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**ARTICLE**

# Development of a simultaneous evaluation method of radioactivity in soil and dose rate using $CeBr_3$ and $SrI_2(Eu)$ scintillation detectors for environmental monitoring

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The simultaneous evaluation of dose rate and radioactivity using newly developed scintillation detectors, i.e. $CeBr_3$ and $SrI_2(Eu)$ was invesigated for environmental radiation measurement. Energy dependence and linearity of both detectors were verified by measurements in a low backgrould laboratory operated by the German metrological insititute and found to be applicable to enviromental monitoring purposes. As a result of environmental gamma ray measurements in normal situation, evaluated air kerma rate and radioactivity in/on soil for $^{137}Cs$, $^{134}Cs$ and $^{40}K$ 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; $CeBr_3$ scintillation detector; $SrI_2(Eu)$ 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” $CeBr_3$ and a 1”φ × 1”$SrI_2(Eu)$ cylindrical scintillation detectors were employed for this study. This is because they have relatively lower self-contamination and similar energy resolution, compared with a $LaBr_3(Ce)$ 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 $CeBr_3$ and the $SrI_2(Eu)$ 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 $CeBr_3$ and the $SrI_2(Eu)$ scintillation detectors. The G(E) functions for each scintillation detector enable to directly estimate

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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 spectrum 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$^{-2}$)$^{-1}$) or $N_f/A_m$ (cps ((Bq g$^{-1}$)$^{-1}$), as shown the following equation.

$$A_a or A_m = {}^{C_f}\!/_{(N_f/A_a)} \, or \, {}^{C_f}\!/_{(N_f/A_m)}, \quad (1\text{-a})$$

and

$$(N_f/A_a) or (N_f/A_m) = \left(\frac{N_f}{F_\varphi} \times \varphi_{Aa}\right) or \left(\frac{N_f}{F_\varphi} \times \varphi_{Am}\right) \quad (1\text{-b})$$

For employing the peak area method, energy dependent peak counting rate per photon fluence rate from the ground surface, $N_f/F_\varphi$(cps (photons cm$^{-2}$ s$^{-1}$)$^{-1}$), so-called the peak efficiency for each detector is required and obtained by measurements and the Monte Carlo calculations. Results were shown in **Figure 1**.

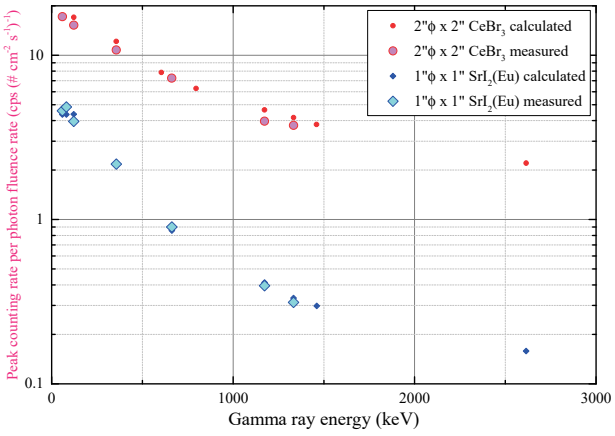


Figure 1. Comparison of the peak efficiency curve for the $CeBr_3$ and the $SrI_2$(Eu) scintillation detectors.

The peak efficiency were obtained by measuring pulse height spectra by gamma rays from point sources ($^{241}$Am, $^{152}$Eu, $^{133}$Ba, $^{60}$Co and $^{137}$Cs) and calculated using the Monte Carlo calculations using MCNP-4C [6]. Series of measurement were performed at the height of 1.0 m from the point source put on the floor. The height should be the same as trial measurement performed in an open field. Then, peak counting rate per activity, $N_f/A_a$ and $N_f/A_m$ were derived from multiplying $N_f/F_\varphi$ with photon fluence rate per unit surface deposition density, $\varphi_{Aa}$ (photons cm$^{-2}$ s$^{-1}$ (Bq cm$^{-2}$)$^{-1}$) and activity concentration in soil, $\varphi_{Am}$ (photons cm$^{-2}$ s$^{-1}$ (Bq g$^{-1}$)$^{-1}$). Both $\varphi_{Aa}$ and $\varphi_{Am}$ are independent of the detector used and were calculated analytically or taken from the Table A.1 in ICRU 53 [2].

$$A_a or A_m = {}^{F_\varphi}\!/_{\varphi_{Aa}} \, or \, {}^{F_\varphi}\!/_{\varphi_{Am}} \quad (2)$$

This study also demonstrates how to determine the surface deposition densities on soil by directly evaluating the photon fluence rate, $F_\varphi$(photons cm$^{-2}$ s$^{-1}$) at 1.0 m height from the surface. The $F_\varphi$ corresponding to each radioactive cesium and natural radionuclides such as $^{40}$K and $^{208}$Tl was determined by the unfolding method introduced in this study. According to the equation (2), $A_a$ (Bq cm$^{-2}$) and $A_m$ (Bq g$^{-1}$) were also evaluated by dividing $F_\varphi$ for each radionuclide by the photon fluence rate per unit activity, $\varphi_{Aa}$ and $\varphi_{Am}$. To evaluate the surface deposition density on soil for $^{137}$Cs and $^{134}$Cs, the migration of soil from the ground surface and the depth profile of concentration of radioactive Cs in soil should be accounted for. The parameter called the relaxation mass per unit area, β (g cm$^{-2}$), is introduced, in order to express the vertical distribution of radioactive Cs in soil. According to the recommendation of the ICRU 53 [2], β was set to 3. This is because almost six years have passed since initial deposition of radionuclides took place at the measurement date.

## 3. Results and discussion

### *3.1. Energy dependence and linearity check of scintillation detectors*

The energy dependence of evaluated air kerma rate from both detectors was assessed and compared. In **Figure 2**, 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**. Air kerma

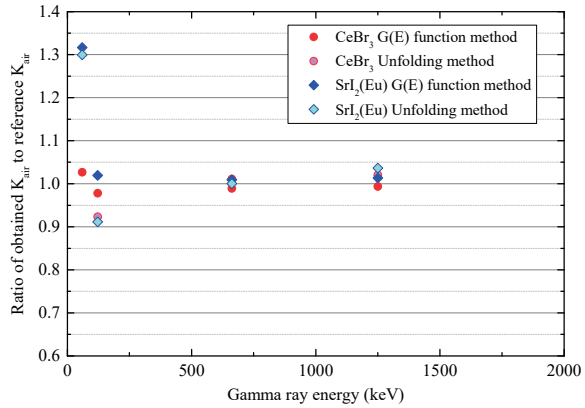


Figure 2. Comparison of the energy dependence of the $CeBr_3$ and the $SrI_2(Eu)$ scintillation detectors. air kerma rate was evaluated by two different methods, i.e., G(E) function and unfolding methods.

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 $h^{-1}$.

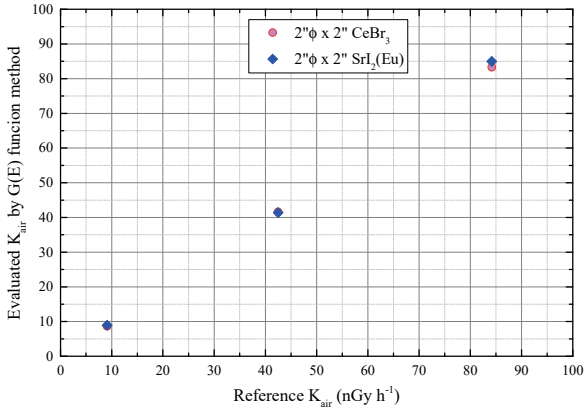


Figure 3. Linearity check for air kerma rate evaluated from pulse height spectra obtained by each scintillation detector.

**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 also evaluated by the G(E) function method, for comparison with those obtained by a commercial NaI(Tl) scintillation survey-meter.

Table 1. Comparison of the intrinsic BG of scintillation detectors obtained in the underground laboratory. Ambient dose equivalent rates were derived by applying G(E) function method to pulse height spectra obtained from each detector.

| Detector | $CeBr_3$ (2"φ x 2") | $SrI_2(Eu)$ (1"φ x 1") |
|---|---|---|
| intrinsic BG (nGy $h^{-1}$) | 6.1 | 5.3 |

Table 2. Comparison of ambient dose equivalent rates, $H^*(10)$ and air kerma rates, $K_{air}$ obtained by the $CeBr_3$ and the $SrI_2(Eu)$ scintillation detectors.

| Scintillation detector | $H^*(10)$ by NaI(Tl) survey meter (nSv $h^{-1}$) | $H^*(10)$ by G(E) function method (nSv $h^{-1}$) | $K_{air}$ by G(E) function method (nGy $h^{-1}$) | $K_{air}$ by unfolding method (nGy $h^{-1}$) |
|---|---|---|---|---|
| $CeBr_3$ | 109 | 112 | 92.7 | 94.7 |
| $SrI_2(Eu)$ | 102 | 107 | 85.0 | 84.1 |

* All measurements were conducted at the field in the Nuclear Science Research Institute (Tokai), JAEA

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.

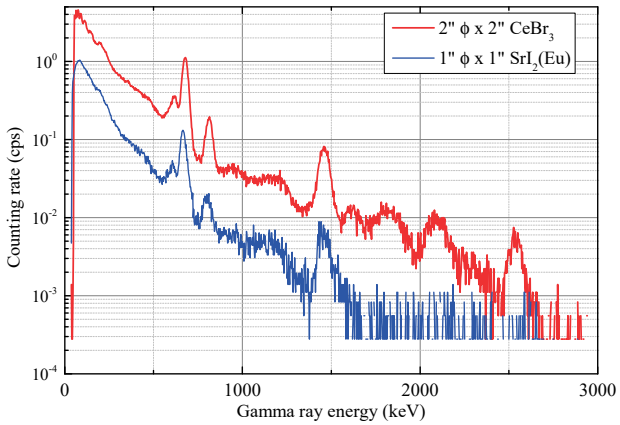


Figure 4. Pulse height spectra of the $CeBr_3$ and the $SrI_2(Eu)$ scintillation detectors measured in an open and plain field in Tokai, JAEA. Measurements were made at the height of 1.0 m above ground and the measuring time was 3600 s. Cesium 134 and 137 due to the Fukushima nuclear accident were clearly identified.

As shown in **Figure 4**, no clear peak can be identified which corresponds to gamma rays from $^{208}Tl$ (2614 keV) in the spectrum from the $SrI_2(Eu)$ detector. Radioactivity concentrations in soil for $^{40}K$ 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 $CeBr_3$ and the $SrI_2(Eu)$ scintillation detectors.

(a) $CeBr_3$ scintillation detector.

| Radio-nuclide | Peak area method (A) | Unfolding method (B) | B/A |
|---|---|---|---|
| Artificial | Surface contamination density on soil (Bq $cm^{-2}$) | | |
| $^{137}Cs$ | 3.09 | 3.21 | 1.04 |
| $^{134}Cs$ | 0.441 | 0.405 | 0.92 |
| natural | Radioactivity concentration in soil (Bq $g^{-1}$) | | |
| $^{40}K$ | 0.429 | 0.438 | 1.02 |
| $^{208}Tl$ | 0.020 | 0.017 | 0.82 |

(b) $SrI_2(Eu)$ scintillation detector.

| Radio-nuclide | Peak area method (A) | Unfolding method (B) | B/A |
|---|---|---|---|
| Artificial | Surface contamination density on soil (Bq $cm^{-2}$) | | |
| $^{137}Cs$ | 3.13 | 3.19 | 1.02 |
| $^{134}Cs$ | 0.473 | 0.454 | 0.96 |
| natural | Radioactivity concentration in soil (Bq $g^{-1}$) | | |
| $^{40}K$ | 0.446 | 0.437 | 0.98 |
| $^{208}Tl$ | - | - | - |

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 $CeBr_3$ and/or $SrI_2(Eu)$ scintillation detector to emergency radiation monitoring*

From the series of investigation, the simultaneous determination method using the $CeBr_3$ and/or $SrI_2(Eu)$ 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 $CeBr_3$ and/or $SrI_2(Eu)$ 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" $CeBr_3$ scintillation detector could provide an accurate ambient dose equivalent rate of 10 μSv $h^{-1}$ and that the detector can work properly in the $^{137}Cs$ 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 $CeBr_3$ and/or $SrI_2(Eu)$ 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 $CeBr_3$ and/or $SrI_2(Eu)$ 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 $^{134}Cs$ and $^{137}Cs$. 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 $CeBr_3$ and/or $SrI_2(Eu)$ 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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