ra/^{235}U interference. Apparent disequilibrium between ^{226}Ra and ^{214}Pb, ^{214}Bi was found in the results for the soil sample, with about 10% overestimation of the two decay products with respect to the parent ^{226}Ra. The reason

for this effect was identified in a different distribution of radon gas between the sample liquid/solid phase (both reference solution and soil samples) and air phase of the top air gap.

A systematic investigation was then started with the aim to develop a procedure for correction of radon diffusion/distribution, inside the container, between matrix and top air volume for different matrix volumes. This paper describes the proposed radon diffusion model, the tools especially developed for calculation of the radon diffusion correction and the ENEA-INMRI results for the IAEA-448 soil reference material characterization.

Uncertainties are reported as standard uncertainty components or combined standard uncertainties ($k=1$).

2. Model

With reference to Fig. 1, a cylindrical sample container, internal volume V, is filled with a ^{226}Ra-matrix of activity A up to a filling volume (F.V.) V_2, leaving a top air gap of volume V_1. Under the assumption of a radon-tight container the ^{222}Rn, generated by ^{226}Ra decay, reaches secular equilibrium with its parent and its total activity will reach 99.57% of the activity value, A, in 1 month (30 d). While ^{226}Ra remains in the original matrix, radon, thanks to its high permeability and diffusivity, tends to escape from the matrix to the air gap. As a result, short lived radon decay products (^{214}Pb and ^{214}Bi) are generated not only in the matrix but also in the air gap. From here, in the stationary state, they are presumably adsorbed on the surfaces in contact with the air gap. As a consequence, the source geometry for ^{226}Ra will differ from that for ^{214}Pb and ^{214}Bi.

Then, for radon and its decay products in volumes 1 and 2 we have

$$A = A_1 + A_2, \quad V = V_1 + V_2. \tag{1}$$

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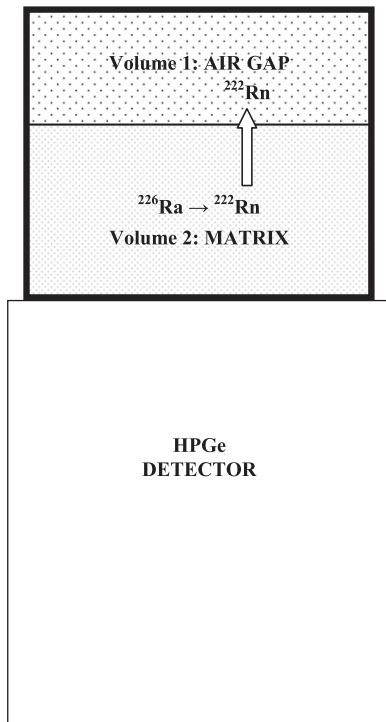E-mail address: pierino.defelice@enea.it (P. De Felice).

Fig. 1. Scheme of a sample container in which a ^{226}Ra matrix is put in the bottom and an air gap is left on top. Radon generated from ^{226}Ra decay diffuses through the matrix and fills the air gap.

The radon activity concentrations, $C_1 = A_1/V_1$ and $C_2 = A_2/V_2$, in the matrix and air gap respectively, are related through $\zeta = C_1/C_2$, the radon distribution coefficient between air and matrix. For a liquid sample ζ corresponds to the inverse of the radon solubility, while for soil it has to be derived from radon diffusion coefficients and soil granulometry. Then, the fractions of the radon activity in the air gap and in the matrix, $\eta_1 = A_1/A$ and $\eta_2 = A_2/A$, are given by the following expressions:

$$\eta_1 = \frac{\zeta c}{1+\zeta c}, \quad \eta_2 = \frac{1}{1+\zeta c}, \tag{2}$$

where $c = V_1/V_2$. When the container is positioned on an HPGe detector for spectrum acquisition, the efficiency ε_1 for detecting a photon emitted from V_1 will be different from the efficiency ε_2 for detecting a photon emitted from V_2 and the total count rate, R, will be $R = A_1\varepsilon_1 + A_2\varepsilon_2$, where the photon emission probabilities have been incorporated in the efficiencies (nuclide efficiency). The effective counting efficiency, ε_{eff}, will be

$$\varepsilon_{eff} \equiv \frac{R}{A} = [\eta_2 + (1-\eta_2)k]\varepsilon_2, \tag{3}$$

where $k = \varepsilon_1/\varepsilon_2$ is the efficiency ratio.

In gamma-ray spectrometry, where sample measurements are carried out with reference to calibration sources, the above described effect can occur both for calibration source and samples and Eq. (2) can be written in both cases. Applying the efficiency transfer approach, the effective efficiency for the sample, ε_{eff}^s, can be obtained from the effective efficiency for the calibration
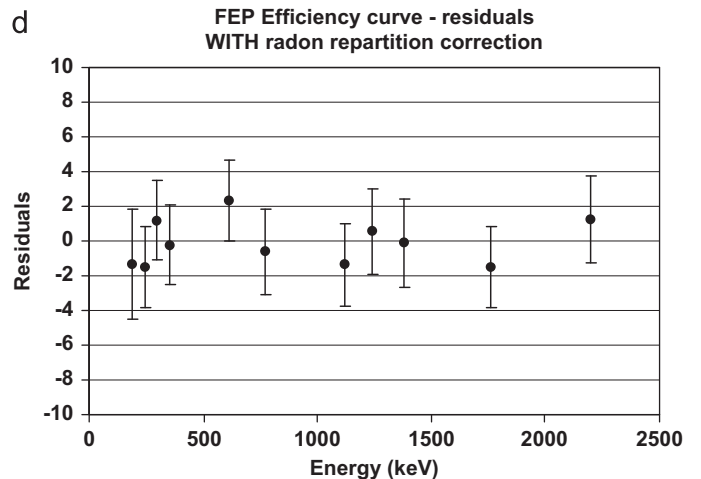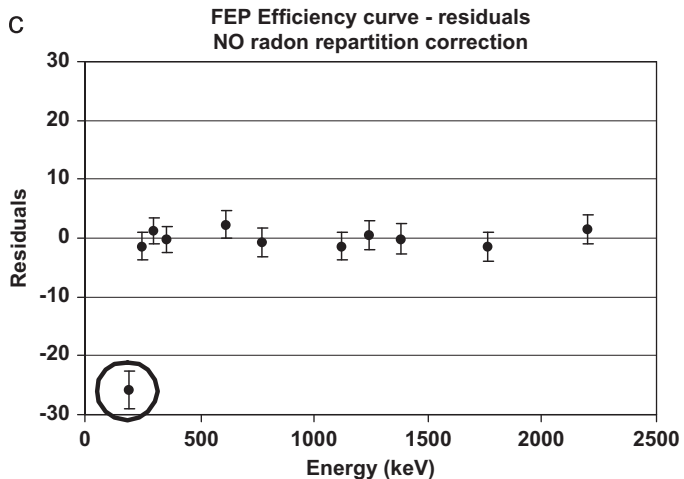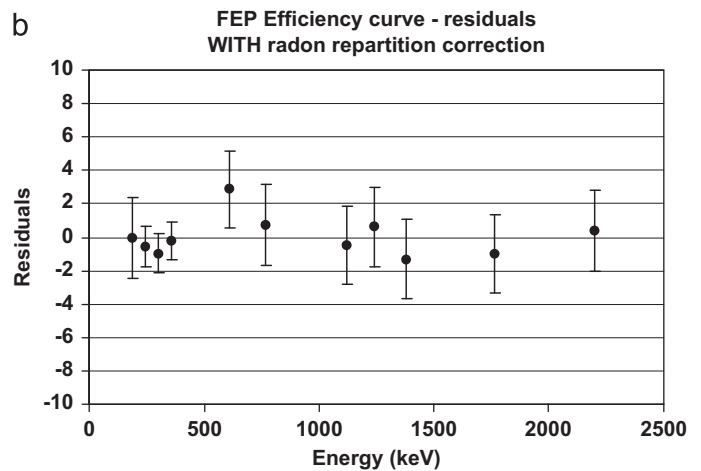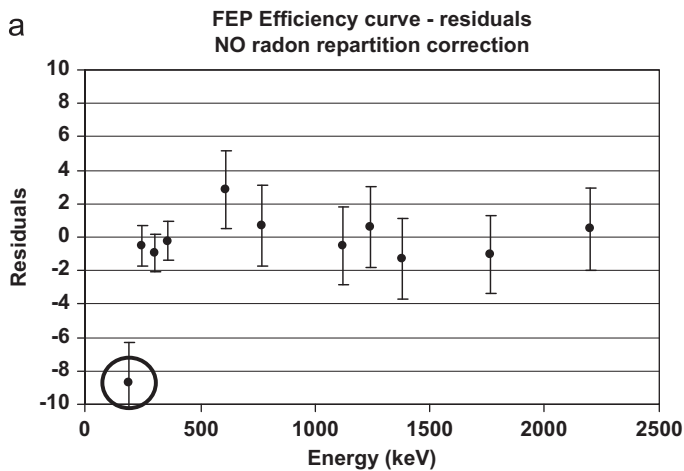

Fig. 2. Residuals of FEP efficiency values interpolation before and after correction for radon diffusion/distribution. Figures (a) and (b) refer to 100% F.V. while figures (c) and (d) refer to 70% F.V.