Pore-Filled Scintillating Membranes For Simultaneous Alpha-Emitting Radionuclides Preconcentration And Detection

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1. Introduction

Continuous monitoring of radionuclides in the environment is important from health and safety point of view. Of these, presence of alpha emitting radionuclides is of more concern due to their high internal hazards. In environment, wherever low amount of these radionuclides is present, the detection and quantification step needs to be preceded by a preconcentration step, where the radionuclide is extracted and preconcentrated to give detectable signal. Various methods like ion exchange, solvent extraction and chromatographic techniques have been developed for actinide preconcentration but these methods have drawback of multiple steps involved in quantification of actinides. Simultaneous preconcentration and determination can be carried using extractive scintillating materials in which the organic phase containing extractant and scintillator is impregnated in polymer matrices. Various forms of extractive scintillating materials have been developed like fibers, beads and membranes [1]. However, one of the major problems associated with such materials, developed so far, is the loss of stability of the extractant and scintillator in the polymer phase during loading and elution cycle [1]. This loss of extractant during contact and elution will alter the results. Therefore, the stability of the extracting medium during the period of equilibration is essential. The systems can be made stable by replacing the mobile carrier in a liquid membrane with a covalently bonded carrier. The latter class of membranes is termed as a fixed-site/ pore-filled membrane. In this, functionalized monomer is covalently anchored on the polymer chains. Along with good stability, the advantage of the pore-filled membranes is high flux through these membranes.

In the present work, pore-filled scintillating membranes have been prepared for simultaneous preconcentration and detection of alpha emitting radionuclide. Two different types of base polymers (polypropylene (PP) and polyethersulfone (PES)) have been used for grafting the phosphate bearing monomer that is known for its good affinity towards actinides. The membrane composition has been optimized and its stability and reusability has been checked.

2. Experimental

The host PP and PES membranes (4 X 4 cm^2 size) were initially soaked in methanol for half an hour. Then the membranes were soaked for 3 h in grafting solutions containing: Bis [2-(methacryloxy)ethyl] phosphate (MEP) as extractant, 2,5-diphenyloxazole (PPO) as a primary scintillators and α,α`-dimethoxy-α-phenyl acetophenone (DMPA) as UV-initiator. In some membranes, 1,4-bis(2-methylstyryl)benzene (MSB) was also added in the grafting solution as a wavelength shifter. After soaking, the membranes were UV irradiated at 365 nm for 15 min. Membranes were then washed in DMF: water mixture for one hour and vacuum dried at 50^0C. The extent of grafting (%) was obtained gravimetrically as:

$$Grafting\ (\%) = \frac{(W_i - W_f)}{W_i}\ X\ 100 \qquad (1)$$

where, W_i and W_f are the weights of the membranes, before and after grafting respectively. The grafted membrane samples of 1cm × 3cm were used to equilibrate with ^{241}Am in pH 2 solution for overnight. The scintillation response of these ^{241}Am loaded membranes monitored by counting these samples in liquid scintillation counter in a fixed geometry as shown in figure 1.

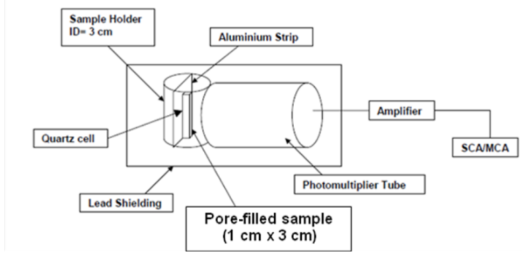


Fig. 1. Typical detection set-up used for membrane scintillation counting.

3. Results and Discussion

For both PP and PES membranes, an avera[…]fting of 140 % was obtained.

(a)

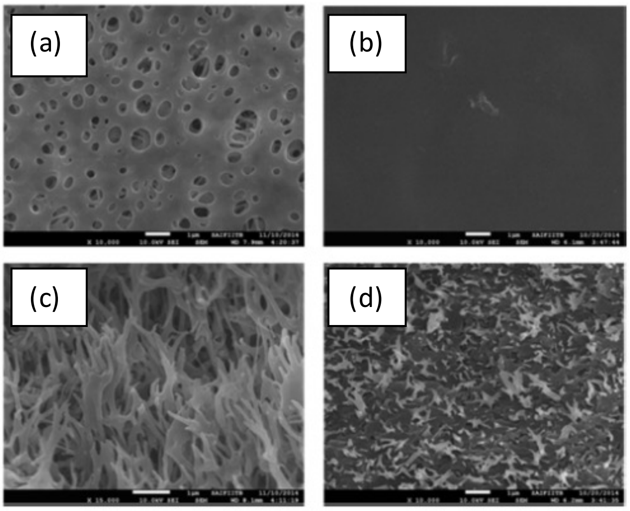
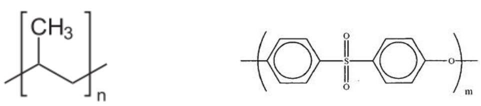


Fig 2. FEG-SEM images of (a) top, and (c) cross-section of the nascent membrane and (b) top, and (f) cross-section of the grafted PES membrane.

The mass gain (%) > 100 was obtained for both substrates showing a high degree of pore-filling. This is also evident from the FEG-SEM images in Fig. 2a-d. As seen from the images of the nascent and grafted membranes, the pore openings are completely and uniformly filled by the polymer microgel.

The counting efficiency of the ^{241}Am loaded grafted samples was obtained by comparing the membrane scintillation counts with the corresponding counts for liquid scintillation counting. For the same ^{241}Am activity loaded in both PP and PES samples, PES based membranes were found to have much higher counting efficiency (87 %) compared to PP sample (11 %) in a fixed geometry. This can be explained based on the difference in chemical structures of the two host membranes. PP membrane has aliphatic backbone whereas PES membrane has aromatic membrane as shown in Fig. 3.



PP PES

Fig. 3. Structures of (a) PP and (b) PES membrane.

The aromatic environment has been proven to be better for the fluorescence processes of absorption and emission [2]. This is because the π cloud of the aromatic ring provides a target for radiation interaction, capturing the energy of the incident particle more efficiently. This is also apparent from the pulse height spectra of the two membranes and a standard liquid scintillator (Fig. 4). For PP membrane, only low energy component and no peak has been observed while for PES membrane, although at lower energy than liquid scintillators, a peak is observed.

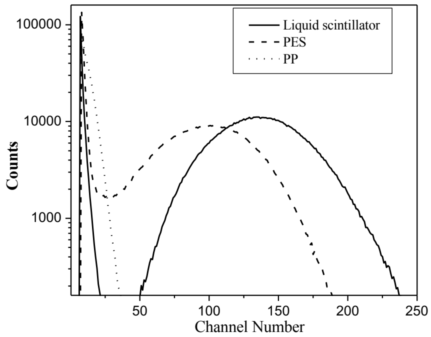


Fig.4.Pulse height spectra of grafted membranes and liquid scintillator.

To increase the scintillation efficiency of the PP based grafted membranes, wavelength shifter (MSB) was also added in some membranes during polymerization. But no effect on the scintillation efficiency of PP based membranes was observed. For PES membranes also, no change in the scintillation efficiency was observed due to addition of MSB. This can be explained based on the fluorescence spectra of these membranes. The emission of these membranes was found to be in visible region as seen in Fig. 5. Therefore, there was no significant change on addition of wavelength shifter in the membrane matrix. Due to the high efficiency of PES membranes, further studies were concentrated on these substrates.

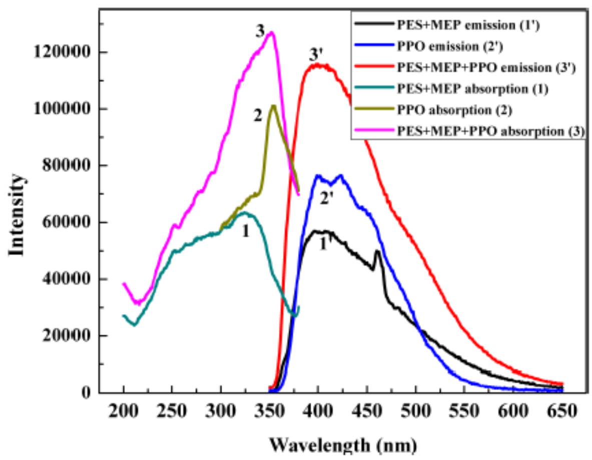


Fig.5. Absorption and emission spectra of PES membranes, with and without organic fluor, PPO, and of PPO solution.

The linear response of the membranes was checked by loading the membranes with different amounts of ^{241}Am activities as shown in Fig. 6 The slope of the line has been found to be 0.84, in concordance with the 87% counting efficiency of the membrane samples.

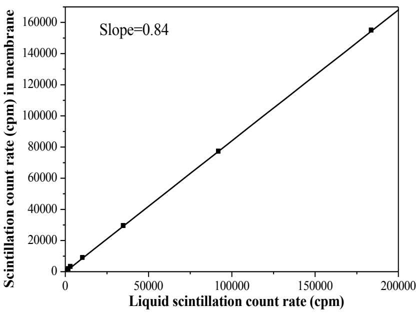


Fig. 6. Response of the PES grafted membranes for different activities.

The stability and reusability of the grafted PES membranes were also checked by subjecting same PES membrane with repeated cycles of ^{241}Am loading and deloading. At each step, the membrane was counted for its scintillation response. Figure 7 shows that even after three cycles, reproducible response of the membrane was obtained. This shows that the membrane is stable after repeated use, with no leaching of PPO.

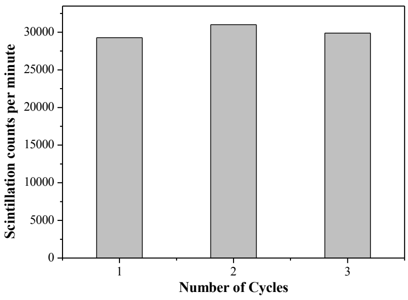


Fig. 7. Scintillation counts of PES membrane for its repeated use.

4. Conclusion

The present study shows that scintillating pore-filled membranes can be used to simultaneously preconcentrate and detect alpha emitting actinides. The studies show that the substrate of the pore-filled membrane plays important role in designing these pore-filled membranes. The membranes have been found to stable and reusable.

References

1. S. Sodaye, Y.M. Scindia, A.K. Pandey, A.V.R. Reddy, Sensors and Actuators B Vol. 123, pp.50,2007 and references therein.
2. W. Moser, W. F. Harder, C. R. Hurlbut and M. R. Kusnjzr, Radiat. Phys. Chem. Vol. 41, pp. 31, 1993