Temperature Sensitive Strippable Coating for the Removal of Radioactive Cesium From Contaminated Surface

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1. Introduction

The nuclear accident at the Fukushima Daiichi nuclear power station in 2011 released a huge quantity of radioactive contaminants into atmosphere. They were deposited by wind and rain on the various surfaces of objects such as soil, road, and building in terrestrial and city areas [1]. Various surface decontamination methods using tools such as scabbling and milling have been tested for the removal of ^{137}Cs from contaminated surfaces. A strippable coating that can be easily peeled off from the surface upon drying is a widely used method to avoid the destruction of the original surface [2]. Although strippable coating showed good effectiveness, it becomes radioactive waste itself after use, requiring costly waste disposal. Therefore, new surface decontaminants that produce a low amount of radioactive waste after use are still desired.

In the present study, we developed a new strippable coating consisting of magnetic adsorbents having Cs adsorption ability [3] and an polyvinyl alcohol-borate based hydrogel for the removal of ^{137}Cs from contaminated surfaces and the volume reduction of radioactive waste after use.

2. Experiment

2.1 Fabrication of adsorbent/PVA-borate-based surface decontaminant

Magnetic adsorbents were synthesized by following the procedure previously reported by our group (Yang et al. 2016b). Briefly, 2.7 g of FeCl3•6H2O and 7.2 g of sodium acetate were dissolved in 100 mL of ethylene glycol. Then, the mixture solution was transferred to a Teflon-lined stainless-steel autoclave. The autoclave was sealed and heated to 200 °C for 8 h. After washing with excess ethanol and deionized water several times, the aqueous solution of magnetic nanoparticles (0.5 mg/mL) at pH 2 was reacted with a 2.5 mM $K_4[Fe(CN)_6]$ solution. After stirring for 1 h, the black particles were washed with deionized water several times. Finally, the product was dried in vacuum at 60 °C overnight.

For the fabrication of the surface decontaminant, the desired amount of sodium borate decahydrate was first dissolved in water or 0.1 M NH_4Cl solution. Then, the PVA(avg Mw 13000 – 24000) was dissolved by stirring and heating at 80 °C in sealed vials to prevent water from evaporating. The wt% of PVA ranged from 4 to 6 wt%, and the wt% of borax was fixed at half that of PVA. Finally, the magnetic adsorbents were added to the PVA-borate hydrogel solution.

2.2 Surface decontamination procedure .

Cement was deposited on the surface of a planchet (diameter = 4 cm) and then coated with paint (Ilshin Co. Ltd., South Korea) to serve as a model surface. The painted cement was contaminated with ^{137}Cs by dropping and evaporating a known amount of ^{137}Cs solution on the surface. While the temperature of decontamination solution was maintained at 50°C, the contaminated surface was treated with decontamination solution. Within a few minutes, an adsorbent/PVA-borate hydrogel film was generated and then peeled off the surface 3 h later. The radioactivity of the painted cement before and after treatment was measured using an automatic low-background Alpha/Beta counting system. Then, the adsorbent/PVA-borate hydrogel film was added to 20 g of water, and the adsorbent was magnetically collected using an external magnet.

3. Result and Discussions

It has been recently discovered that PVA-borate hydrogel have temperature sensitive phase transition behavior [4], which allows them to be peeled from a surface after surface decontamination. For the surface decontamination, the aqueous polymeric solution containing adsorbent, PVA and borate at 50^{o}C was applied to the contaminated surface by brushing to solubilize the ^{137}Cs. Then, the strippable PVA-borate hydrogel/adsorbent composite film was formed through reversible the cross-linking of the PVA with borate at room temperature for easy removal. After surface decontamination, the adsorbent capturing the ^{137}Cs was magnetically separated for the volume reduction of radioactive waste.

Fig. 1(a)) show that magnetic nanocluster was successfully coated with gray cubic shapes of PB (XRD data in fig. 1(b)). It exhibited superparamagnetic behavior, and the saturation magnetization of the magnetic nanoadsorbent at 1.5 T was 27.8 emu/g, which is sufficiently high to collect the adsorbent in water by using an external magnet. (fig 1(c)).

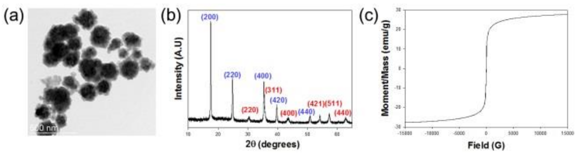

Fig. 1. (a) TEM image and (b) XRD pattern of the magnetic adsorbent (red: Fe_3O_4, blue: Prussian blue), and (c) Magnetization curve of magnetic adsorbent, as measured using a vibrating sample magnetometer.

To evaluate the decontamination ability of the adsorbent/PVA-borate complex, 137Cs-contaminated painted cement was used as a model surface. After generation of adsorbent/PVA-borate hydrogel film at room temperature, the films were easily removed from the surface (Fig. 2(a)). Fig. 2(b) and (c) show that the adsorbent/PVA-borate hydrogel film was completely removed from the surface in one piece and did not leave any residue.

The strippable coating displayed a good removal efficiency of 83.46% and a DF of 6.05 from the ^{137}Cs-contaminated surface due to the presence of the adsorbent, which had a large distribution coefficient for Cs (3.34×10^{4} mL/g). Moreover, the PVA-borate complex can be reused following a simple addition of water via the magnetic separation of the adsorbent, which can capture 99.071% of the ^{137}Cs in the used hydrogel film.

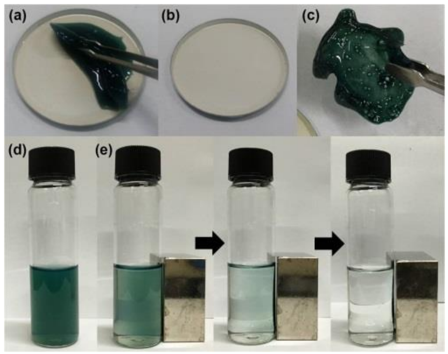

Fig. 2. Digital images showing (a) the stripping coating on a ^{137}Cs-contaminated surface, (b) lack of film residue of the adsorbent/hydrogel film on the surface after treatment, (c) the stripped adsorbent/hydrogel film, (d) aqueous solution of stripped coating (e) magnetic separation of adsorbent from solution using an magnet.

4. Conclusion

In this study, temperature sensitive hydrogel based strippable coating was successfully fabricated for the surface decontamination. The films displayed a good removal efficiency of 83.46% from the ^{137}Cs-contaminated surface. Thus, our strippable coating has good potential as a new surface decontaminant.

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