

PREPARATION AND PROPERTY OF THE POLYMER SCINTILLATOR FOR THE ALPHA CONTAMINATION MONITORING

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ABSTRACT

In the operation of nuclear research facilities, such as the Korean Research Reactor 1&2 and the Uranium Conversion Plant, a significant amount of nuclear wastes are produced, which work as α -particle emitting sources. The wastes contaminated with alpha include pipes, reaction vessels, and storage vessels, and their contamination status must be surveyed for a disposal and reuse in the future. For the quantification of the surface contamination of nuclear wastes, a polymer-wiping medium applied without a scintillation cocktail was formulated. The medium, scintillator-embedded polymer membrane for the alpha-ray detection, was prepared by impregnating organic scintillators in a membrane structure. The medium or plastic scintillator consists of polysulfone as a matrix with PPO as an organic scintillator and POPOP as a wave-shifting agent dissolved in the matrix. The emitting wavelength and intensity of the scintillator were measured to evaluate the efficiency of the scintillator. Also, in order to determine the optimum thickness of the membrane, the variation of the detection ability with the membrane thickness was assessed.

INTRODUCTION

A pilot Uranium Conversion Plant (UCP) was constructed in 1982 for the purpose of the technology development and the localization of nuclear fuels for a HWR. Its operation was stopped in 1993. Considering the needs to block the leakage of radioactive contaminants, reduce the extra costs of maintenance, and build up the decommissioning and dismantling technology of nuclear fuel cycle facilities, the dismantling of the conversion facility was decided upon, and the Korea Atomic Energy Research Institute (KAERI) started a decommissioning program for the plant in 2004. In the process of dismantling, a large volume of nuclear wastes would be generated. Typical wastes consist of contaminated ducts, storage tanks, a reactor batch, etc. These wastes have to be carefully managed, by being kept safe and secure from an environmental context. Therefore, the precise estimation of the radioactive contamination of the wastes is a prerequisite to determining the disposal or decommissioning of those wastes.

Surface contamination can be classified into either that fixed for a deposit or that able to be removed by wiping or cleaning the surface. The fixed contamination can be quantified by applying a survey meter directly to the contaminated site. To obtain reliable data, the possible

impedance from outside radiation sources needs to be controlled. On the contrary, radioactive contamination due to a removable one can be estimated by the wipe test, which requires a wiping and a subsequent counting of the contaminants attached to the wiping medium. The wipe test or indirect survey has to be carefully operated during a wiping, so as not to disturb the wastes on the wiping area and be absorbed by the handler. The wipe test involves a smearing process with either a plain, a dry adsorbent such as a filter paper or a specific membrane holding scintillation agents inside the membrane matrix. This kind of specific membrane can be applied without the aid of a scintillation cocktail, which is required for the plain medium.

A specific scintillation membrane, which detects a radioactive contamination, has to be differentiated in the matrix structure as well as in the scintillating agents embedded in the structure, depending on the radiation source. And research has been progressed to formulate specific scintillator-impregnated membranes [1-5]. In this report, a new plastic scintillator was prepared and characterized, which is applicable to the wiping and a subsequent detection without an extra scintillation cocktail.

EXPERIMENTALS AND METHODS

Preparation of the Plastic Scintillator

Plastic scintillation membranes consisting of PSF (polysulfone) as polymer matrix, PPO (2,5-diphenyloxazole) as a primary scintillator, and POPOP (1,4-bis-2-(5-phenyl-oxazolyl)) as a wave-shifting agent were prepared through the solidification of polymeric solutions. To formulate a casting solution, PPO and POPOP were dissolved in MC (methylene chloride) and then PSF was added and homogenized using a mechanical stirrer. The prepared solution was cast on a glass plate using the automatic casting machine, shown in Figure 1 and was coagulated via a solvent evaporation at the atmosphere, resulting in a solid film.



Fig. 1. Automatic casting machine for the polymer membrane preparation

Estimation of the Radiation Detection Ability of the Plastic Scintillator

The amount of scintillation light formed by the reaction between the scintillators and the α -particles was counted using a PMT (photomultiplier tube) and LB (low background α/β counter, Canberra, S5XLB). For the radioactive detection test of the prepared membranes, a radioactive solution of a common radionuclide, Am-241, was spotted on a membrane, and the emitted light was quantified by a PMT and LB.

RESULTS AND DISCUSSTION

In a previous paper [6], we reported that the membrane containing CAYS (Cerium Activated Yttrium Silicate) in the matrix can be employed as a “wiping scintillator”, which can be used as a wiping medium applicable for a direct detection without a scintillation cocktail. The CAYS-impregnated membrane can provide a reliable detection capacity in a scintillation reaction of a low-energy beta-ray emitter.

For a comparison with new plastic scintillation membranes, the CAYS-impregnated membrane was applied for the detection of the α -particle, Am-241. The membrane, holding CAYS in the matrix, did not show any energy spectrum related to the α -particle, as shown in Figure 2, indicating that the detectable spectrum is not formed due to the long decay time of the scintillation light. On the contrary, the PSF scintillation membrane, containing PPO and POPOP as scintillating agents, responds to the same α -particle, providing a detectable energy spectrum from the reaction with the radionuclide (Figure 3).

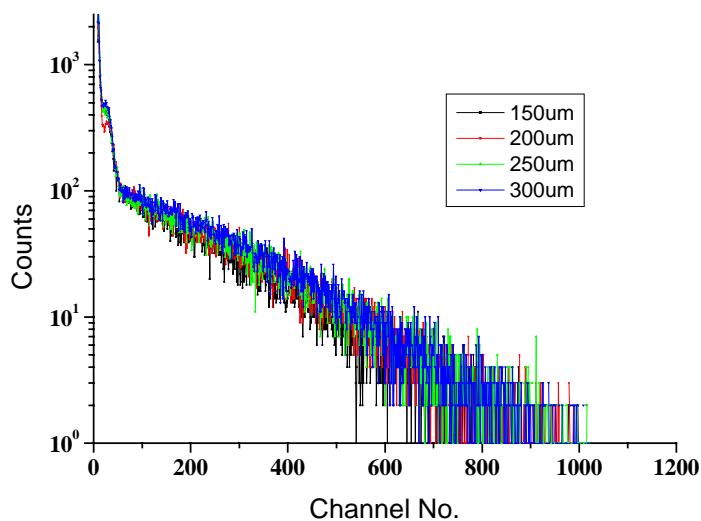


Fig. 2. Spectra for the alpha response of the CAYS-impregnated membrane

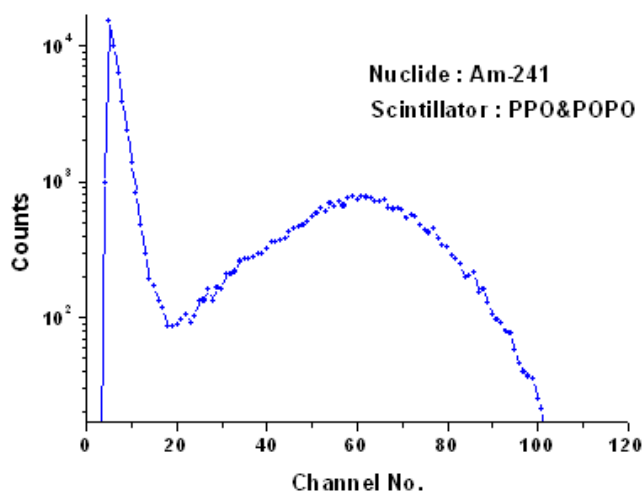


Fig. 3. Spectra for the alpha response of the polymer membrane containing PPO and POPOP

The PMT, for detecting the scintillated light, reacts effectively near a specific wavelength of 425 nm. Therefore, the scintillation reaction needs to be controlled to emit that particular spectrum. To evaluate the scintillation characteristics of PPO and POPOP, the wavelengths emitted from those agents were measured. The peak of PPO was detected near 410 nm and that of POPOP near 425 nm, revealing that they can be highly efficient for a PMT measurement (Figure 4).

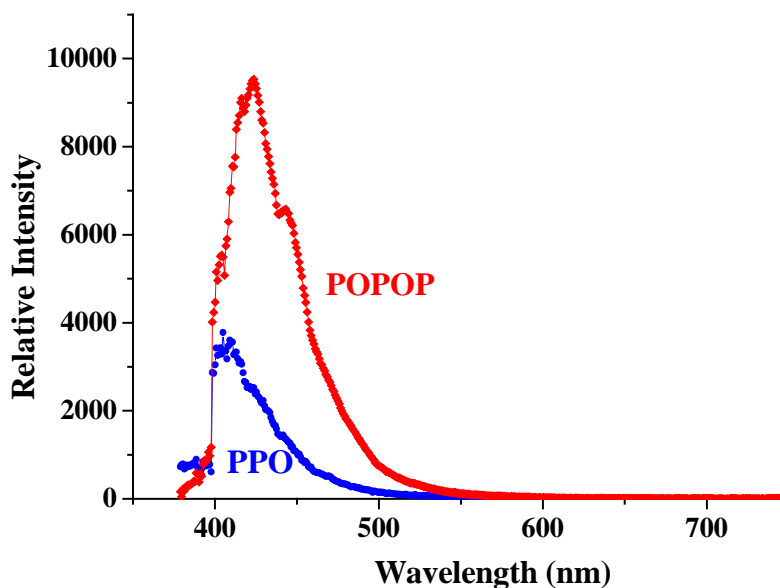


Fig. 4. Emission spectrum of PPO and POPOP

The light transmission rate through a membrane has a very significant impact on its scintillation detection efficiency, since the scintillated light has to travel inside the membrane structure to reach a PMT. Therefore, the scintillation membrane itself needs to be transparent or highly transmittable. As shown in Figure 5, the transmission rates of the prepared scintillation membranes are more than 85% near 425 nm, thus revealing a high transparency.

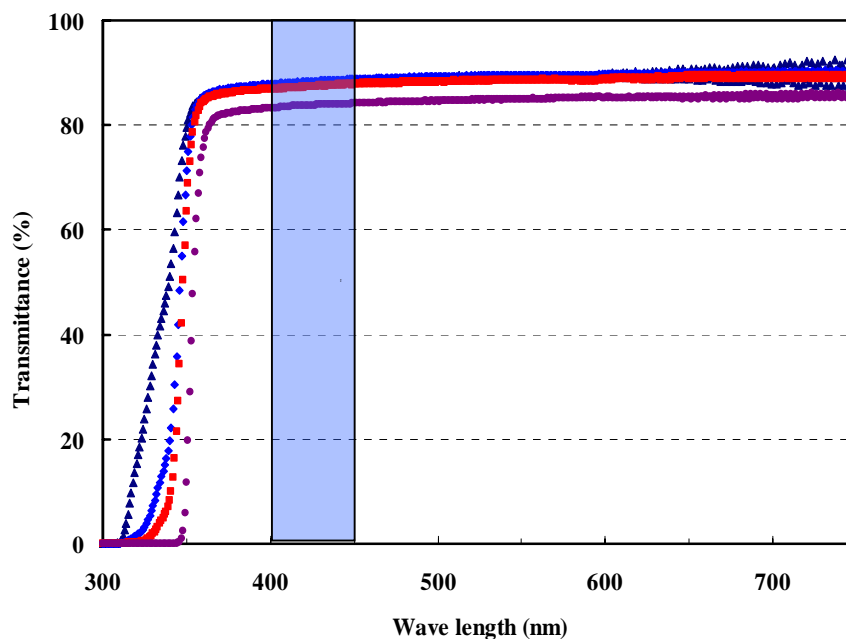


Fig. 5. Transparency of the scintillation membrane near a visible light

The results above demonstrate that the new scintillator membranes can be applicable to the detection measurement of α -particle emitting radionuclides. To evaluate the impact of the scintillator concentration on the detection efficiency, the relationship between the concentration of the scintillating agents and the corresponding counting results was investigated. Table I shows the variation in the preparation conditions. Even though the scintillating agents play a key role in the detection mechanism, they need to be regulated at a certain amount in the matrix because too high a concentration causes a reduction of scintillation efficiency due to the self-absorption of the scintillator against the scintillated light. As shown in Figure 6, the α -particle spectrum moves to the right with an increase of the PPO concentration.

An α -particle's penetration into a medium is limited within a short distance. Therefore, the membrane thickness is critical for optimizing its detection capacity. The membrane should be thick enough to react with most of the deposited α -particles, but also thin enough not to induce a significant quenching of the scintillated light. To optimized the membrane thickness, various membranes were prepared with thickness ranging from 150 to 300 μm , and the corresponding solidified membranes' detection capacity were compared with each other (See Figure 7). Until the casting thickness of the polymer solution reaches 250 μm , the detection capacity improves with an increase of the membrane thickness. Beyond 250 μm , however, the detection capacity decreases rather than increases, even though the amount of the overall scintillating agents is

increased with the thickness. These results indicate that the α -particle's penetration into these specific membranes is limited to less than 50 μm in a solid membrane, prepared with 300 μm of a solution casting thickness.

Table I. Preparation conditions for the organic scintillator-impregnated membrane

No.	Polymer Concentration (g)		Organic Scintillators (g)		Casting Thickness (μm)	Coagulation Method
	solvent (MC)	polymer (PSF)	primary (PPO)	secondary (POPOP)		
01	40	12.5	0.0188	0.00188	300	Evaporation
02	40	12.5	0.0375	0.00375	300	Evaporation
03	40	12.5	0.0500	0.00500	300	Evaporation
04	40	12.5	0.0625	0.00625	300	Evaporation

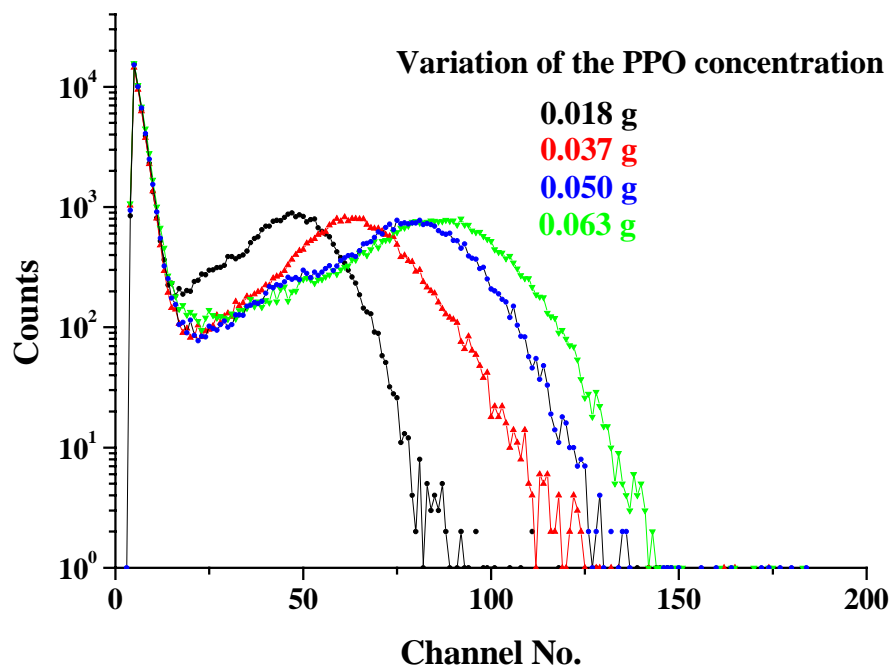


Fig. 6. Alpha spectrum variation of the scintillation membrane with the PPO concentration

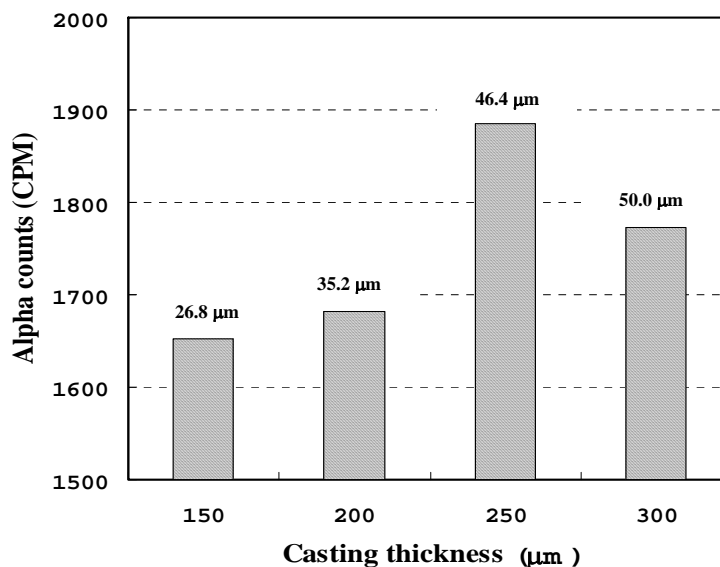


Fig. 7. Variation in radioactive detection capacity with membrane thickness

CONCLUSION

Scintillation plastic membranes were formulated to detect an α -particle contamination on the surface of nuclear wastes. The membrane is composed of a polysulfone matrix containing organic scintillators, its structure being solidified from a homogeneous casting solution. The efficiency of the α -particle detection of the prepared membranes is characterized with variations of the preparation conditions such as the concentration of the scintillation agents and the membrane thickness. The membrane shows a reliable detection capacity for the detection of the α -particle, and can be applied to the on-site estimation of a radioactive surface contamination.

REFERENCES

- [1] S. W. Wunderly and J. F. Quint, US Patent 4,916,320, April 10 (1990).
- [2] K. A. Schellenberg, US Patent 4,562,158, December 31 (1985).
- [3] L. F. Costa, D. C. Harrington, and R. S. Miller, US Patent 4,692,266 (1985).
- [4] C. G. Potter and G. T. Warner, "Scintillation counting of harvested biological samples with low energy beta emitters, using solid scintillant filters", In H. Ross, J. E. Noakes, and J. D. Spaulding (Eds.), *Liquid Scintillating Counting and Organic Scintillators*, Chelsea, Michigan, Lewis Publishers (1991).
- [5] M. J. Han, P. M. Bummer, and M. Jay, "Solid scintillation proximity membranes . Characterization of polysulfone-inorganic fluor morphologies precipitated from NMP Solutions", *J. Membrane Sci.*, 140, 235 (1998).
- [6] Myeong-Jin Han, Kune Woo Lee, Bum-Kyoung Seo, Preparation and characterization of a double-layered porous film to assay for surface radioactive contamination, *J. Membrane Sci.*, 223, 59 (2003).