Efficient and ion-selective desalination of radioactive iodine species in water by using nano-adsorbents immobilized composite membranes

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1. Introduction

Efficient treatment of radioactive wastes arising from overuse of radioisotopes in medical institutes and nuclear reactor accidents has recently been an important issue, and thus much effort has been devoted to protecting the environment from hazardous radioactive wastes. Among them, radioactive iodines (e.g. I-131, I-129) in aqueous media is regarded as one of the most palpable threats to humans, because it is generated by post-use in medical and industrial applications. Despite recent progress in the development of advanced adsorbents for desalination of radioactive iodine, the establishment of a desalination method with high removal efficiency, ion-selectivity, and sustainability under continuous flow has not yet been reported. In this paper, we would like to report the fabrication and application of nano-hybrid composite membranes, composed of gold nanoparticles and cellulose acetate membranes, which have the potential to be used for efficient removal of radioactive iodine.[1]

2. Methods

2.1 Preparation of nanocomposite membranes

A commercially available cellulose acetate membrane (CAM) filter (pore size = 0.20 μm, diameter = 25 mm) was washed with 10 mL of deionized water by using a syringe. In the next step, 10 mL of citrate stabilized AuNPs (concentration: 10 nM, average size: 15 nm) was passed through the filter by using syringe at the rate of 1 mL/sec. The membrane filter was then washed with pure water several times to give a deep-wine colored filter. Gold nanomaterials were observed using FEI Verios 460L field emission scanning electron microscope (SEM) under the high performance conditions with accelerating voltages up to 15 kV. Elemental composition of gold nanomaterial was analyzed by SEM-energy dispersive X-ray (EDX) (AMITEC) analysis with accelerating voltages up to 20 kV. EDX spectrum was recorded in the area scan mode by focusing the electron beam onto a region of the sample surface. AuNPs immobilized cellulose acetate membrane (Au-CAM) filter was kept under ambient conditions until it was applied to the filtration experiment.

2.2 *Desalination of radioactive iodine using Au-CAM filters unit under a continuous in-flow condition*

To evaluate the efficiency of Au-CAM filter unit under a continuous flow condition, [^{125}I]NaI (3.7 MBq) was diluted with 50 mL of aqueous media (pure water, 1 x PBS, 1.0 M NaCl, 0.1 M NaOH, 0.1 M HCl, 10 mM CsCl, 10 mM, $SrCl_2$, synthetic urine, and seawater). An aqueous radioactive iodine solution was then passed through Au-CAM at an in-flow rate of approximately 1.5 mL/sec (by using a syringe pump or by hands equipped with lead gloves). The amount of residual radioactivity in the filtrate was measured by using a γ-counter (PerkinElmer, 2480 Automatic γ-counter).

3. Results

We have demonstrated simple methods for the fabrication of Au-CAM by using citrate-stabilized AuNPs and cellulose acetate membrane (Figure 1a). The surface of Au-CAM were observed by SEM images which showed that the nanomaterials were incorporated stably on the cellulose nanofibers (Figure 2). The nanoparticles incarcerated on the membrane were sustained stably and were not released from the membrane by continual washing with aqueous solutions such as 1.0 M NaCl. The adsorption capacity of an Au-CAM was approximately 12.2 μmol of iodide anion/g of AuNPs. To evaluate the desalination performance, Au-CAM filter was applied to a continuous in-flow desalination process. The radioactive iodine solutions (3.7 MBq/50 mL) were passed through a filter unit containing Au-CAM at an in-flow rate of 1.5 mL/s (Figure 1b). The amount of the residual radioactivity in the filtrate were measured by using a γ-counter. The removal efficiency (%) was defined by the following equation (1).

$$\text{Removal efficiency (\%)} = (C_0 - C_e)/C_0 \times 100 \qquad (1)$$

(C_0: the concentration of radioactive iodine before filtration step; C_e: the concentration of radioactive iodine after filtration step)

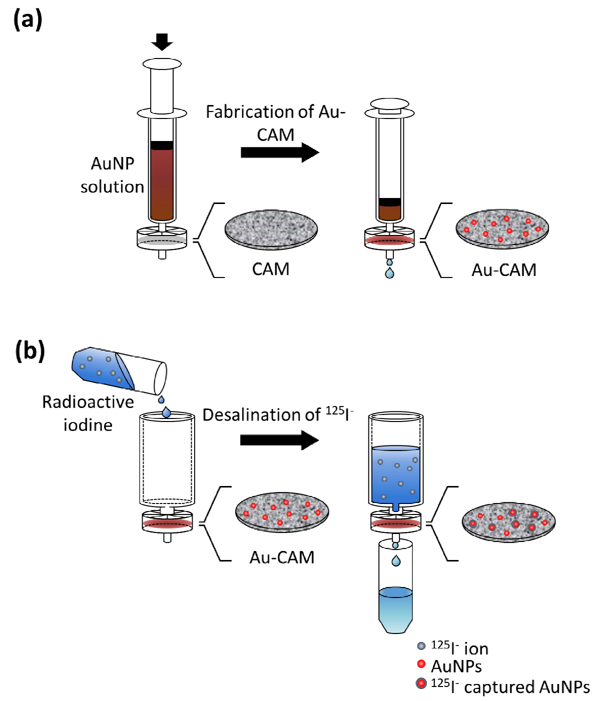

Figure 1. (a) Preparation of Au-CAM, (b) Desalination of radioactive iodines using Au-CAM

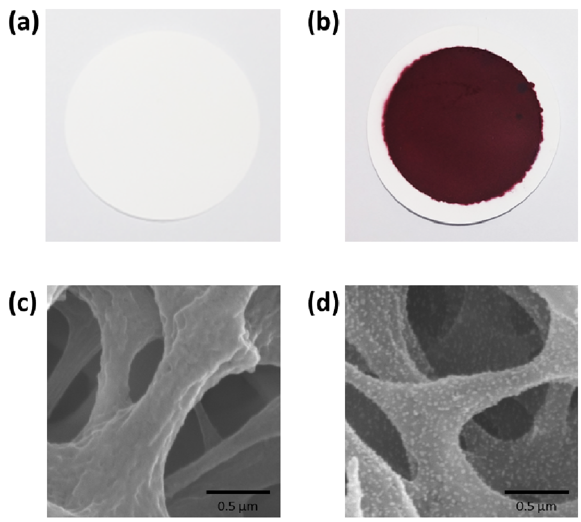

Figure 2. A photographic image (a) cellulose acetate membrane (diameter 47 mm), (b) Au-CAM (diameter 47 mm), SEM image (c) cellulose acetate membrane, (d) Au-CAM.

As shown in figure 3, the concentration of radioactive iodine was decreased significantly and the excellent efficiency was obtained through a simple filtration step. In particular, the desalination performance of Au-CAM was not suppressed by high concentration of inorganic salts such as sodium, cesium, and strontium and several organic substances. In all cases, the removal efficiency of Au-CAM was higher than 99.5%. Au-CAM showed high removal efficiency under neutral and basic condition (up to pH 13), however, it was dropped to ca. 90% under acidic condition (pH 1). Furthermore, Au-CAM could be reusable for repetitive desalination of radioactive iodine in a synthetic urine and seawater. During the consecutive filtration process, more than 99% of radioactivity in aqueous media was captured efficiently by using a single Au-CAM filter unit.

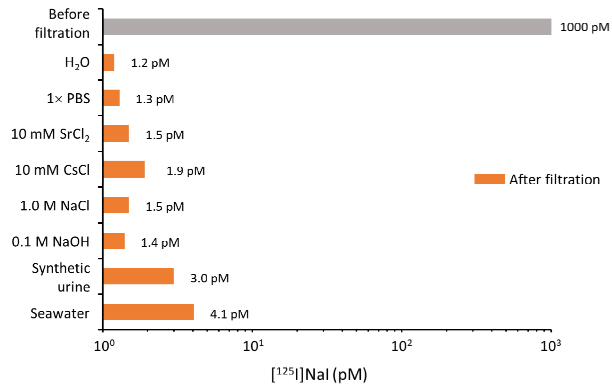

Figure 3. Filtration of radioactive iodine anions in several aqueous solutions using Au-CAM

4. Discussion

In this study, we demonstrated highly useful method for rapid and efficient separation of radioactive halogen species. By using citrate-stabilized AuNPs and a commercially available cellulose acetate membrane, Au-CAM can easily be prepared and the fabrication step is highly reproducible. As iodide anions is spontaneously chemisorbed on the surface of AuNPs, Au-CAM can be applied to the remediation of radioactive iodines in various aqueous media.[1-4] The reactivity of I-125 is identical with other iodine isotopes, and thus this method will be utilized to remove more hazardous radioelements such as I-129 and I-131. In the presence of high concentration of competing anions such as phosphate, chloride, and hydroxide, the nano-hybrid membrane (Au-CAM) showed excellent desalination efficiency and good reusability. Another significant advantage is that immobilized nanoparticles on a cellulose acetate membrane is stable under high salt conditions and varied pH. It appears like that AuNPs on the membrane of carbohydrate were stabilized by oxygen atom containing functional groups including hydroxyl and carbonyl groups. Thus, the hybrid membrane can be stored for several weeks without loss of its performance and chemical stability.

5. Conclusion

The continuous process in the present study is superior to conventionally used methods in terms of removal efficiency, ion-selectivity and reusability. By using a single Au-CAM (diameter: 25 mm), ca. 90 mL of aqueous waste can be purified in a minute. It is anticipated that a lot of Au-CAM filters will be easily produced in a short time, because large-scale synthesis and characterization of the citrate-stabilized AuNPs were well-established. Taken together, Au-CAM will be a promising adsorbent system worth to investigate for the practical remediation of industrial and medical radioactive iodine wastes.

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