

Rheological and Uranium Decontamination Property of Chemical Gels

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Abstract: The rheological and decontamination behaviors of chemical gels prepared in a Ce(IV)–HNO₃ or HNO₃ based solution for tile surfaces contaminated with uranium radionuclides were investigated. A chemical decontamination gel was prepared by adding gelling agents composed of a stable pyro silica viscosifier and PEG-based non-ionic coviscosifiers, into a Ce(IV)–HNO₃ solution or HNO₃ solution.

Removal percentage of uranium with a coviscosifier type is high according to the following order: diethylene glycol hexyl ether (DGHE) > tripropylene glycol dodecyl ether (TPGDDE) > tripropylene glycol butyl ether (TPGBE). It maybe was caused by the difference in the length of hydrophobic alkyl chains of coviscosifier.

The chemical gel prepared in a HNO₃ based solution shows a higher removal efficiency of 89-98% over five decontamination steps and a satisfactory rheological property in removing uranium contaminants from a tile surface, compared to that in a Ce(IV)–HNO₃ based solution.

Key words: Uranium, Gel, Decontamination, Cerium, Nitric acid, Diethylene glycol hexyl ether (DGHE), Tripropylene glycol dodecyl ether (TPGDDE), Tripropylene glycol butyl ether (TPGBE)

1. Introduction

Chemical decontamination technology represents a highly effective removal of radioactive contamination through chemical dissolution and/or a redox reaction. However, the production of large amounts of waste limits its in-situ use. It is necessary to develop processes using chemical gels instead of chemical solutions, in order to avoid the well-known disadvantages of chemical decontamination techniques while retaining their high decontamination efficiency [1-3].

This method is effective in situations in which long contact times are required, and when a need to minimize waste exists. A chemical decontamination gel can be prepared by adding gelling agents composed of a viscosifier and a coviscosifier to the chemical decontamination agents used in traditional decontamination processes [4].

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A chemical gel decontamination process consists of applying a gel by spraying it onto any vertical or complex surface of the large area components to be decontaminated, as shown in Fig. 1. The gel adheres to the surface due to its thixotropic properties and operates by dissolving the radioactive deposit, along with a thin layer of gel support, so that the radioactivity trapped at the surface can be removed. Upon drying, this gel forms a strong film that can be peeled from the surface. The appropriate combination of viscosifier and coviscosifier is a very important factor in the control of the viscosity and adhesion properties of chemical decontamination gels [5].

Gelling agents were prepared by adding PEG-based non-ionic coviscosifiers (diethylene glycol hexyl ether and tripropylene glycol butyl ether, and tripropylene glycol dodecyl ether) into a stable pyro Si viscosifier. The decontamination and rheological behaviors of chemical gels prepared in a Ce(IV)-HNO₃ or HNO₃ based solution for tile surfaces contaminated with uranium radionuclides were investigated.

2. Experimental

A Ce(IV)-HNO₃ chemical decontamination agent was prepared by dissolving 0.5M Ce(IV) in 2M concentrated nitric acid. The gelling agents were composed of a viscosifier and a coviscosifier. Pyro Si(CAB-O-SIL M-5), which is stable in an acidic medium, and is easily gellated in small amounts compared to Al, was selected as a viscosifier and used in a 5-10 wt.% concentration during the experiment.

As shown in Figure 1, SEM images for the Pyro Si exhibited a network structure, which consists of the primary particles with a size of 30-50nm in diameter. As a coviscosifier, diethylene glycol hexyl ether (DGHE), tripropylene glycol butyl ether (TPGBE), and tripropylene glycol dodecyl ether (TPGDDE) were selected among PEG-based non-ionic surfactants, which are chemically stable and easily dissolved, and were tested within a range of 0.1-1.0 wt.%. The physical properties of the viscosifier and 3 coviscosifiers are shown in Table 1.

A chemical decontamination gel was sprayed onto the surface of a tile specimen contaminated with uranium radionuclides. The gel adheres to the surface of the specimen and operates by dissolving a radioactive deposit, along with a thin layer of the gel support, so that the radioactivity trapped at the surface can be removed. The efficiency of the radioactivity removal from the surface of the tile sample, expressed by the decontamination factor (DF), was calculated by measuring the radioactivity concentration of uranium radionuclides using MCA (Canberra, 2025).

Thixotropy, defined as a decrease in the apparent viscosity under stress, followed by a gradual recovery at rest, is a rheological phenomenon. The rheogram curves at various shear rates according to the gel formulations were obtained using a rheometer (Brookfield Eng. & Lab. Inc., R/S-CPS plus) and Rheo-3000 software.

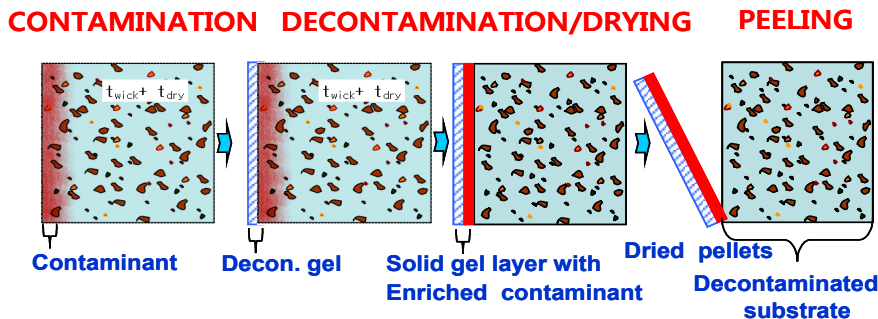


Figure 1: Surface decontamination using chemical gel.

Table 1
Physical properties of the viscosifier and coviscosifiers

Viscosifier (Fumed silica, CAB-O-SIL M-5)	BET S/A (m ² /g)	pH	Size (nm)
	200	3.7-4.3	200-300
Surfactant	Structure		HLB
DGHE (Diethylene glycol hexyl ether)	$\text{HO} \left(\text{CH}_2 \text{CH}_2 \right)_2 \text{C}_6 \text{H}_8$		11
TPGBE (Tripropylene glycol butyl ether)	$\text{HO} \left(\text{CH}_2 \text{CH}_2 \right)_3 \text{C}_4 \text{H}_9$		15.4
TPGDDE (Tripropylene glycol dodecyl ether)	$\text{HO} \left(\text{CH}_2 \text{CH}_2 \right)_3 \text{C}_{12} \text{H}_{25}$		10.6

3. Results and Discussion

Figure 2 provides a schematic rheogram of various chemical gels, which shows the effects of the formulation on the rheological properties. After shearing for 60s at a shear rate of 500 s⁻¹ simulating the spraying conditions and a consecutive shear rate of 5 s⁻¹ representing a stationary state is imposed. Chemical gels have a viscosity ranging from 4,000 to 8,000 cP at 5 s⁻¹ depending on the gel formulation, and are high in viscosity according to the following order: DGHE>TPGBE>TPGDDE. This is due to the difference in the length of hydrophobic alkyl chains, except TPGDDE, which have a difficulty in dispersing in the solution. However a re-build time of below 5 seconds for all chemical gels was attained.

As shown in Figure 3, the removal percentage of uranium with the number of decontaminations for a Ce(IV)-HNO₃ solution containing a various coviscosifier is high according to the following order: DGHE (88-98%)> TPGDDE (87-96%)> TPGBE (83-91%), owing to the difference in the length of hydrophobic alkyl chains of coviscosifier, and initial radioactivity level of U-contaminated specimens (DGHE: 24.5Bq/g, TPGDDE:59.0 Bq/g, TPGBE:33.6 Bq/g).

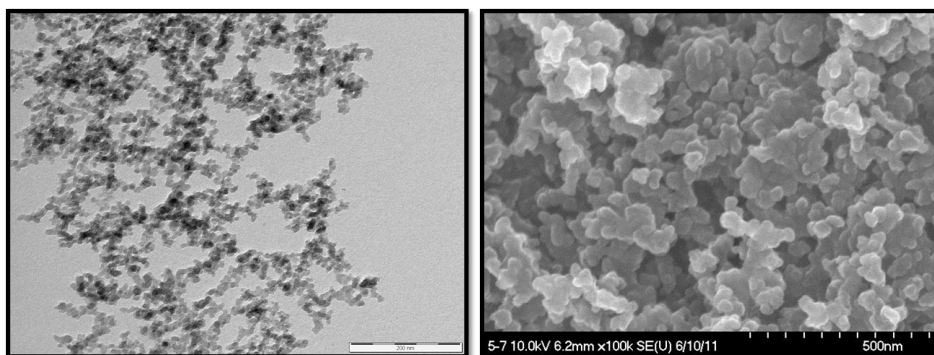


Figure 2: TEM & SEM images for the pyro Si, M-5.

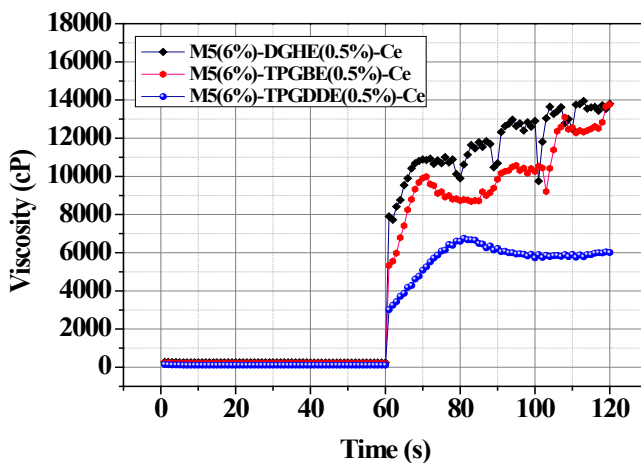


Figure 3: Rheogram of various chemical gels: shearing for 60s at a shear rate of 500/s followed by a shearing for 60s at 5/s.

Figure 4 shows the removal of uranium with the number of decontaminations for a Ce(IV)-HNO₃ and HNO₃ solution containing a TPGDDE coviscosifier. A chemical gel containing a HNO₃ solution, compared with Ce(IV)-HNO₃ gel, is more effective as a

decontaminating property except for the first decontamination step. The HNO_3 -based chemical gel shows a high removal efficiency of 89-98% over five decontamination steps for the uranium contaminations.

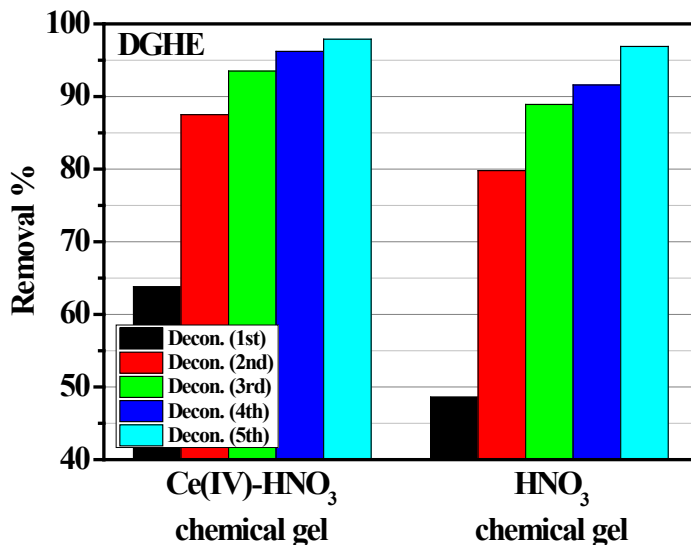


Figure 4: Removal percentage of uranium with the number of decontaminations for a Ce(IV)-HNO_3 & HNO_3 solution containing a DGHE coviscosifier.

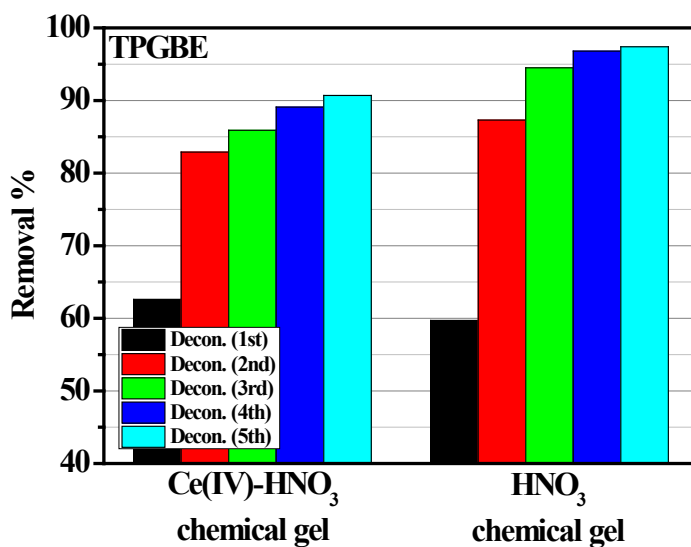


Figure 5: Removal percentage of uranium with the number of decontaminations for a Ce(IV)-HNO_3 & HNO_3 solution containing a TPGBE coviscosifier.

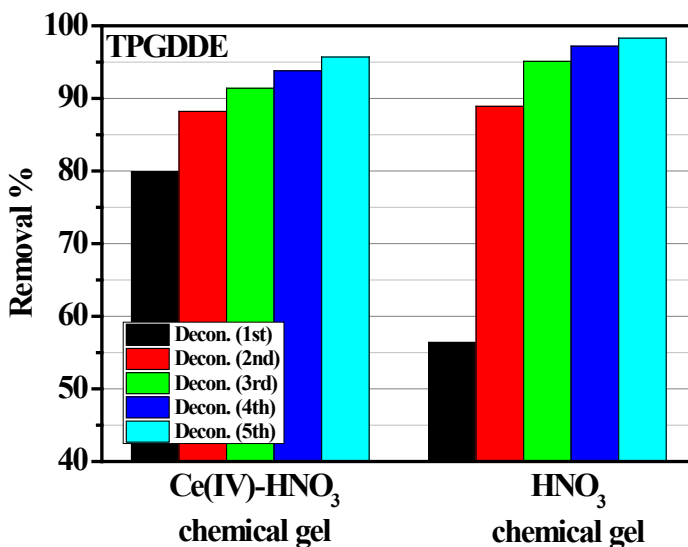


Figure 6: Removal percentage of uranium with the number of decontaminations for a Ce(IV)-HNO₃ & HNO₃ solution containing a TPGDDE coviscosifier.

4. Conclusions

The decontamination and rheological behaviors of chemical gels for the surfaces of tile contaminated with uranium radionuclides were investigated. A chemical decontamination gel composed of a pyro silica viscosifier and PEG-based non-ionic coviscosifiers in a Ce(IV)-HNO₃ solution or HNO₃ solution was prepared. Removal percentage of uranium with various coviscosifiers is high according to the following order: DGHE>TPGDDE>TPGBE₃, owing to the difference in hydrophobic alkyl chain length of coviscosifier, and initial activity level of uranium specimens.

The HNO₃ solution based chemical gel shows a high removal efficiency of 89-98% over five decontamination steps and a satisfactory rheological property in removing uranium contaminants from a tile surface.

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