Journal of Radioanalytical and Nuclear Chemistry, Vol. 269, No.1 (2006) 141–145

Sample geometry and efficiency determination of bremsstrahlung radiation of ^{90}Sr on gamma detection systems

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(Received January 4, 2006)

The bremsstrahlung radiation energy spectra were produced by hard beta-emitters the ^{90}Sr–^{90}Y contaminated tea sample sources placed in a copper cylinder (cylinder counting geometry) and encapsulated in two Cu discs (sandwich counting geometry). These energy spectra were directly measured by using two separate gamma-ray spectrometers with a coaxial 110% efficient HpGe detector and a 110 cm^3 HpGe well-type detector. The minimum detectable activity and the absolute efficiency of beta-activity for the sandwich and cylinder geometries were found to be 23 Bq·kg^{-1} and (1.67±0.04)% and 55 Bq·kg^{-1} and (2.61±0.05)%, respectively. These results indicate that the bremsstrahlung radiation counting method can be applied to some environmental studies when high efficient HpGe detectors, especially well-type HpGe are used.

Introduction

It is known that ^{90}Sr is one of the most hazardous components of long-lived fission products. It is produced essentially by the ^{235}U and ^{239}Pu fission reaction and is released to the environment by global fallout following atmospheric nuclear explosions, by nuclear waste discharges and fallout from nuclear accidents. Owing to its chemical and biochemical similarities to calcium, ^{90}Sr tends to deposit in bone and blood-forming tissues. Further, ^{90}Sr is easily assimilated by animal and plant tissues and incorporated in the food chain and makes a remarkable contribution to internal dose since ^{90}Sr is a pure beta-emitter ($E_{\beta max} = 0.546$ MeV) and characterized by a long physical half-life ($T_{1/2} = 28.79$ y) and biological half-life (about 7 y), which is also in secular equilibrium with ^{90}Y the decay product ($E_{\beta max} = 2.228$ MeV, $T_{1/2} = 64$ h).

After the Chernobyl nuclear accident, analysts needed a useful and suitable method for a rapid ^{90}Sr determination in environmental and food samples. Recently published many techniques in the literature related to chemical separation[1–4] and determination of radiostrontium using Cherenkov radiation counting of ^{90}Sr's decay product ^{90}Y accelerate the analysis dramatically.[5–9] Additionally, the bremsstrahlung radiation produced by high energy beta particles from ^{90}Sr–^{90}Y decay, which earlier known and applied direct beta-measurements to determine in vivo whole-body activities of ^{90}Sr might be used as a pure instrumental method[10] for the determination of ^{90}Sr. The possibility of application of bremsstrahlung radiation from a commercially available solid ^{90}Sr–^{90}Y beta-source to some environmental studies has been described by MIETELSKI et al.[11,12]

The aim of the present work was to investigate the influence of the detector parameters (detector type and its active crystal volume) and sample counting geometries on the efficiency determination of bremsstrahlung radiation of ^{90}Sr. Two separate gamma-ray spectrometers with a coaxial 110% efficient HpGe detector for sandwich counting geometry and a 110 cm^3 HpGe well type detector for cylinder counting geometry were used to obtain an experimentally evidence on the selection criteria of the sample geometry together with a suitable detector type and its active volume.

Experimental

Five g tea samples weighted with a balance having an accuracy of 0.1% were used. Each of five samples was contaminated with a different activity of 62.2, 124.4, 186.6, 248.8, 466.5 Bq (as the sum of ^{90}Sr and ^{90}Y activities) using a ^{90}Sr standard solution obtained from Isotope Products Inc. traceable to NIST. Samples were separately ashed at 450 °C in a furnace, and finally formed the pellet by applying 15 ton/inch.^{2}

Two commercially available high-resolution gamma-spectrometry systems were used, in turn, in the measurements of the energy spectra obtained from the bremsstrahlung. One of the counting systems was equipped with a p-type HpGe well (GWL-110220) with total active volume of 110 cm^3 manufactured by EG&G Ortec Inc. The crystal size of HpGe well detector was 5.48 cm in diameter and 6.11 cm in length. The detector has 4.0 cm in active well depth and 1.6 cm in well inside diameter. Its measured energy resolution was 2.07 keV at 1332.5 keV of ^{60}Co and of 1.29 keV at 122 keV of ^{57}Co. The detector was shielded by lead lined with cadmium and copper on sides. However, the top surface of the shield is only covered by a 3 mm steel without lead. In the HpGe well detector system, the preamplifier signal is processed through a Spectroscopy Amplifier and an ADCAM Multichannel Buffer interfaced to a personal computer for the function of data

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0236–5731/USD 20.00

Akadémiai Kiadó, Budapest
Springer, Dordrecht

acquisition/storage and display/analysis by using a software package named OMNIGAM.

The other counting system was equipped with a coaxial p-type HpGe detector (GC11021) with an active volume of 451 cm^3 manufactured by Canberra Inc. The HpGe detector has a relative efficiency of 110%, an energy resolution of 2.1 keV at 1332.5 keV of ^{60}Co and of 1.3 keV at 122 keV of ^{57}Co, and a peak-to-Compton ratio of 85 : 1. For gamma-ray shielding a front opening split-top shield (Canberra Model 767) was used to reduce background. It features 10 cm lead thickness, which is jacketed by a 9.5 mm steel outer housing. The graded liner comprised 1 mm thick tin layer and 1.5 mm thick Cu layer to prevent interference by lead X-rays. To minimize scattered radiation from the shield, the detector was centered in it. The detector was interfaced to the DSA-1000 Digital Spectrum Analyzer which was a full featured 16 K channel multichannel analyzer on advanced digital signal processing techniques (DSP). DSA-100 operates through Genie-2000 gamma-spectroscopy software including peak searching, peak evaluation, energy/efficiency calculation mode, nuclide identification and etc. The end-caps of both HpGe detectors are made of aluminum.

Each of the ashed tea samples contaminated with different activities as powder was filled in a cylindrical Cu tube of 1.5 cm in external diameter, 5 cm in length and 2 mm in wall thickness, then the sample tube was placed in the HpGe well type detector (cylinder counting geometry shown in Fig. 1a). Each of the ashed tea samples in pellet form was sandwiched between two Cu discs of 1 mm thickness and 4 cm of diameter, and then the samples were put directly on the coaxial HpGe detector end-cap (sandwich counting geometry shown in Fig. 1b).

The predetermined counting time for each measurement was 6000 seconds and it was enough to obtain good counting statistics. Thus, the brems-strahlung radiation energy spectra were obtained for the cylinder counting and the sandwich counting geometries. The background spectra were also collected separately for both geometries. Each bremsstrahlung radiation energy spectra was corrected for the background in the regions of interest (ROI).

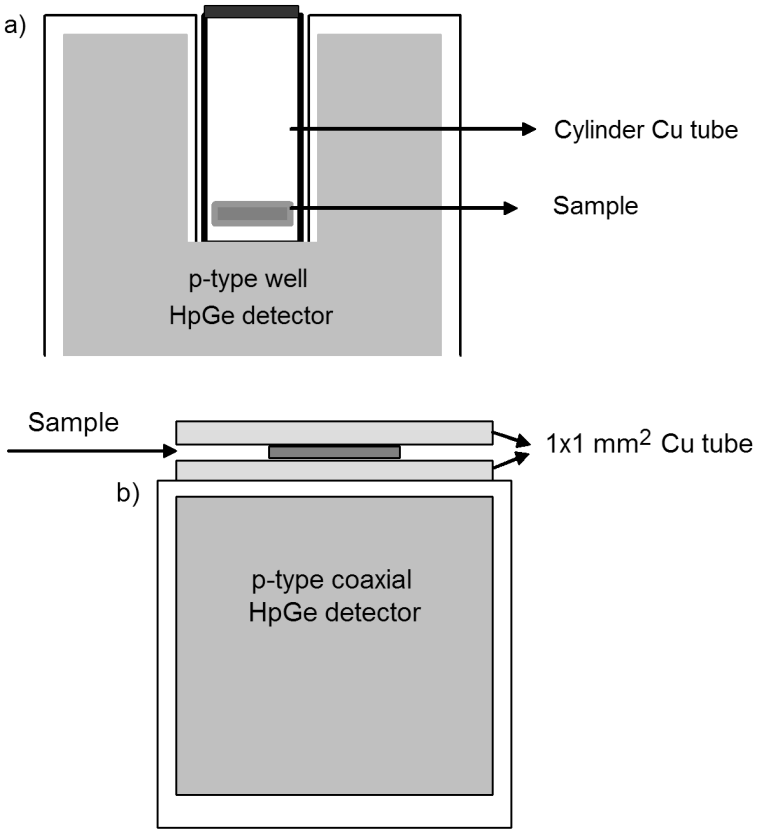

Fig. 1. Schematic representation of the experimental geometry: (a) cylinder counting geometry for the HpGe well type detector and (b) sandwich counting geometry for the coaxial HpGe detector

Results and discussion

Two detectors with high efficiency, but having different counting geometries were used to demonstrate that the present method is applicable to the determination of ^{90}Sr contained in samples. For this purpose, the bremsstrahlung radiation energy spectra of the equilibrated powder sample sources of ^{90}Sr–^{90}Y with 62.2, 124.4, 186.6, 248.8, 466.5 Bq of total activity for the cylinder counting geometry were measured by using the HpGe well type detector. The bremsstrahlung radiation energy spectra of the equilibrated sample sources of ^{90}Sr–^{90}Y in the form of pellets, with the same total activity mentioned above for the sandwich counting geometry were measured by using the coaxial HpGe detector. The ROIs of these net-bremsstrahlung radiation spectra were chosen in the energy range of 50 to 500 keV for the well HpGe and 60–490 keV for the coaxial HpGe, because of the maximum output of bremsstrahlung radiation from ^{90}Sr–^{90}Y beta-energies in Cu absorber. As an example, the considered ROI is shown in Fig. 2 for the sandwich counting geometry.

The results indicated that the count rate in the chosen ROIs increased with increasing activity of the sample for both detectors. As well, the count rate in the ROI is proportional to ^{90}Sr activity contained in samples. Thus, the net count rates obtained for the each of the ashed tea samples are plotted versus their ^{90}Sr–^{90}Y total activities of 62.2, 124.4, 186.6, 248.8, 466.5 Bq (shown in Fig. 3). As seen in Fig. 3, the net count rate in the ROI has a good linear relation with the activity of the sample. From the measured data in Table 1 for both counting geometries, the absolute efficiency for ROI specified for each of the detectors has been determined using linear regression. The slope of the fitted line obtained for the counting geometry of interest, as seen in Fig. 3, is interpreted as an absolute efficiency of measurement of beta-activity of ^{90}Sr. Hence, the absolute efficiency was found to be (1.667±0.036)% for the sandwich counting geometry with the 110% efficient coaxial HpGe and (2.611±0.053)% for the cylinder counting geometry with the 110 cm^3 HpGe well detector. However, the maximum response factor (2.611±0.053)% was achieved for the cylinder Cu tube with increasing its thickness up to 2 mm by using the HpGe well type detector. The uncertainty sources of the absolute efficiency were the peak statistical error for the chosen ROIs (0.98% for the coaxial HpGe and 0.44% for the HpGe well), the certified activity uncertainty (2%) for the standard solution used, and other estimated systematic errors (0.5%). Hence, the total uncertainties for absolute efficiency determination were calculated to be about 2% for both the coaxial HpGe detector and the HpGe well detector according to the known error propagation law.

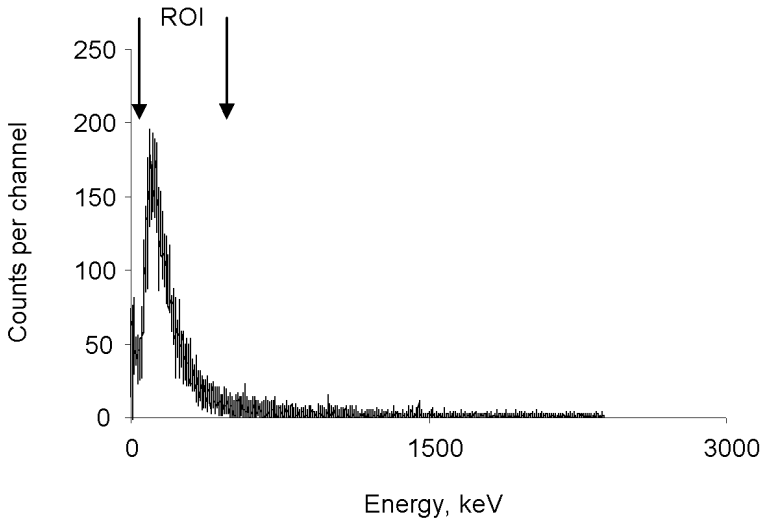

Fig. 2. Bremsstrahlung radiation energy spectra of the ^{90}Sr/^{90}Y activity (466.5 Bq) placed between 1 mm thick Cu discs, obtained with a 110% HpGe detector (ROI determined from 60 to 490 keV and background subtracted)

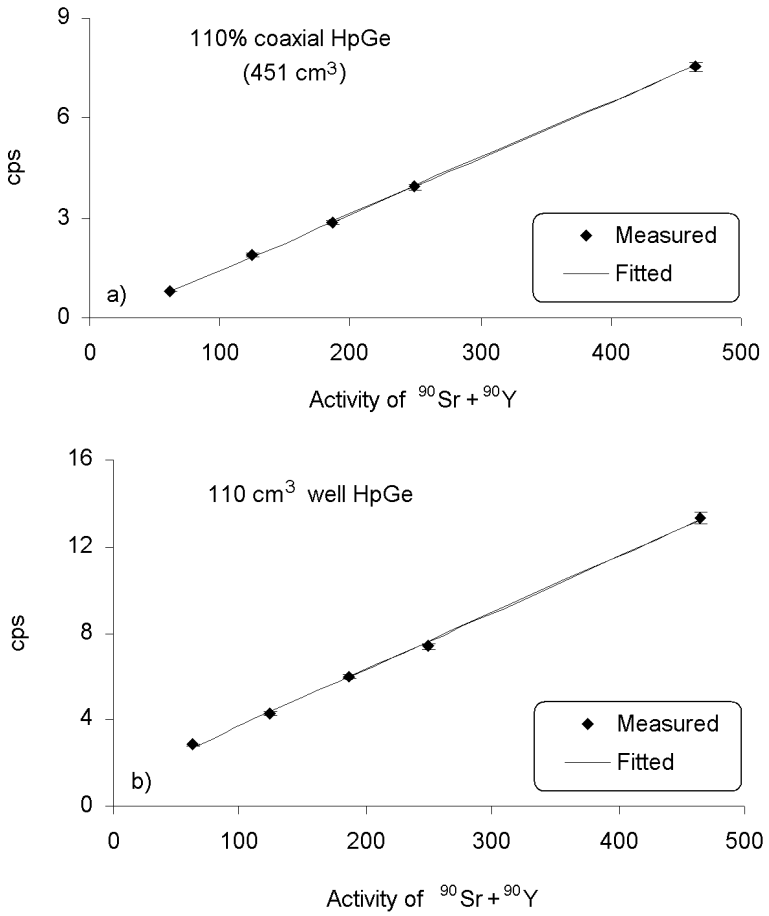

Fig. 3. Correlation plots of the bremsstrahlung intensity count rate versus ^{90}Sr activity for sandwich counting geometry (a) and cylinder counting geometry (b)

Table 1. Experimental data of the sadwich and cylinder counting geometries

Spectrometer	Geometry	ROI,[a] keV	Slope,[b] (counts/s)/Bq	Intercept, counts/s	Regression coefficient, R^2
Coaxial HpGe detector	Sandwich	60–490	0.01667 ± 0.00035	–0.21535	0.999
HpGe well type detector	Cylinder	50–500	0.02611 ± 0.00053	1.09403	0.999

[a] ROI: was chosen to be the same all samples.
[b] The slope is equal to the absolute efficiency for the measurement system.

It is well known that for the environmental applications, the minimum detectable activity (MDA) in a nuclear analytical method is another important parameter. Therefore, the MDA for determination of ^{90}Sr in the ashed tea samples in the present measurement systems is calculated as:

$$\mathrm{MDA} = \frac{\mathrm{LLD}}{\varepsilon \cdot t \cdot w} \qquad (1)$$

where MDA is in Bq·kg^{-1}, t is the measurement time in seconds, w is the weight of the sample in kg and LLD is the lower limit of detection, as defined by:[13]

$$\mathrm{LLD} = 2.706 + 4.653\sigma_{\mathrm{N_B}} \qquad (2)$$

where $\sigma_{\mathrm{N_B}}$ is the standard deviation of the background in the ROI and equals square root of the number of counts for the background spectrum. For a background measurement time of 4 days, the MDA of the presented counting geometries was calculated to be 23 Bq·kg^{-1} and 55 Bq·kg^{-1} for sandwich counting geometry and cylinder counting geometry, respectively. It was expected that the MDA for the cylinder counting geometry used for the HpGe well type detector would be lower than that of the sandwich counting geometry used for the coaxial HpGe detector. However, it is shown that the effectiveness of the home made shielding of the HpGe well type detector is poorer than that of

commercial shielding used for the coaxial HpGe detector. This implies that the HpGe well type detectors and cylinder counting geometry will still be useful for the determination of ^{90}Sr in the environmental samples by gamma-ray spectrometry.

Conclusions

It is concluded that the bremsstrahlung radiation counting method may be used for the determination of ^{90}Sr in real environmental samples both as sandwich counting geometry with Cu absorber using a high efficiency coaxial HpGe detector and as Cu-cylinder counting geometry using HpGe well type detector, especially in an emergency cases. It is worth noting that when a higher efficient HpGe well type detector with a larger crystal is used in measurement of bremsstrahlung radiation energy spectra for rapid ^{90}Sr determination, the cylinder counting geometry can be more useful and practical for environmental samples. However, some radiochemical separation procedures for the bremsstrahlung radiation counting method are still necessary to isolate ^{90}Sr from other beta-emitters such as ^{144}Ce, ^{137}Cs, ^{125}I, ^{99}Tc, ^{40}K, etc. present in a real sample because of the interference of these radionuclides to the ^{90}Sr to be measured.

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Turkish Atomic Energy Authority supported this work. The authors wish to thank Mr. Mustafa VURAL, researcher in SNRTC, for his technical assistance.

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