Asian Journal of Chemistry; Vol. 28, No. 6 (2016), 1285-1287

ASIAN JOURNAL OF CHEMISTRY

http://dx.doi.org/10.14233/ajchem.2016.19655

Effect of Chemical Formulations for Uranium Decontamination by Chemical Gels

CHONG-HUN JUNG*, WANG-KYU CHOI and JEI-KWON MOON

Decontamination & Decommissioning Research Division, Korea Atomic Energy Research Institute (KAERI), P.O. Box 105, Taejon 305-600, Republic of Korea

*Corresponding author: Fax: +82 42 8684797; E-mail: nchjung@kaeri.re.kr

Received: 7 October 2015; *Accepted*: 9 January 2016; *Published online*: 29 February 2016; AJC-17792

The decontamination and rheological behaviours of chemical gels prepared in a Ce(IV)-HNO_3 or HNO_3 based solution for tile surfaces contaminated with uranium radionuclides were investigated. Gelling agents were prepared by adding PEG-based non-ionic coviscosifiers into a stable pyro Si viscosifier. Dispersion of gelling agents in a Ce(IV)-HNO_3 solution or HNO_3 solution produces a chemical decontamination gel. The order of removal percentage of uranium with various co-viscosifiers was diethylene glycol hexyl ether (DGHE) > tripropylene glycol dodecyl ether (TPGDDE) > tripropylene glycol butyl ether (TPGBE).This is considered to be due to the difference in the length of hydrophobic alkyl chains of coviscosifier and initial activity level of contaminated specimens.

Keywords: Uranium, Gel, Decontamination, Cerium.

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 gel decontamination process can be applied by spraying a gel onto any surface of the large area components to be decontaminated. 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 [4,5].

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 [6]. 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.

Gelling agents were prepared by adding PEG-based non-ionic coviscosifiers (diethylene glycol hexyl ether, tripropylene glycol butyl ether and tripropylene glycol dodecyl ether) into a stable pyro Si viscosifier. The decontamination and rheological behaviours of chemical gels prepared in a Ce(IV)-HNO_3 or HNO_3 based solution for tile surfaces contaminated with uranium radionuclides were investigated.

EXPERIMENTAL

A Ce(IV)-HNO_3 chemical decontamination agent was prepared by dissolving 0.5 M Ce(IV) in 2 M 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 Fig. 1, SEM images for the Pyro Si exhibited a network structure, which consists of the primary particles with a size of 30-50 nm 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 viscosifier and three 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).

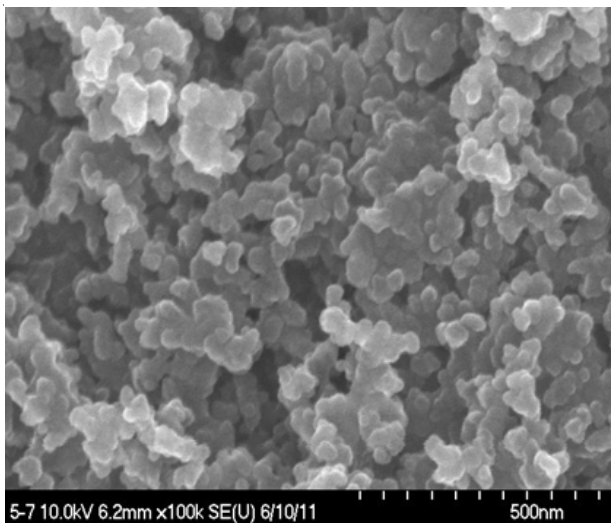
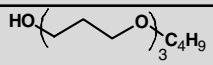
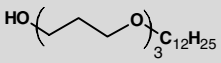
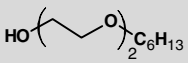


Fig. 1. SEM images of pyro Si, M-5

TABLE-1
PHYSICAL PROPERTIES OF
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)			11.0
TPGBE (Tripropylene glycol butyl ether)			15.4
TPGDDE (Tripopylene glycol dodecyl ether)			10.6

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.

RESULTS AND DISCUSSION

Fig. 2 provides a schematic rheogram of various chemical gels, which shows the effects of the formulation on the rheological properties. After shearing for 60 s 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 rebuild time of below 5 s for all chemical gels was attained.

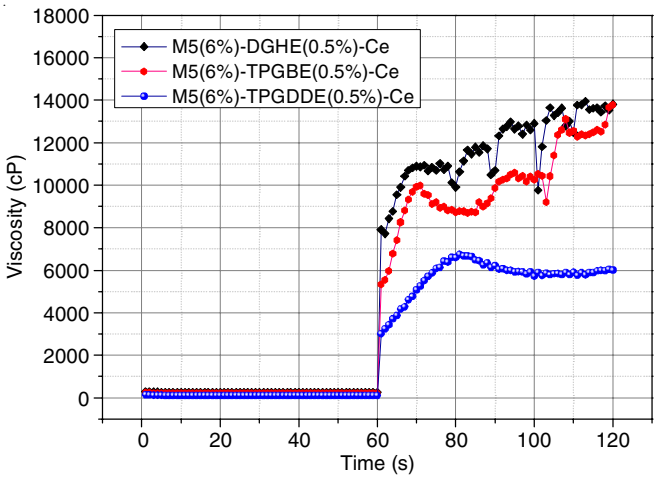


Fig. 2. Rheogram of various chemical gels: shearing for 60 s at a shear rate of 500 s^{-1} followed by shearing for 60 s at 5 s^{-1}

As shown in Fig. 3, the removal percentage of uranium with the number of decontaminations for a Ce(IV)-HNO_3 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: 24.5 Bq/g, TPGDDE:59.0 Bq/g, TPGBE:33.6 Bq/g).

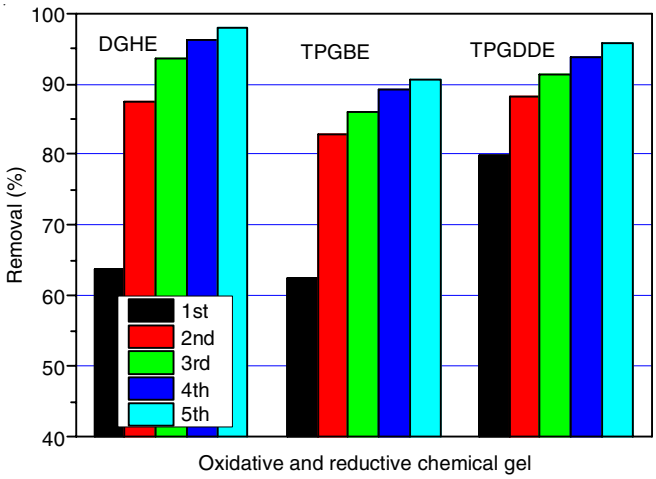


Fig. 3. Removal percentage of uranium with the number of decontaminations for a Ce(IV)-HNO_3 solution containing various coviscosifiers

Fig. 4 shows the removal of uranium with the number of decontaminations for a Ce(IV)-HNO_3 and HNO_3 solution containing a TPGDDE coviscosifier. A chemical gel containing a HNO_3 solution, compared with Ce(IV)-HNO_3 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.

Fig. 5 shows the removal concentration of the radioactivity with the applying decontamination times. With the number of times of the decontamination, the radioactivity concentration is decreased and the decontamination efficiency was increased. It accounted for about 90 % of total radioactivity removed from first and the second decontamination, indicating that

Fig. 6. Optical images of the surfaces (a) before drying, (b) after drying and (c) after detachment of decontamination chemical gel

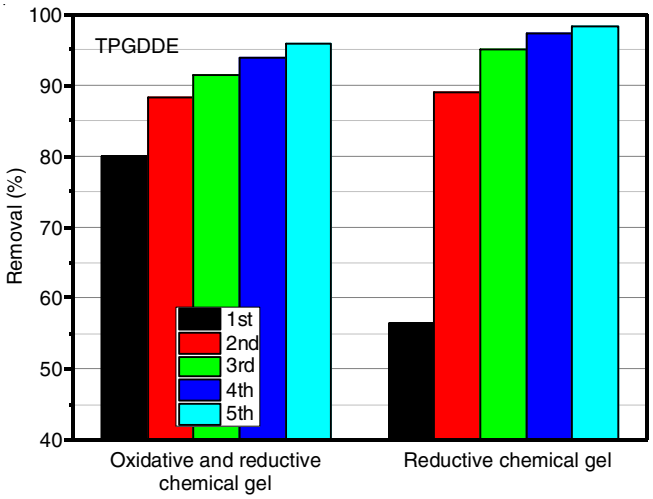


Fig. 4. Removal percentage of uranium with the number of decontaminations for Ce(IV)-HNO_3 and HNO_3 solution containing TPGDDE coviscosifier

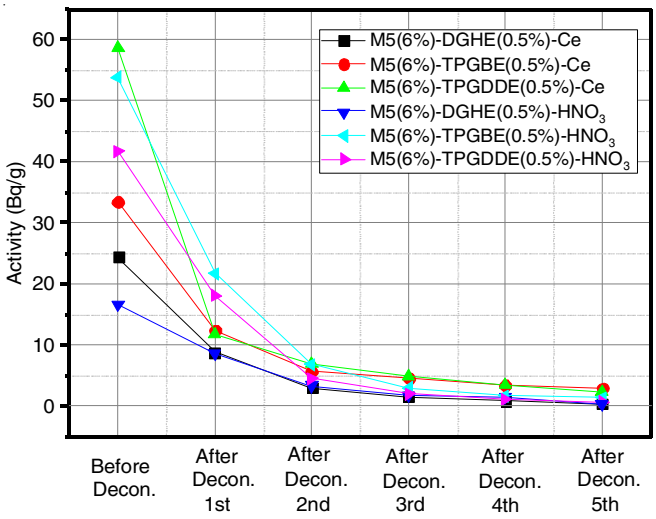


Fig. 5. Removal Concentration of radioactivity with the number of times of decontamination

the initial decontamination steps can be a significant fraction of the total activity removed.

Fig. 6 showed that the optical images of the surfaces after drying and detachment of the decontamination chemical gel reveals that almost of radioactivity was effectively removed.

Conclusion

The decontamination and rheological behaviours of chemical gels for tile surfaces contaminated with uranium radionuclides were investigated. A chemical decontamination gel composed of a pyro Si viscosifier and PEG-based non-ionic coviscosifiers in a Ce(IV)-HNO_3 solution or HNO_3 solution was created. Removal percentage of uranium with various coviscosifiers is high according to the following order: DGHE > TPGDDE > TPGBE. It is due to the difference in hydrophobic alkyl chain length of coviscosifier and initial activity level of uranium specimens.

The HNO_3 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.

ACKNOWLEDGEMENTS

This work has been carried out under the Nuclear R&D Program (2012M2A8A5025655) funded by Ministry of Science, ICT & Future planning.

REFERENCES

1. S. Faure, Innovative Processes for Nuclear Decontamination Solids, Exchange Meeting Between KAERI-CEA (2008).
2. A. Purohit, M.D. Kaminski and L. Nunez, Method for the Decontamination of Metallic Surfaces, US Patent US6504077 B1 (2003).
3. J.-P. Gauchon, J.-L. Alcaraz and L. Gallo, Gel Decontamination et Son Utilisation pour la Decontamination Radioactive de Surfaces FR-A-2656949 (1990).
4. W. Gaul, An Improved Polymer-Based Hydrogel for Decontamination of Hard Assets, Waste Management Conference, March 01 (2007).
5. G. Edgington, Decontamination of Nuclear Medicine Isotopes from Hard Surfaces using a Peelable Polymer-Based Hydrogel, HPS Annual Meeting, July 12 (2007).
6. A.L. Taboas, A.A. Moghissi and T.S. LaGuardia, The Decommissioning Handbook, ASME, pp. 4-22 (2004).