Development of the Dual Scintillator Sheet and Phoswich Detector for Simultaneous Alpha- and Beta-rays Measurement

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ABSTRACT

Thin sheet type of ZnS(Ag)/plastic dual scintillator for simultaneous counting of alpha- and beta-particles using a organic and inorganic scintillator widely used in the radiation measurement was manufactured, which could be applicable in the contamination monitoring systems. Counting materials were manufactured by solidification of the scintillator solution which mixed scintillator, solvent, and polymer. Prepared dual scintillator is a counting material which can simultaneously measure the alpha- and beta-particles. It was divided into two parts : an inorganic scintillator layer for alpha-particle detection and an organic one for beta-particle detection. The organic layer was composed of 2,5-diphenyloxazole [PPO] and 1,4,-bis[5-phenyl(oxazolyl)benzene] [POPOP] acting as the scintillator and polysulfone acting as the polymer. The inorganic layer was composed of ZnS(Ag) as scintillator and polysulfone as paste. The ZnS(Ag) scintillator layer was printed onto the organic layer using screen printing method. To estimate the detection ability of the prepared counting materials, alpha-particle emitting nuclide, Am-241, and beta emitting nuclide, Sr/Y-90, were used. The scintillations produced by interaction between radiation and scintillator were measured by photomultiplier tube. The overall counting results reveal that the developed detector is efficient for simultaneous counting of alpha- and beta-particles. For application test, the dual scintillator was fabricated with a phoswich detector for monitoring the in-pipe alpha and beta contamination. To deploy inside a pipe, two types of phoswich detectors, sheets and cylinders, were prepared. For in-pipe monitoring, it was found that the cylindrical type was excellent. In the study, polymer composite counting material and phoswich detectors were prepared using organic and inorganic scintillator for detecting different radiations. In the future, it will be applied to the contamination monitoring system for nuclear decommissioning sites, waste treatment sites, and similar areas.

INTRODUCTION

Generally a great quantity of waste was generated during the decommissioning of nuclear facilities. These wastes are contaminated with various types of alpha, beta, and gamma emitting nuclides. The contamination level of the decommissioning wastes must be surveyed for free release or reuse.

Surface contamination is divided into removable and fixed contamination. Fixed contamination is measured by a direct method with a portable contamination monitor. Rremovable contamination is measured by an indirect method using smear paper and a low background proportional counter. A direct method can't distinguish between the removable and fixed contamination. Although the high background disturbance would degrade the detection accuracy of a direct measurement device, the device is commonly applied for on-site measurement at the contamination sites. This kind of direct surface contamination measurement is carried out with a gas or scintillation counter. Large area scintillation detectors can be used for simultaneous detection of alpha- and beta- particles.

However, it is very difficult to count the radioactive contamination level inside a pipe using conventional counting methods because of the small diameter of the piping. Also, it would be useful if the surface contamination of the alpha and beta-ray could be simultaneously measured in the nuclear facilities. Such a contamination measurement could be conducted by proportional counter or phoswich detectors. But the proportional counter is very difficult to make in a small enough size to allow inserting into a pipe. It is possible to manufacture the phoswich detector for small size and simultaneous counting of the alpha- and beta-ray.

The phoswich detector is convenient for monitoring alpha and beta contamination using only a single detector, composed of a ZnS(Ag) scintillator for counting the alpha particles and a plastic detector for beta particles [1-5]. Silver activated zinc sulfide is one of the older inorganic scintillators. It has a very high scintillation efficiency, comparable to that of NaI(Tl), but is only available as a polycrystalline powder. As a result, its use is limited to thin screens used primarily for alpha particles or other heavy ion detection. The scintillator for counting beta particles is typically made from plastic.

To measure the radioactive contamination in piping, the prepared composite is formulated as a phoswich detector type. The phoswich detector unit has a combined structure of a photomultiplier tube [PMT] and the prepared composite, rolled up like a cylindrical tube to be inserted into the piping. Using the prepared phoswich detector, the detection of radioactive contamination in piping is proceeded by distinguishing the signals obtained from the detector through the pulse shape discrimination technique, to evaluate the amounts of alpha and beta particles separately. The overall counting results reveal that the developed phoswich detector is efficient for simultaneous counting of alpha- and beta-ray in a pipe.

EXPERIMENTAL DESIGN AND METHODS

Preparation of the dual scintillator sheet

The preparation procedure of the dual phosphor is shown in Fig. 1. Scintillation detectors for the simultaneous detection of alpha and beta particles can be produced by coating of ZnS(Ag) on the top of a thin (about 0.25 mm) plastic scintillator for beta particle detection. In this research, scintillation composites are prepared as a dual type structure capable of detecting alpha and beta particles simultaneously. The composite is formulated by coating ZnS(Ag) particles for alpha-ray detection over the plastic scintillator for beta-ray detection as well as mechanical support of the composite itself. The bottom plastic layer is obtained by the solution casting and subsequent evaporation technique, using various types of polymers. The ZnS(Ag) top layer for alpha-ray detection is coated over the base layer through the screen-printing technique.

When the radiation energy from radioactive contamination sites works on the plastic scintillator and is transported into the base polymer, the energy transferred in polymer is released by the polymer relaxation. The released energy is picked up by the first dissolved solute, inducing the emitting of scintillation photons near the wavelength of ultraviolet [UV]. To optimize the PMT's detection efficiency, the wavelength of the scintillation photons can be shifted by dissolving the second luminescent organic solute in a plastic base layer. In this research, polysulfone [PSf] is used as the base polymer material, PPO as the first organic solute or the main luminescent additive, and POPOP as the wave-shifter or the additional luminescent additive. For the preparation of the base plastic layer of a scintillator composite, both PPO and POPOP are first dissolved in methylene chloride [MC]. PSf is added in the solution and mixed by using a mechanical stirrer at 150 rpm. The prepared solution is carefully cast on glass plates at the specific thickness using the applicator or the casting knife. Before being cast, the solution is checked to ensure there are no bubbles, which can result in quenching of the scintillation photons. The cast film is allowed to set for 24 hours in the atmosphere for the formation of solid base plastic scintillator.

The top layer of the composite is attached into the base layer using the screen printing technique in which the paste consisting of a ternary solution of inorganic scintillator ZnS(Ag), binder and solvent is squeezed over the screen mesh, and the solution passing through the screen is coated on the sub-layer. For the paste, PSf is dissolved in dimethylformamide [DMF] and ZnS(Ag) is added into this solution and mixed to be formed a homogeneous paste. The paste is screen printed on the prepared plastic scintillator, using the urethane squeezer. To avoid a humidity impact during the adhesion of two layers, the composite containing the coated ZnS(Ag) layer is placed on the oven of 80^0C for 24 hours. The prepared composite scintillator has a tightly combined double layer structure.

Dual scintillator phoswich detector

The combination of two dissimilar scintillators optically coupled to a single PMT is often called a phoswich (or phosphor sandwich) detector. The scintillators are chosen to have different decay times so that the shape of the output pulse from the PMT is dependent on the relative contribution of scintillation light from the two scintillators (Table I).

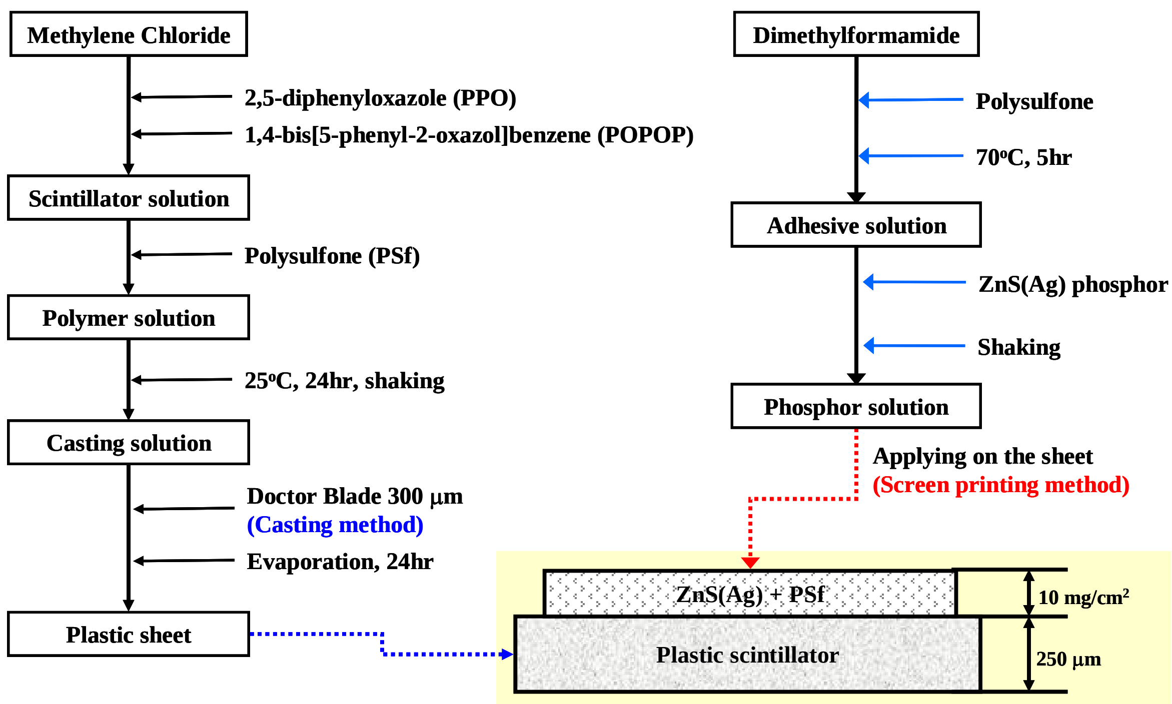

Fig. 1. Preparation procedure of the dual scintillator sheet for simultaneous counting of alpha- and beta-ray.

Table I. Characteristics of the phoswich detector used in this study

	Plastic	ZnS(Ag)
Scintillator	PPO & POPOP	ZnS(Ag)
Density	1.32 g/cc	10 mg/cm^2
Light Output (% Anthracene)	64	300
Refractive Index	1.58	-
Decay Time (ns)	2.1	200
Wavelength of Max. Emission (nm)	425	450

The zero crossing method of pulse shape discrimination (PSD) has widely used in the phoswich detectors. Measurement of the zero crossing time of the bipolar pulse determines the particle type. The distribution of zero crossing times can be used to determine a PSD figure of merit M.

$$M = T / (t_a + t_b)$$

T is the separation between the time peaks and t_a and t_b are the respective full width at half maximum [FWHM] of the zero crossing time distributions for particle a and b, respectively.

An Am-241 source for alpha particle counting and Sr/Y-90 for beta particles counting were used. Most of pulse-height and pulse-shape distributions were obtained by measuring the respective sources for 1,000 s.

RESULTS AND DISCUSSION

The dual scintillator sheet

To be a good plastic scintillator, a base polymer has to keep a transparent feature as well as the ability of good beta particle detection. Various types of polymers possess both the good radioactive energy transfer and transparency. Among these polymers, PSf, polystyrene [PS], Estyrene, and Poly(bisphenol A Carbonate) [PBAC]can be good candidates for this specific scintillator, requiring the good mechanical strength and the beta particle detection ability. Using these polymers, the plastic scintillators were prepared through the same preparation protocol, and their beta particle detection capacity was measured. As shown in Fig. 2, PS has the best detection capacity among the prepared scintillators, but the prepared composite sheet was too brittle to be formed into a cylindrical shape. PSf, Estyrene, and PBAC show the similar detection efficiency. In the case of PBAC, the raw chips are not dissolved easily in the solvent. Therefore, PSf and Estyrene are best fit into the required properties and selected as base polymer materials for the solvent preparation technique.

To evaluate the relationship between the detection capacity and the composite thickness of the prepared scintillators, several composites from 0.14 to 0.45 mm in thickness were formulated and compared with each other. In the formulation for lessening the gamma-ray impact, the thickness of the prepared scintillators was kept at 0.25 mm.

The transmission rates of the prepared base plastic scintillators are more than 85% near 425 nm, the wavelength of the emitted scintillation photons.

By using PSf as the binder for adhering the ZnS(Ag) layer into the base PSf layer, the composite can hold a monolithic structure without any extra adhesive.

In the case of ZnS(Ag) scintillator, the optimal thickness for alpha-ray detection was 10 mg/cm^2. If the thickness of ZnS(Ag) layer is greater than 25 mg/cm^2, it is useless as a scintillation detector because the generated scintillation will not reach the PMT. Also, in the case of low thicknesses, the ZnS(Ag) layer can't absorb the whole energy of the alpha-ray, so it brings about a small detection efficiency. Therefore, the thickness of the ZnS(Ag) layer plays a key roll in the detection ability, so in this study it was kept at 10 mg/cm^2.

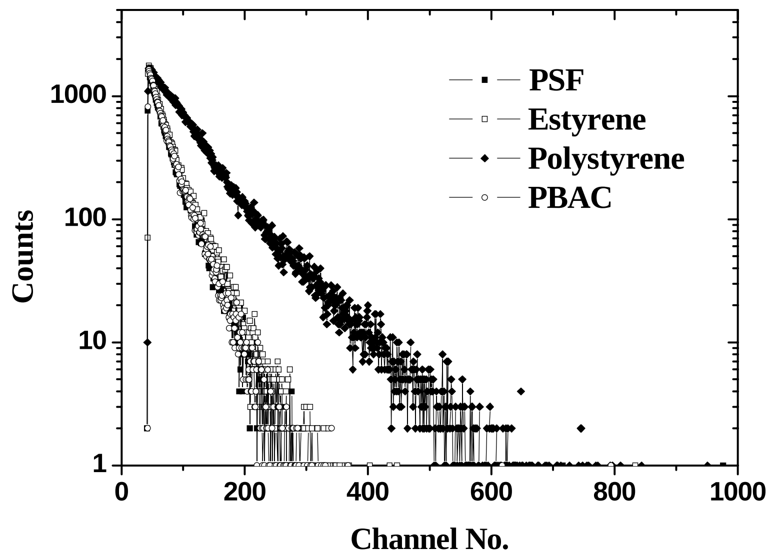

Fig. 2. Beta-ray spectrum of the plastic sheet for beta-ray counting according to polymer materials.

Phoswich detector for alpha- and beta-ray detection

A conceptual diagram of the phoswich detector for simultaneous counting of alpha- and beta-rays in a pipe is shown in Fig. 3. The prepared dual scintillator was formed into a cylinder.

Each radiation must be discriminated in a phoswich detector to allow simultaneous measurement of different type radiations using single-detector system. The pulse-shaped discrimination and pulse-height discrimination were generally used to discriminate each radiation. Pulse heights generated by alpha- and beta-rays were discriminated using the energy level discriminator for the pulse-height discrimination method. But, the accurate measurements can't be performed because of overlap of alpha and beta events for low level activity. Therefore, the pulse-shape discrimination method is generally used to discriminate each other. The pulse-shape discrimination is the method discriminating rise-time of scintillation formed in each scintillator. The pulse-shape distribution spectra for alpha- and beta-ray measured with the phoswich detector are shown in Fig. 4.

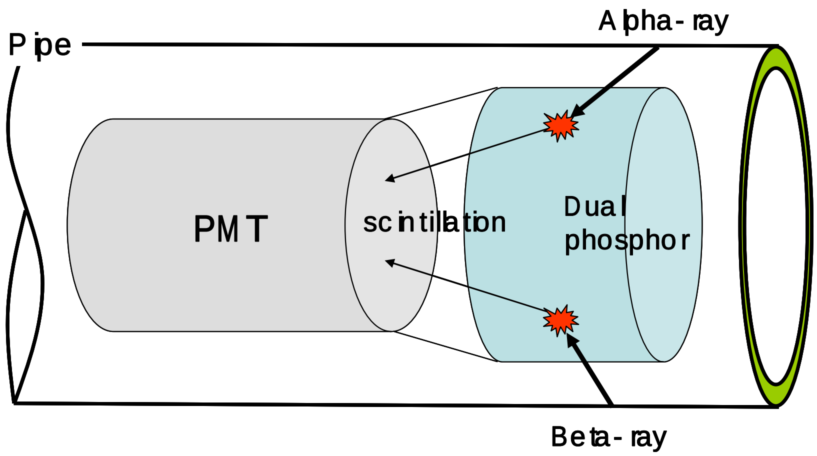

Fig. 3. Conceptual diagram of the phoswich detector for simultaneous alpha- and beta-particles counting in a pipe.

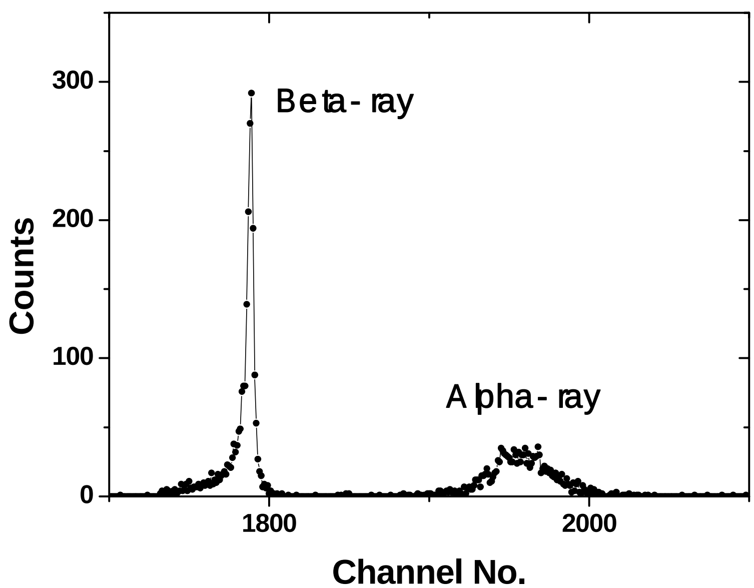

Fig. 4. Pulse-shape distribution of alpha- and beta-rays measured with dual scintillator phoswich detector in a pipe.

CONCLUSION

Polymer composites of a ZnS(Ag)/plastic dual scintillator sheet were prepared. The composite was composed of both the base plastic scintillator, formulated through the solvent casting method, and the top ZnS(Ag) layer, coated through the screen printing on the base layer. It had good detection capability of the simultaneous detection of alpha and beta particles, and was relatively easy to make, compared with making the composite by the conventional heat-melting method. The thickness of a base layer is 0.25 mm, to lessen the gamma ray hindrance, and the density of the ZnS(Ag) layer is 10 mg/cm^2.

The dual scintillator sheet was fabricated with phoswich detector for monitoring the in-pipe alpha- and beta-rays contamination. In order to apply pipe inside, a cylindrical dual scintillator sheet was fabricated. In the near future the phoswich detector will be tested in the decommissioning site of the Korea Research Reactor 1&2 and the uranium conversion facility.

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