Radiation measurements with heat-proof polyethylene terephthalate bottles

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This study demonstrates that the energy resolution of a newly developed 100 per cent pure polyvinyltoluene (PVT) plate allows its use as a base material for a plastic scintillator. The energy resolution, which is a key element for high-performance radiation detectors, was \( \Delta E/E = 8.41 \pm 0.07\% \) (full width at half maximum (FWHM)) for 976 keV K-line conversion electrons from a \(^{207}\)Bi source. On the basis of results from \(^{207}\)Bi and \(^{137}\)Cs sources, the observed energy resolution of the PVT plate, \( \Delta E/E = 8.7/E^{1/2}\% \) (FWHM), was slightly better than that of a typical plastic scintillator (BC-408), \( \Delta E/E = 8.7/E^{1/2}\% \) (FWHM), with \( E \) in units of MeV. These results prompted us to search for other new base materials for plastic scintillators. In this study, we examined polyethylene terephthalate (PET) bottles, a common source of domestic plastic waste. We demonstrated that a lump of heat-proof PET bottles is fluorescent; moreover, there is excellent compatibility of the fluorescence with the quantum efficiency of typical photomultiplier tubes. This inexpensive source of plastic appears suitable for radiation measurements and as a base material for plastic scintillators. Future studies on the radiation response of plastics should lead to the development of higher performance and more eco-friendly radiation detectors.

Keywords: polyethylene terephthalate bottle; fluorescence; radiation measurement; scintillator; photosensor

1. Introduction

Organic scintillators have been used as radioactivity monitors in a wide range of scientific fields, including medical science, physical science and engineering. Plastic scintillators, first developed about 60 years ago (Schorr & Torney 1950), are a form of organic scintillator and are commonly used in radiation measurement and radiation protection. Because these scintillators are easy to manufacture and handle, they have become very popular in research.

The basic material of a plastic scintillator is a plastic that contains some form of aromatic hydrocarbon such as polyvinyltoluene (PVT) or polystyrene (Bondansky & Eccles 1957). Generally, a plastic scintillator is produced by doping

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the plastic with a few wavelength shifters. This allows absorbed ultraviolet light to be emitted as more easily measurable visible light (Curran 1953; Swank & Buck 1953; Thornton 1954).

There are at least three reasons to dope plastics with wavelength shifters. (i) Wavelength shifters increase the fluorescent yield (Birks 1964), which influences the performance of the base material (i.e. the photon collection and energy resolution—key elements for high-performance radiation detectors). (ii) The transmitting efficiency of ultraviolet light is low because of self-absorption in the base material, which influences the manufacture of large pieces of the base material. (iii) When photosensors were developed about 60 years ago, they had low sensitivity for ultraviolet light, making it difficult to measure ultraviolet light directly. Presently, state-of-the-art photosensors with high ultraviolet sensitivity are being developed to resolve this difficulty.

Research on base materials and wavelength shifters has been ongoing for at least half a century (Schram & Lombaert 1963). Thus, there is much quantitative data in terms of the fluorescent yield and the transmission efficiency of small pieces (a few cubic centimetres) of base materials in the fields of physics and chemistry (Moser et al. 1993). However, in terms of energy resolution, there is little quantitative data available for any base material. It is important to be able to estimate the energy resolution of a base material. Because of remarkable advances in technology in recent years, especially in the improvement of photon collection, it is now possible to evaluate the energy resolution and also use a medium-sized (approx. 10 cm$^3$) piece of material. Therefore, in the present paper, we describe the energy resolution of a newly developed 100 per cent pure PVT plate of medium size. The PVT plate was found to have a slightly better energy resolution than a typical plastic scintillator in common use (BC-408; manufactured by Saint-Gobain Ceramics & Plastics Inc., France).

In addition, we also investigated a new base material for plastic scintillators. We focused on a source of waste consumer plastic, namely polyethylene terephthalate (PET) bottles. Unlike specialized scintillator plastics, these are readily available worldwide. In this paper, we demonstrate that a lump of heat-proof PET bottles is fluorescent and can be used for radiation measurements. Moreover, we show that the fluorescence from the PET lump has an excellent compatibility with the quantum efficiency of a typical photomultiplier tube in common use.

2. Material and methods

To evaluate the energy resolution of the base material of a plastic scintillator, a 100 per cent PVT plate of medium size (62 × 62 × 10 mm$^3$) was manufactured in cooperation with Saint-Gobain Ceramics & Plastics Inc. Ionization in the PVT plate produces ultraviolet light with a maximum emission wavelength of approximately 330 nm. Energy resolution is known to depend on light collection (Nakamura et al. 2010), which is thus a key element for achieving adequate energy resolution.

To efficiently detect ultraviolet light generated in the PVT plate, the four smaller (i.e. 62 × 10 mm$^2$) sides of the PVT plate were viewed with the commonly used photomultiplier tubes (H7195, Hamamatsu Photonics K.K.,

Japan) connected with optical grease (BC-630; Saint-Gobain Ceramics & Plastics Inc., France). This system was placed in a black box to shield it from light.

Conversion electron sources are very useful for calibration and evaluation of a radiation detector because conversion electrons are monoenergetic (Leo 1992). However, conversion electron sources emit not only conversion electrons but also gamma rays with approximately the same energy. There is a possibility that the gamma rays compete in the energy window selected for the conversion electrons. In the present study, the emitted conversion electrons from the conversion electron sources were measured through a lead collimator used not only to collimate the radiation but also to reject accidental coincidence events between the conversion electrons and gamma rays with approximately the same energy.

The lead collimator consisted of two lead blocks, each with a surface area of $30 \times 30 \text{ mm}^2$ and a 10 mm diameter hole in the centre. The heights of the lead blocks were 8 and 2 mm, respectively. The lead collimator was located at the centre of the larger side of the PVT plate. The experimental set-up is shown in figure 1. Here, a $^{207}\text{Bi}$ source (BIRB4391, High Technology Source Ltd) and a $^{137}\text{Cs}$ source (CS21, Japan Radioisotope Association, Japan) were used as the conversion electron sources (table 1).

<table>
<thead>
<tr>
<th>isotope</th>
<th>half-life (years)</th>
<th>type</th>
<th>energy (keV)</th>
</tr>
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<tr>
<td>$^{207}\text{Bi}$</td>
<td>32</td>
<td>gamma ray</td>
<td>569</td>
</tr>
<tr>
<td></td>
<td></td>
<td>K-line conversion electron</td>
<td>482</td>
</tr>
<tr>
<td></td>
<td></td>
<td>gamma ray</td>
<td>1064</td>
</tr>
<tr>
<td></td>
<td></td>
<td>K-line conversion electron</td>
<td>976</td>
</tr>
<tr>
<td></td>
<td></td>
<td>gamma ray</td>
<td>1770</td>
</tr>
<tr>
<td></td>
<td></td>
<td>K-line conversion electron</td>
<td>1682</td>
</tr>
<tr>
<td>$^{137}\text{Cs}$</td>
<td>30</td>
<td>gamma ray</td>
<td>662</td>
</tr>
<tr>
<td></td>
<td></td>
<td>K-line conversion electron</td>
<td>626</td>
</tr>
<tr>
<td></td>
<td></td>
<td>beta particle</td>
<td>511 ($E_{\text{max}}$)</td>
</tr>
</tbody>
</table>
3. Results and discussion

(a) A 100 per cent pure polyvinyltoluene plate

To calibrate the energy scale of the PVT plate, 976 keV K-line conversion electrons from the $^{207}$Bi source that was placed on the top of the lead collimator were measured. The energy spectra obtained by summing the signals from the four photomultiplier tubes are shown in figure 2.

The shape of the energy peak of the 976 keV K-line conversion electron line is asymmetrical because some energy of the conversion electrons was deposited in the radioisotope source. Thus, the energy of the conversion electrons observed by the radiation detector was less than the primary energy, which is equal to the difference between the excitation energy and the atomic binding energy (Nakamura et al. 2008). For the $^{207}$Bi source, the energy peak of the 976 keV K-line conversion electron line was shifted lower by approximately 10 keV.

The energy resolution of the PVT plate was obtained as $\Delta E/E = 8.41 \pm 0.07\%$ (full width at half maximum (FWHM)) for the 976 keV K-line conversion electrons. The energy resolution for the 482 keV K-line conversion electrons from the $^{207}$Bi source was $\Delta E/E = 12.22 \pm 1.88\%$ (FWHM). The energy spectra obtained from the $^{137}$Cs source are shown in figure 3. For the $^{137}$Cs source, not only conversion electrons and gamma rays but also beta particles were measured. The energy resolution obtained was $\Delta E/E = 10.73 \pm 0.21\%$ (FWHM) for the 625 keV K-line conversion electrons from the $^{137}$Cs source.

Figure 4 shows the observed energy resolutions of the PVT plate and a typical plastic scintillator (BC-408, with a size similar to the PVT plate), which were obtained by the same procedure. The observed energy resolution of the PVT plate is well reproduced by $\Delta E/E = 8.2/E^{1/2}\%$ (FWHM) with $E$ in units of MeV (Knoll 2000). The observed energy resolution of the PVT plate, $\Delta E/E = 8.2/E^{1/2}\%$ (FWHM), was slightly better than that of a typical plastic scintillator, $\Delta E/E = 8.7/E^{1/2}\%$ (FWHM).

This study demonstrates that a PVT plate of medium size is capable of obtaining good energy resolution and that it can detect various forms of radiation. In addition, even a photomultiplier tube with normal specifications is sufficient to detect ultraviolet light. Use of a photomultiplier tube that has higher sensitivity to ultraviolet light would result in better energy resolution. On the basis of these results, high-performance radiation detectors can be developed in the future.

(b) Lump of polyethylene terephthalate bottles

We examined whether waste PET bottles might be used to detect radiation in place of the typical base materials of plastic scintillators. We investigated a lump of heat-proof PET bottles (provided by Mitsui Chemicals Inc., Japan). Since the PET lump was a sample, it had a complicated shape (figure 5), with outer dimensions $110 \times 50 \times 5 \text{mm}^3$. Its properties are listed in table 2.

The principal ingredient of the PET lump was polyethylene terephthalate, which fluoresces. Therefore, we measured the emission distribution of the PET lump using a spectrometer (F-2700; Hitachi High-Technologies Corp., Japan). Figure 6 shows the emission spectrum obtained. The emission peak of the PET
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Figure 2. Energy spectrum of conversion electrons and gamma rays from the $^{207}$Bi source measured by summing the energy signals from the four photomultiplier tubes. The peaks shown by the dashed lines are the K, L1, L2 and L3 conversion electron lines. These show that the peak is asymmetrical owing to energy deposition in the $^{207}$Bi source. The taller dashed line is the 976 keV K-line conversion electron line. The dotted lines show the energy distributions by Compton scattering. Solid red line, fitting line; dotted lines, gamma ray.

lump around 380 nm was in good agreement with the quantum efficiency peak of the same photomultiplier tube that had been used to evaluate the PVT plate. Since this indicated a strong possibility of using the PET lump as a scintillator, even without wavelength shifters, we investigated its ability to measure radiation from a radioisotope source.

In the experiment, one of the same photomultiplier tubes that had been used to evaluate the PVT plate was attached to the bottom surface of the PET lump and connected with optical grease. The lead collimator was also located on the top surface of the PET lump.

The conversion electrons from a $^{207}$Bi source were measured through the lead collimator that was placed on the top surface of the PET lump. The obtained energy spectrum of the PET lump is shown in figure 7. A sufficiently large peak representing the 976 keV K-line conversion electrons was observed, similar to the results seen for the PVT plate.

The obtained energy resolution of the PET lump was $\Delta E/E = 24\%$ (FHWM) for the 976 keV K-line conversion electrons. The energy resolution was worse than that of the PVT plate and the plastic scintillator. However, there is the potential to improve the energy resolution of the PET lump. The PET lump was whitish in appearance because of the cooling process during recycling of the PET

Figure 3. Energy spectrum of conversion electron lines, gamma rays and beta particles from a $^{137}$Cs source obtained by summing the signals from the four photomultiplier tubes. The dashed lines show the K, L1, L2 and L3 conversion electron lines. The energy distributions from Compton scattering are shown as the dotted line. The dot-dashed line shows the beta particles. Solid red line, fitting line; dotted line, gamma ray.

bottles and doping of the materials for heat-proofing. Therefore, the ultraviolet light generated in the PET lump is absorbed and scattered within the material. The energy resolution of the PET lump might be improved by using a better cooling process during recycling and by investigating the use of other types of PET bottles.

4. Conclusions

We were able to successfully detect radiation using the PET lump, and we showed that the PET lump has the potential to detect different types of radiation. Because of its complicated shape and without adopting surface finishing (which causes poor photon propagation efficiency), a quantitative performance evaluation of the PET lump itself could not be performed; however, the performance of the PET lump was experimentally ascertained. Studies to provide quantitative data for the PET bottle are in progress.

The CROSS (Correlation Response Observatory for Scintillation Signals) project was initiated in October 2008. This project aims to develop medical diagnostic equipment with performance that is equivalent or slightly superior to that of the existing positron emission tomography equipment, but with

Figure 4. Energy resolutions of the PVT plate obtained from monoenergetic 976 keV and 482 keV K-line conversion electrons from a $^{207}$Bi source and monoenergetic 626 keV K-line conversion electrons from a $^{137}$Cs source. Here, the energy resolution of the PVT plate is $\Delta E/E = 8.2/E^{1/2}\%$ (FWHM) and that of the plastic scintillator (BC-408) is $\Delta E/E = 8.7/E^{1/2}\%$, with $E$ in units of MeV (FWHM). Dashed line, plastic scintillator; solid line, polyvinyltoluene.

Figure 5. Photograph of the lump of heat-proof PET bottles. As is shown, this sample is whitish in appearance, a complicated shape and no surface finishing.
Figure 6. Correlation between the emission distributions of the scintillators and the quantum efficiency of the photomultiplier tube. The dashed line is the emission spectrum of the PET lump. The dotted line is the emission spectrum of the PVT plate. The dot-dashed line is the emission spectrum of the plastic scintillator (BC-408). The solid line shows the quantum efficiency of the photomultiplier tube (H7195).

Table 2. Properties of the scintillators.

<table>
<thead>
<tr>
<th>material</th>
<th>100% pure plastic scintillator lump of heat-proof PET bottles</th>
</tr>
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<tbody>
<tr>
<td>supplier</td>
<td>Saint-Gobain</td>
</tr>
<tr>
<td>base</td>
<td>(C9H10)n</td>
</tr>
<tr>
<td>density (g cm(^{-3}))</td>
<td>1.03</td>
</tr>
<tr>
<td>index</td>
<td>1.58</td>
</tr>
<tr>
<td>wavelength max. emission (nm)</td>
<td>330</td>
</tr>
</tbody>
</table>

production costs that are lower than that of the existing medical diagnostic equipment. In the project, plastic materials are used as gamma ray scattering detectors. We know that the interaction rate of gamma rays in a material depends on its density. Thus, when medical diagnosis equipment is developed, it is necessary to select the plastic materials by considering not only the energy resolution but also the properties of the materials. Discussion of this will be reported in a separate publication.
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Figure 7. Energy spectrum of the radiation emitted from the $^{207}$Bi source obtained by using the lump of heat-proof PET bottles. By comparing this figure with figure 2, the peak is shown to represent the 976 keV K-line conversion electrons.

One feature of the PET bottle material is that oxygen is present as a principal ingredient (table 2). As a result, the density of the PET lump is higher than that of the plastic scintillator and PVT. Therefore, by using PET bottles as the plastic material, we may be able to produce cost-effective positron emission tomography detectors.

The lives of many people could potentially be saved with an ecologically friendly instrumentation solution.

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References
