Development of real time $^{90}$Sr Counter applying Cherenkov light detection

H. Ito*, S. Iijima, S. Han, H. Kawai, S. Kodama, D. Kumogoshi, K. Mase, M. Tabata
Department of Physics, Graduate School of Science Chiba University, Chiba, Chiba, Japan
E-mail: hiroshi@hepburn.s.chiba-u.ac.jp

Radioisotopes have been emitted around Japan due to a nuclear accident at the Fukushima daiichi nuclear power station in March 2011. A problem is the contaminated water including the atomic nucleus which relatively has a long half-life time such as $^{90}$Sr, $^{137}$Cs generated from $^{235}$U used for nuclear fuel. Internal exposures by $^{90}$Sr are more dangerous than $^{137}$Cs’s because it has a long biological half-life (49 years). Therefore, real-time $^{90}$Sr counter has been required. It is relatively easy to identify a nucleus emitting gamma ray, but it is more difficult to identify a nucleus emitting pure beta ray such as $^{90}$Sr. Typically, measurement of a radioactivity absolute value of $^{90}$Sr takes a month at least to give a result. At first, we aim to identify $^{90}$Sr / $^{137}$Cs by threshold type Cherenkov detection. It needs radiator which has less than 1.0492 of refractive index for identification of $\beta$-ray with maximum energy of 2.28 MeV from $^{90}$Sr and 1.17 MeV from $^{137}$Cs. Recently, The material satisfying this condition does not exist except the silica aerogel. We produced a prototype counter and evaluated performance. The sensitivity is observed $(5.49 \pm 0.06) \times 10^{-3}$ Hz/Bq and $(1.12 \pm 0.66) \times 10^{-5}$ Hz/Bq of $^{90}$Sr and $^{137}$Cs, respectively. And the counter achieved $(2.0 \pm 1.2) \times 10^{-3}$ of sensitivity ratio between $^{90}$Sr and $^{137}$Cs.
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1. Introduction

An accident at Fukushima daiichi Nuclear power station in March 2011 has been emitting about 900 PBq as a conversion of iodine with Internal Nuclear Event Scale (INES-estimation) of the radioisotope into the atmosphere, which is equivalent to one sixth of the nuclear accident at Chernobyl power plant. From the accident, a larger area (1800 km$^2$) inside of Fukushima Prefecture in Japan has become to dangerous area when possible of emitting is 5 mSv/year or more [1]. Main radioisotope produced by nuclear reactor is known as $^{131}$I, $^{129m}$Te, $^{238}$Pu, $^{90}$Sr, $^{134}$Cs, $^{137}$Cs, $^{133}$Xe, $^{106}$Ru and so on. Just after the accident occurred, we had been careful about internal exposure at the thyroid gland by accumulating $^{131}$I through a breath, which has a short half-life (8 days). Currently (2014), we are careful about exposure through the contamination water having $^{90}$Sr, $^{137}$Cs and so on, which have a long half-life.

$^{90}$Sr has a same chemically property with Calcium (Alkaline-earth metal), physical half-life of 29.1 years, effective half-life of 18 years, decay mode: $^{90}$Sr $\rightarrow ^{90}$Y $\rightarrow ^{90}$Zr and emitting two $\beta$-rays with maximum energy of 0.55 MeV and 2.28 MeV. On the other hand, $^{137}$Cs has a same of chemically property with potassium (Alkali material), physical half-life of 30.1 years, effective half-life of 70 days, two decay mode; emitting $\beta$-ray with maximum energy of 0.51 MeV and $\gamma$-ray with energy of 662 keV at 95% and emitting $\beta$-ray with maximum energy of 1.17 MeV at 5%. So, $^{90}$Sr is more dangerous than $^{137}$Cs in the contamination water because of the comparison with each effective half-life. A problem is difficult to identify $^{90}$Sr because it has only $\beta$ decay mode. Currently, identification of $^{90}$Sr with extraction chemically is necessary of one month because of its long half-life. We have been developing $^{90}$Sr counter applying Cherenkov light detection [2]. This counter would be need instantaneous measurement for sea food accumulated by contamination water leaked into the Pacific ocean and monitoring the drainage for cooling the reactor in Japan.

2. Real time $^{90}$Sr Counter

This counter composes of aerogel Cerenkov counter, trigger scintillation counter and veto counter. It has sensitivity to only $^{90}$Sr and a character of real time measurement because of photon counting. In this section, the devices composed in the counter and mechanism of identification for $^{90}$Sr is described.

2.1 aerogel Cherenkov counter

Threshold type Chernkov detectors make a yes/no decision based on whether the particle is above or below the Cherenkov threshold velocity $v_c = 1/n$ [3], when $n$ is refractive index. To react on the $\beta$-ray from $^{90}$Sr and not to react on the $\beta$-ray from $^{137}$Cs, a refractive index of the radiator is required between 1.017 and 1.0492. Silica aerogel [2] as a radiator has following properties; low density, high transparency and configurable index during production. The index for this counter is 1.045. photo-detection was employed light guide made from WLS (wavelength shifter) fibers and small PMTs (photomultiplier tube) for extensible effective area of this counter and reducible noise from $\gamma$-ray. WLS fiber made by Kuraray Co. Ltd. has following properties; several kinds of absorption and emission wavelength (B-3, Y-11), trapping efficiency of 5.4% by double cladding, attenuation length of about 3 m and 0.2 mm minimal diameter [5]. The PMT used in it
is R9880U-210, which has following properties; compact size(photo-cathode of 8mm diameter), metal package, fast time response, high quantum efficiency [7]. All PMT used in the counter is this series.

2.2 Trigger Scintillation counter

It is important to use thinner type trigger for the entering β-ray to aerogel with less energy loss from a sample. Trigger Scintillation counter composed of a sheet formed from scintillating fibers with size of 0.2 mm in diameter, SCSF-87M by Kuraray Co. Ltd., and two small PMTs. The sheet have dead space of 12% by the gap between the each fiber’s cladding. Trigger counter has been obtained 54.4% of detection efficiency by the test using the β-ray from $^{90}$Sr source, where the efficiency is a reaction of both PMTs connected to end of the scintillating fibers simultaneously when a trigger scintillation counter for the test at backward reacts on the β-ray.

2.3 Veto counter

A size of plastic scintillator used in this counter for veto of cosmic ray is $200 \times 400 \times 5$ mm$^3$. Scintillation light is transferred to the PMT through WLS fiber (Y-11) sheet from all side face of the scintillator. The device has been checked enough performance as the mean number of photoelectrons 7.4 p.e. by the cosmic ray test.

2.4 Mechanism of identification of $^{90}$Sr

A mechanism of the identification of $^{90}$Sr is explained in following. In case of $^{90}$Sr, when the trigger counter reacts on the β-ray, aerogel emits the Cherenkov light and WLS fiber reacts and read the Cherenkov light. On the other hand, in case of $^{137}$Cs, the trigger counter reacts on the β-ray, aerogel stops the beta-ray and Cherenkov light is not emitted. Therefore, aerogel has a function as a radiator and shielding material. the γ-ray is not reacted on the lower density detector. So, the noise is reducible. Cosmic ray is also reacted the veto counter.

Detection logic for this counter is shown in Figure 1. The electric circuit of this counter has a simple logic and lower cost, which has power supplies and discriminators for each PMT, coincidence, Fan-in/Fan-out (or coincidence level 2) for PMTs reading Cherenkov light and scaler (Figure 2).

<table>
<thead>
<tr>
<th>Table 1: component of $^{90}$Sr counter</th>
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<tbody>
<tr>
<td>the device</td>
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<tr>
<td>Cherenkov counter</td>
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<tr>
<td>Trigger counter</td>
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<tr>
<td>veto counter</td>
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3. Performance Measurement for a prototype counter

We produced a prototype counter and estimated this performance using radioactive sources. Radioactivities defined by Japan Isotope Associate: $^{90}$Sr (25kBq), $^{137}$Cs (25kBq), $^{60}$Co (100kBq) and $^{22}$Na (100kBq) were put the center of sensitive area on the counter, and a number of count per one minute was measured. A sensitivity of each radioactivity $\eta$ is defined as detection rate per unit of radioactivity,

$$\eta = \frac{1}{A} \left( \frac{N_{RA}}{t} - R_{BG} \right),$$

(3.1)

where $N_{RA}$ is a number of count in each radioactivity, $t$ is measurement time [sec], $R_{BG}$ is background rate [Hz] and $A$ is each radioactivity [Bq]. As the result, a background rate $R_{BG}$ is $(1.48 \pm
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$0.46 \times 10^{-1}$ Hz, and this counter obtained $^{90}$Sr sensitivity of $(5.5 \pm 0.06) \times 10^{-3}$ Hz/Bq and $^{137}$Cs sensitivity of $(1.12 \pm 0.66) \times 10^{-5}$ Hz/Bq in this measurement. The other sensitivity is shown in Table 2. A sensitive ratio of $^{137}$Cs/$^{90}$Sr is significant for this motivation, and the ratio is estimated as $(2.0 \pm 1.2) \times 10^{-3}$. Therefore, this prototype counter has achieved a performance of higher sensitivity of $^{90}$Sr and lower sensitivity of $^{137}$Cs. The prototype counter has effective area size of 30 cm $\times$ 10 cm and lower sensitivity on the center of area by PMTs connected both end of side. Position uniformity measurement on seven points of incident was obtained sensitive fluctuation less than 10%.

**Figure 3:** This counter obtained mean $^{90}$Sr sensitivity of $5.2 \times 10^{-3}$ Hz/Bq in the incident position measurement of seven positions (top). The position uniformity of $^{90}$Sr sensitivity was measured as the fluctuation less than 10% (bottom).

**Table 2:** performance estimation of a prototype counter

<table>
<thead>
<tr>
<th>performance</th>
<th>parameter</th>
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<tbody>
<tr>
<td>$^{90}$Sr Sensitive</td>
<td>$(5.49 \pm 0.06) \times 10^{-3}$ Hz/Bq</td>
</tr>
<tr>
<td>$^{137}$Cs Sensitive</td>
<td>$(1.12 \pm 0.66) \times 10^{-5}$ Hz/Bq</td>
</tr>
<tr>
<td>$^{60}$Co Sensitive</td>
<td>$(8.77 \pm 0.39) \times 10^{-5}$ Hz/Bq</td>
</tr>
<tr>
<td>$^{22}$Na Sensitive</td>
<td>$(5.65 \pm 0.33) \times 10^{-5}$ Hz/Bq</td>
</tr>
<tr>
<td>background rate</td>
<td>$(1.48 \pm 0.46) \times 10^{-1}$ Hz</td>
</tr>
<tr>
<td>sensitivity ratio of $^{137}$Cs/$^{90}$Sr</td>
<td>$(2.0 \pm 1.2) \times 10^{-3}$</td>
</tr>
<tr>
<td>sensitive area</td>
<td>30 cm $\times$ 10 cm</td>
</tr>
<tr>
<td>sensitive uniformity</td>
<td>$&lt; 10%$</td>
</tr>
</tbody>
</table>
4. discussion and conclusion

From the result of performance measurement for prototype counter, we have a discussion about limit of detection for $^{90}\text{Sr}$ radioactivity. The limit of detection to identify between $^{90}\text{Sr}$ and $^{137}\text{Cs}$ at relativity of 99% or more is imported from

$$N_{Sr} = aSxt + cSt$$ \hspace{1cm} (4.1)
$$N_{Cs} = bkSxt + cSt$$ \hspace{1cm} (4.2)
$$N_{Sr} > N_{Cs} + 2.58\sqrt{N_{Cs}}.$$ \hspace{1cm} (4.3)

where $N_{Sr}$ is Number of count for $^{90}\text{Sr}$, $N_{Cs}$ is Number of count for $^{137}\text{Cs}$, $a$ is sensitivity of $^{90}\text{Sr}$ [Hz/Bq], $b$ is sensitivity of $^{137}\text{Cs}$ [Hz/Bq], $c$ is background rate per unit area [Hz/cm$^2$], $S$ is effective area [cm$^2$], $k$ is amounts ratio of $^{137}\text{Cs}$ / $^{90}\text{Sr}$ at measurement environment, $t$ is measurement time [sec], $x$ is radioactivity of $^{90}\text{Sr}$ [Bq]. The value $x$ satisfied Eq. (4.1), (4.2) and (4.3) means a limit of detection of $^{90}\text{Sr}$ and depends on effective area $S$, measurement time $t$ and environment of the ratio of $^{137}\text{Cs}/^{90}\text{Sr}$ existence $k$. Figure 4 shows the limit of detection at $k$ of the ratio and $t$ of measurement time. These plots present that the limit of detection is improved depending on the measurement time and not deteriorated well depending on the environment of the ratio of $^{137}\text{Cs}/^{90}\text{Sr}$ existence. Therefore, this prototype counter has a enough performance of identifiable $^{90}\text{Sr}$ with 99% or more in the environment where $^{137}\text{Cs}$ exists much.

![Figure 4: The limit of detection for $^{90}\text{Sr}$ at measurement of 1 sec (blue), 60 sec (Black), 600 sec (red), 3600 sec (green) in the environment of the ratio of $^{137}\text{Cs}/^{90}\text{Sr}$. These plots present that the limit of detection is improved depending on the measurement time and not deteriorated well depending on the environment of the ratio of $^{137}\text{Cs}/^{90}\text{Sr}$ existence.](image)

In conclusion, we have developed real time $^{90}\text{Sr}$ counter applying Cherenkov light detection, produced a prototype counter and measured the performance estimation. From the measurement using radioactivity, a prototype counter has the enough performance of higher sensitivity of $^{90}\text{Sr}$, lower sensitivity of $^{137}\text{Cs}$ and identifiable $^{90}\text{Sr}$ with 99% or more in the environment where $^{137}\text{Cs}$ exists much.
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References


