

Reversibly Cross-Linked and Strippable Hydrogels for the Removal of Radioactive Cesium From Surface

Hee-Man Yang*, Chan Woo Park, Kune-Woo Lee, Ilgook Kim, and In-Ho Yoon
Decommissioning Research Division, Korea Atomic Energy Research Institute, 111,
Daedeok-daero 989beon-gil, Yuseong-gu, Daejeon, Korea
*hmyang@kaeri.re.kr

1. Introduction

Radioactive materials have been extensively used in various industrial and research fields, particularly for nuclear research and industry. During their utilization, various surfaces, such as concrete, steel, and paint, in most nuclear facilities become contaminated with radioactive materials. Thus, surface decontamination is a very important technology for radioactive material utilization and even for decommissioning nuclear facilities because surface contamination can be transferred to workers by contact, and it may become airborne if not treated [1]. Meanwhile, the nuclear accident at the Fukushima Daiichi nuclear power station in 2011 unfortunately released large amounts of radioactive cesium into the environment, which also affected various urban structures, such as roofs, building exterior surfaces, and roads in a wide area [2]. Although current surface decontamination methods, including physical or chemical methods, in nuclear facilities are well established, surface decontamination for wide-area remediation after nuclear accidents is still challenging due to some drawbacks of the current surface decontamination techniques [3]. Consequently, new surface decontaminants are still desired for a facile and cost effective preparation and one-step surface decontamination.

2. Experiment

2.1 Fabrication of adsorbent

Magnetic adsorbents were synthesized by following the procedure previously reported by our group [4]

2.2 Fabrication of surface decontaminant

The surface decontaminant was prepared by mixing a Polyvinyl alcohol (PVA) solution containing PB or

adsorbent with a borax solution. First, the PVA was dissolved in an NH_4Cl solution by stirring and heating at 80°C in sealed vials to prevent water from evaporating. Then, the desired amount of PB or adsorbents were also added into the PVA solution. Finally, the aqueous borax solution was added dropwise by stirring (vortex apparatus). In addition, the molar concentration of NH_4Cl in the solution was fixed at 0.1 M.

2.3 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. The contaminated surface was treated with decontamination solution. Within a few minutes, an adsorbent/hydrogel film was 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

The storage modulus (G') of the PB/PVA-borate hydrogel was measured to evaluate the strippable property of the hydrogel at room temperature. Although the PB/PVA-borate hydrogel exhibited a gel-like nature with a strong elastic character, the values of G' and G'' decreased compared with those of the PVA-borate hydrogel (Fig. 3(c) and Table 1). As the wt% of PB increased in the PB/PVA-borate hydrogel, the values of G' and G'' decreased. This is probably due to the expansion of the PVA-borate network due to the electrostatic repulsion between the negatively charged

PB and the negatively charged PVA-borate network induced by the borate ion ($B(OH)_4^-$) during the PVA-borate complexation [16]. Although the presence of PB in the PVA-borate hydrogel decreased the values of G' and G'' , the values are still high enough that it can be anticipated that the PB/PVA-borate hydrogel has good peeling-off properties that make it suitable for surface decontamination applications.

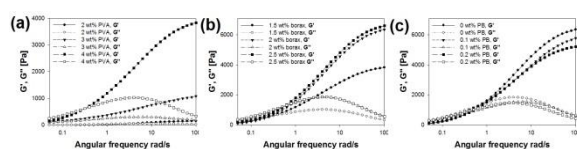


Fig. 1. Frequency sweep curves of the (a) PVA-borate hydrogel formed using 1.5 wt% borax and various wt% PVA solutions, (b) PVA-borate hydrogel formed using 4 wt% of PVA and various wt% borax solutions, and (c) PVA-borate/PB hydrogel formed by using 4 wt% PVA, 2 wt% borax and a 0.1 or 0.2 wt% PB solution.

To evaluate the decontamination ability of the PB/PVA-borate hydrogel, ^{137}Cs -contaminated stainless steel, aluminum, and painted cement were used as model surfaces. After 3 h of application of the PB/PVA-borate hydrogel onto the various contaminated surfaces, the hydrogel was removed from the surfaces. Fig. 5(a) shows that the PB/PVA-borate hydrogel was completely removed from the surfaces in one piece and did not leave any residue. As shown in scheme 1, the ^{137}Cs that was present on the contaminated surfaces was solubilized by the ion exchange of NH_4^+ for Cs^+ and was further captured by the PB, which has an adsorption selectivity for Cs in the hydrogel, until the PB/PVA-borate hydrogel was removed. The DF and R values are listed in Table 1. Surface decontamination using distilled water without the PB/PVA-borate hydrogel was also conducted using the same procedure as a control. The R values for the surface decontamination using water were 53.27% for the painted cement surface, 87.78% for the aluminum surface, and 72.54% for the stainless steel surface. The lower R value for the painted surface compared with the other surfaces is attributed to the rough surface of the painted cement, whereas the other samples had smooth surfaces. All of the R values increased when the PB/PVA-borate hydrogel was used to treat the contaminated surfaces, which indicated that the PB/PVA-borate hydrogel was more effective than conventional surface decontamination using distilled water. These results were attributed to the good surface

exchange properties of NH_4^+ for Cs^+ and the excellent Cs adsorption property of PB.

Table 1. Removal performance of the PB/PVA-borate hydrogel on ^{137}Cs -contaminated surfaces of painted cement, aluminum, and stainless steel (A_0 is the initial radioactivity before decontamination, and A_f is the final radioactivity after decontamination)

		$R = (A_0 - A_f) / A_0 \times 100$	$DF = A_0 / A_f$
Painted cement	Water	53.27	2.14
	Coating	64.91	2.85
Al	Water	83.90	6.21
	Coating	89.85	9.85
Stainless steel	Water	72.54	3.64
	coating	85.09	6.71

4. Conclusion

In this study, a hydrogel displayed a good removal performance for ^{137}Cs -contaminated aluminium and stainless steel surface, and enhanced removal efficiency for painted cement surface compared with water washing. Furthermore, the simple preparation, reusability of hydrogel make the material convenient, eco-friendly, and cost-effective. Therefore, hydrogel has good potential as a new surface decontaminant

Acknowledgement

This research was supported by the Korea Ministry of Education, Science, technology Grant (No.2017M-2A8A5015148)

REFERENCES

- [1] E. Calarese, Proc Natl Acad Sci U S A 108, 19447-19448 (2011).
- [2] N. Kinoshita, K. Sueki, J. Sasa, J. Kitagawa, S. Ikarashi, T. Nishimura, Proc. Natl. Acad. Sci. USA 108, 19526-19529 (2011).
- [3] H. N. Gray, B. Jorgensen, D. L. McClaugherty, A. Kippenberger, Ind. Eng. Chem. Res. 40 3540-3546 (2001).
- [4] Yang, H.-M., Jang, S.-C., Hong, S. B., Lee, K.-W., Roh, C., Huh, Y. S., Seo, B.-K., (2016). J. Alloy. Compd. Vol. 657, p. 387-393.