Fabrication of stimuli-sensitive hydrogel for the removal of cesium

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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 the environment.[1] Among these, cesium Cs-137 is the most problematic contaminant due to its long half-life (30.2 years), and high-energy gamma ray (γ-ray) emissions.[2] Various surface including road, roof, house, building were contaminated with Cs-137. For the decontamination of the surface, various coating materials were applied to remove the Cs-137 from the surface and showed good degree of surface decontamination property. However, these coating materials have some problems and limitation such as toxic component, and lack of reusability of materials related to the cost. Thus, a more cost-effective and environmental friendly coating materials is still desired.[3]

In the present study**,** the stimuli-sensitive hydrogel were fabricated for the removal of radioactive Cs from solid surface. We describe the morphology, structure, and physical property of these stimuli sensitive hydrogel. In addition, their ability to eliminate cesium was also evaluated.

2. Methods

Various amounts of acrylamide were dissolved in aqueous solution of sodium alginate. The concentration of sodium alginate was 4 wt%. And the concentrations of acrylamide were varied in the range of 5 wt% to 14 wt%. The N,N'-methylenebisacrylamide(MBAA), ammonium persulphate (APS), and N,N,N',N'-tetramethylethylenediamine (TEMED) were used as a crosslinking agent, thermo-initiator, and accelerator for the synthesis of polyacrylamide, respectively. These reactants were subsequently added into the solution. At 25^{o}C, after about 1 hour, the mixture became a homogeneous and transparent solution. The solution was transferred into a glass mould, and then the mould was put in an oven at 50 ^{o}C for 3 hours. This step produced Na-alginate/PAAm hydrogels.

The morphology change of stimuli-sensitive hydrogel was evaluated by addition of calcium ion into the upper synthesized Na-alginate/PAAm hydrogel. The Na-alginate/PAAm hydrogel solution was soaked in a various concentration of calcium ions solution at room temperature for 3 hours. In this step, the external solution and the hydrogel exchange ions: divalent ions diffuse from the external solution to the hydrogel, and Na^{+} ions diffuse from the hydrogel to the external solution, resulting in highly homogeneous and transparent Ca-alginate/polyacrylamide hydrogel film. For the fabrication of surface decontamination agent, adsorbent having adsorbing property for radioactive cesium were added during the synthesis of the stimuli-sensitive alginate/polyacrylmaide hydrogel.

To measure the swelling ratio (SR) of the adsorbent/hydrogel beads, the beads was freeze-dried (W_d). After the dried bead was immersed in distilled water for 24 h and then the surface moisture was removed by using a filter paper before weighing (W_s).

3. Results

The Fourier transform infrared (FT-IR) spectra of stimuli-sensitive hydrogel after addition of various concentration of calcium ion are shown in Figure 1. Figure 1 showed the characteristic peaks of the alginate and polyacryalmide, indicating Ca-alginate/ polyacryladmie hydrogel were successfully synthesized. Figure 2 showed the digital images of stimuli-sensitive hydrogel films after addition of calcium ions. It can be seen that the hydrogel film were successfully formed by any concentration of calcium ions without NH_4Cl which is widely used materials as a chemical agent to desorb the cesium from the contaminate surface. However, the formation of hydrogel film was interfered by NH4Cl even after addition of calcium ion due to the screen effect of NH_4Cl for the formation of calcium-alginate network. Therefore, the optimum condition of concentration of NH_4Cl needs to be selected to meet two requirements, formation of hydrogel film and good desorption of cesium from the surface.

The water absorbing property of dried stimuli-sensitive hydrogel film was evaluated in terms of the swelling ratio by the following equation.

$$SR\ (\%) = (W_s - W_d)/W_d \times 100$$

The Ca-alginate/polyacrylamide hydrogel films showed various SR values according to the amount of reactant such as acrylamide. As the weight ratio of alginate to acryladmie was increased, the SR values of stimuli-sensitive hydrogels were increased. As the concentration of calcium ions was increased, however, the SR values of stimuli-sensitive hydrogel were deceased. From the SR data, dried stimuli-sensitive hydrogel are potentially useful to absorb the water containing the radioactive cesium desorbed from the contaminated surface by

chemical agent such as NH_4Cl. It is anticipated that the removal efficiency of adsorbent embedded stimuli-sensitive hydrogel are increased as a weight ratio of adsorbent to hydrogel increased. Therefore, adsorbent/hydrogel beads prepared at largest weight ratio of were chosen for further studies.

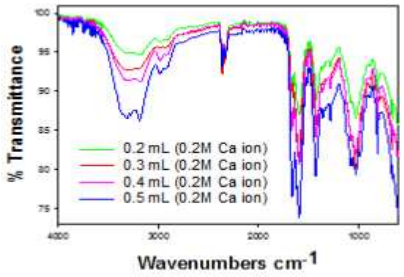

Fig. 1. FTIR spectra of stimuli-sensitive hydrogels containing alginate/polyacrylmiade after addition of various concentration of calcium ions.

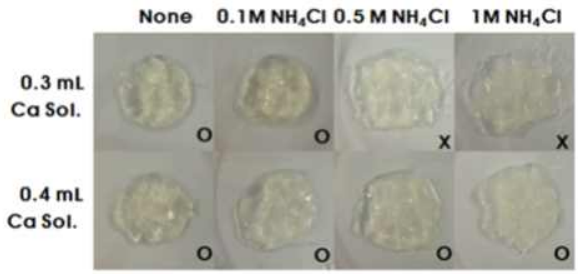

Fig. 2. The digital images of stimuli-sensitive hydrogel films after addition of calcium ions.

Next, the Cs removal efficiency of the stimuli-sensitive hydrogel was examined. Various concentrations of the hydrogel were applied onto the contaminated surface containing Cs. The stimuli-sensitive hydrogel exhibited good removal efficiency for cesium form the surface.

4. Conclusions

In this study, we fabricated stimuli-sensitive hydrogel coating materials for the removal of cesium from the contaminated surface. The smart hydrogel ocating materials showed an excellent morphology change from the liquid to film by addition of Ca ion. Therefore, the stimuli-sensitive hydrogel demonstrated good potential for the treatment of contaminated surface for the removal of radioactive cesium.

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