Development of a simultaneous evaluation method of radioactivity in soil and dose rate using CeBr₃ and SrI₂(Eu) scintillation detectors for environmental monitoring

Munehiko Kowatari*, Yoshihiko Tanimura*, Patrick Kesslerb, Stefan Neumaierb and Annette Roettgerb

aJapan Atomic Energy Agency, 2-4 Shirakata, Tokai-mura, Naka-gun, Ibaraki-ken, 319-1195, Japan; b Physikalisch-Technische Bundesanstalt, Bundesallee 100, Braunschweig, 28116, Germany

The simultaneous evaluation of dose rate and radioactivity using newly developed scintillation detectors, i.e. CeBr₃ and SrI₂(Eu) was investigated for environmental radiation measurement. Energy dependence and linearity of both detectors were verified by measurements in a low background laboratory operated by the German metrological institute and found to be applicable to environmental monitoring purposes. As a result of environmental gamma ray measurements in normal situation, evaluated air kerma rate and radioactivity in/on soil for 137Cs, 134Cs and 40K seemed consistent. Results also support that the method developed in this study is reliable and would be applicable to the environmental radiation measurement.

Keywords: environmental monitoring; CeBr₃ scintillation detector; SrI₂(Eu) scintillation detector; G(E) function method; Unfolding method; ultra-low background laboratory

1. Introduction

Identification and determination of radioactivity on soil in emergency situations is still an ongoing issue. The sophisticated in situ environmental measurement technique using a High Purity Ge (HP-Ge) semiconductor spectrometer [1,2] was not applicable for emergency situations where dose rates exceeded several tenths of μSv h⁻¹. More simplified and easy-to-handle equipment is preferable for emergency situations such as the KURAMA system [3].

The purpose of this study is to investigate the adaptability of newly developed scintillation detectors to radiation monitoring devices which are used for environmental radiation monitoring, particularly in emergency situations. To fulfill the purpose, the authors developed the methodology of the simultaneous evaluation of radiological information derived from the scintillation detectors. A series of in situ gamma ray measurements in the normal environment were conducted as a trial measurement. Then, discussion on applicability of the methodology to in situ radiation measurement in normal and emergency situations was made.

2. Material and methods

2.1. Scintillation detector and in situ environmental measurement

A 2”φ × 2” CeBr₃ and a 1”φ × 1”SrI₂(Eu) cylindrical scintillation detectors were employed for this study. This is because they have relatively lower self-contamination and similar energy resolution, compared with a LaBr₃(Ce) scintillation detector. In this study, air kerma rate and surface deposition density on soil were both evaluated by applying different methods to the same measured pulse height spectra. The so-called G(E) function [4] and unfolding methods were selected for evaluating air kerma rates. To determine radioactivity in soil and surface deposition density on soil, peak area method which are widely recognized for in situ radioactivity measurement and unfolding methods were both employed in this study for comparison.

Trial measurements of environmental radiation measurement using both detectors were performed in the same manner as the ICRU 53 [2] provides for the in situ radioactivity measurement. Each detector coupled with a personal-computer-driven (PC-driven) Multi-Channel Analyzer (MCA) was fixed at a height of 1.0 m from ground surface in an open and plain field in Tokai Research Branch, Japan Atomic Energy Agency (JAEA). Measuring time was set to 3600 s. Obtained pulse height spectra from the CeBr₃ and the SrI₂(Eu) scintillation detectors were then processed with above mentioned methods.

2.2. Air kerma rate

Air kerma rates in the environment were evaluated from the pulse height spectra from both the CeBr₃ and the SrI₂(Eu) scintillation detectors. The G(E) functions for each scintillation detector enable to directly estimate...
air kerma rate from measured pulse height spectrum. They were also evaluated from calculated response functions using EGS4 code [5]. The response matrices and appropriate unfolding code were prepared prior to environmental measurement, in order to apply the unfolding method to environmental radiation monitoring. Response matrices for both scintillation detectors were calculated using the Monte Carlo calculation code, MCNP-4C [6]. Photon fluence spectra from both detectors were obtained by unfolding the measured spectra using MAEXD [7] code. Air kerma rates were then estimated by multiplying the photon fluence rate with fluence-to-air-kerma conversion coefficients provided by ICRP 74 [8]. Prior to environmental radiation measurement, the unfolding and the G(E) function methods employed in this study were thoroughly verified in accredited underground gamma ray reference fields operated in German national metrological laboratory, Physikalisch-Technische Bundesanstalt (PTB) [9,10].

2.3. radio-activity in soil

For evaluating surface deposition densities on soil or radioactivity in soil, two different methods, i.e., the peak area method and the unfolding method were employed.

The surface deposition density on soil, $A_a$ (Bq cm$^{-2}$), or radioactivity concentration in soil, $A_m$ (Bq g$^{-1}$) are determined by dividing a net peak counting rate, $C_f$ (cps) by a peak counting rate per activity, $N_f/A_a$ (cps (Bq cm$^{-2}$)) or $N_f/A_m$ (cps (Bq g$^{-1}$)) as shown in the following equation.

$$A_a or A_m = \frac{C_f}{(N_f/A_a)} or \frac{C_f}{(N_f/A_m)}$$

The energy dependence of evaluated air kerma rate from both detectors was assessed and compared. In Figure 1, responses of each detector are depicted. All measurements in $^{241}Am$, $^{57}Co$, $^{137}Cs$ and $^{60}Co$ gamma ray reference fields were performed in the ultra-low background underground laboratory of PTB [9,10]. Air kerma rates for each reference field ranged between 34 and 84 nGy h$^{-1}$. Except for the results obtained in the $^{241}Am$ reference field, responses for each detector were evaluated to be within 10%. The results imply that reliable air kerma rate could be derived from pulse height spectra obtained in radiation fields with dose rates below 0.1 μSv h$^{-1}$.

For proper evaluation of air kerma rate from pulse height spectra, the linearity of evaluated air kerma rate was also investigated and shown in Figure 3.
rates indicate clear linearity from 10 to 80 nGy h\(^{-1}\) for \(^{137}\)Cs gamma ray. This also supports that the method introduced for dosimetry could enable to assess the variation of air kerma rates even in normal situation which dose rates are to be approximately several tenths of nSv h\(^{-1}\).

Table 1 shows the comparison of the intrinsic background (BG) obtained from both detectors. The intrinsic BG is caused by self-contamination of scintillation crystal and optical window inside detectors. Considering the difference of the crystal volume of each detector, less BG would be observed from the CeBr\(_3\) detector. The very low intrinsic BG of both detectors implies that the minimum detectable limit would be much less than 20 nSv h\(^{-1}\) which is sufficient to identify unforeseen radiological events.

### 3.2. Dosimetry in the environment

The air kerma rates were evaluated by applying the G(E) function and the unfolding methods to the same measured pulse height spectrum shown in Figure 4. Evaluated results were summarized and compared with each other in Table 2. The ambient dose equivalent rates were derived by applying G(E) function method to pulse height spectra obtained from each detector. Consistent air kerma rates were obtained from both methods from each detector. This supports that both methods are applicable to evaluate air kerma rate for environmental radiation monitoring in normal situation.

#### 3.3. determination of radioactivity

##### 3.3.1 \(^{137}\)Cs and \(^{134}\)Cs

In Table 3, surface deposition density on soil for \(^{137}\)Cs and \(^{134}\)Cs evaluated by the peak area and the unfolding methods were summarized. Surface deposition density on soil for radioactive Cs were also determined to be 3.2 Bq cm\(^{-2}\) and 0.44 Bq cm\(^{-2}\) for \(^{137}\)Cs and \(^{134}\)Cs, by a conventional in situ method with the HP-Ge detector. The evaluated surface deposition densities on soil for \(^{137}\)Cs by both methods were found to be in quite good agreement with each other. Results for \(^{134}\)Cs are also consistent, considering the difference in sensitivity of the CeBr\(_3\) and the SrI\(_2\)(Eu) detectors, overlapping interfering gamma rays due to less energy resolution and their accompanying uncertainties.

##### 3.3.2 Natural radionuclides

Table 3 showed comparison of radioactivity concentration in soil. Considering the purpose of in situ radiation measurement in emergency situation, there is no need to determine radioactivity for natural radionuclides. On the other hand, the developed method would make use of identification and quantification of radionuclides of interest if the influence of interfering natural radionuclides were to be properly eliminated.

In Table 3, results of the evaluated radioactivity concentrations in soil for natural radionuclides are listed.
Figure 4. Pulse height spectra of the CeBr₃ and the SrI₂(Eu) scintillation detectors measured in an open and plain field in Tokai, JAEA. Measurements were made at the height of 1.0 m above ground and the measuring time was 3600 s. Cesium 134 and 137 due to the Fukushima nuclear accident were clearly identified.

As shown in Figure 4, no clear peak can be identified which corresponds to gamma rays from ²⁰⁸Tl (2614 keV) in the spectrum from the SrI₂(Eu) detector. Radioactivity concentrations in soil for ⁴⁰K were estimated and found to be quite consistent with those obtained by in situ measurement using a HP-Ge detector (0.42 Bq g⁻¹) at the same open field at Tokai Research branch, JAEA.

Table 3. Comparison of surface deposition density on soil and radioactivity concentrations in soil obtained by the CeBr₃ and the SrI₂(Eu) scintillation detectors.

(a) CeBr₃ scintillation detector.

| Radio-nuclide | Peak area method (A) | Unfolding method (B) | B/A |
|---------------|----------------------|----------------------|-----|
| Artificial    |                      |                      |     |
| ¹³⁷Cs         | 3.09                 | 3.21                 | 1.04|
| ¹³⁴Cs         | 0.441                | 0.405                | 0.92|
| natural       | Radioactivity concentration in soil (Bq g⁻¹) | | |
| ⁴⁰K           | 0.429                | 0.438                | 1.02|
| ²⁰⁸Tl         | 0.020                | 0.017                | 0.82|

(b) SrI₂(Eu) scintillation detector.

| Radio-nuclide | Peak area method (A) | Unfolding method (B) | B/A |
|---------------|----------------------|----------------------|-----|
| Artificial    |                      |                      |     |
| ¹³⁷Cs         | 3.13                 | 3.19                 | 1.02|
| ¹³⁴Cs         | 0.473                | 0.454                | 0.96|
| natural       | Radioactivity concentration in soil (Bq g⁻¹) | | |
| ⁴⁰K           | 0.446                | 0.437                | 0.98|
| ²⁰⁸Tl         | -                    | -                    | -   |

From the results of determination of radioactivity in soil, proper radiation protection information can be properly drawn from the newly developed scintillation detectors by conducting in situ radiation measurement in normal situation.

3.4. Applicability of the simultaneous determination method using the CeBr₃ and/or SrI₂(Eu) scintillation detector to emergency radiation monitoring

From the series of investigation, the simultaneous determination method using the CeBr₃ and/or SrI₂(Eu) scintillation detectors coupled with the PC-driven MCA was confirmed to be reliable and would be applicable to normal radiation monitoring.

For emergency radiation monitoring, rapid and optimal provision of radiological information should be implemented. Not only dose equivalent rate information with time and location [3],[11], but radioactivity on soil of artificial radionuclides should also be monitored. The simultaneous determination method would enable to provide radiological information on both dose rate and radioactivity on soil, by performing the in situ radiation measurement using the CeBr₃ and/or SrI₂(Eu) scintillation detectors coupled with the PC-driven MCA.

With regard to applicability of the detector to high dose rate condition, i.e., > 10 μSv h⁻¹, the authors verified that the developed method using the 2"φ × 2" CeBr₃ scintillation detector could provide an accurate ambient dose equivalent rate of 10 μSv h⁻¹ and that the detector can work properly in the ¹³⁷Cs gamma ray calibration field whose ambient dose equivalent rate is approximately 100 μSv h⁻¹. This implies that in situ radiation measurement could be carried out even in high dose rate condition which is several tenths of μSv h⁻¹ by applying the CeBr₃ and/or SrI₂(Eu) scintillation detectors. The authors confirmed that improper pulse height spectra were obtained by the conventional HP-Ge detector in the area whose dose rate was greater than 40 μSv h⁻¹[12]. This is one of the advantages of the developed method. As shown in Figure 4, both the CeBr₃ and/or SrI₂(Eu) scintillation detectors exhibit a better energy resolution, compared to that of the conventional NaI(Tl) scintillation detector which are generally installed in monitoring stations [11]. The energy resolutions of both detectors are sufficient to identify ¹³⁴Cs and ¹³⁷Cs. Easy-to-handle equipment would be helpful to avoid excess exposure under high dose rate. From this viewpoint, the HP-Ge detector with cooling system is not available, because of the size of the system. As for the stability of the whole system, no peak shift could be found during BG measurements for two hours. This showed that the system has the sufficient stability for in situ radiation measurement.

In summary, the CeBr₃ and/or SrI₂(Eu) scintillation detectors are promising candidates for detectors which are applied to in situ radiation measurement in high dose rate condition, because of the availability in the high dose rate condition, better energy resolution, easy-to-handle system and sufficient stability of the system.
4. Conclusion

This study demonstrated an adaptability of newly developed CeBr$_3$ and SrI$_2$(Eu) scintillation detectors for environmental radioactivity in soil and air kerma rate measurements. By applying conventional peak area method for in situ radioactivity measurement to pulse height spectra obtained from two different detectors, evaluated radioactivity in soil were both considered to be consistent. For simultaneous determination of radioactivity in soil and air kerma rate from the same pulse height spectrum, an unfolding method was introduced in the study. Almost identical air kerma rates were obtained from each scintillation detector. Evaluated surface contamination densities on soil for $^{137}$Cs and $^{134}$Cs by both methods were found to be in agreement with each other, taking into account accompanying uncertainties. This also implies that the simultaneous determination method for dose rate and radioactivity would be applicable even under normal condition whose dose rate is approximately 0.1 μSv h$^{-1}$. By considering radiological emergency situation where dose rates exceed greater than several tenths of μSv h$^{-1}$, our proposed method coupled with both detectors can be a potential option which are applied to rapid and timely evaluation of dose rate and radioactivity concentration simultaneously.

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