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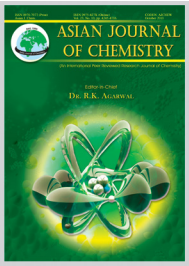

Silica-Based Chemical Gel for Decontamination of Radionuclides†

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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. A chemical decontamination gel was prepared by adding gelling agents composed of a pyro Si viscosifier and PEG-based non-ionic coviscosifier (tripropylene glycol butyl ether and tripropylene glycol dodecyl ether) into a Ce(IV) solution stabilized in concentrated nitric acid. The decontamination and rheological behaviours, along with the drying behaviours of a chemical gel for SUS 304 metallic surfaces contaminated with Co and Cs radionuclides were investigated. A chemical gel containing a 0.5 wt % tripropylene glycol dodecyl ether coviscosifier was more effective in terms of the rheological and drying-detachment properties and the radionucleide decontamination effectiveness in particular, compared to tripropylene glycol butyl ether.

Key Words: Decontamination, Silica, Gel, Radionuclides.

INTRODUCTION

Chemical decontamination technology represents a highly effective removal of radioactive contamination through chemical dissolution or a redox reaction. However, the generation of large amounts of waste limits its use as an *in situ* technology. Therefore, to avoid the well-known disadvantages of chemical decontamination techniques while retaining their high decontamination efficiency, it is necessary to develop processes using chemical gels instead of chemical solutions[1-3]. This method is effective in situations where long contact times are required and when the need to minimize waste exists. A chemical decontamination gel can be prepared by adding gelling agents composed of a viscosifier and coviscosifier to chemical decontamination agents used in traditional decontamination processes[4-7].

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. 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.

This work investigates the decontamination behaviours including rheological and drying behaviours of an inorganic-based chemical gel for SS 304 metallic surfaces contaminated with Co and Cs radioactive materials.

EXPERIMENTAL

Chemical decontamination agents were prepared by dissolving 0.5M Ce(IV) in concentrated nitric acid. The gelling agents were composed of a viscosifier and coviscosifier. Pyro Si, which is stable in an acidic medium and is easily gellated at small amounts compared to aluminium, was selected as a viscosifier and used in a 5-10 wt. % concentration during the experiment. As a coviscisifier, 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. %.

A chemical decontamination gel was sprayed onto the surface of an SUS 304 specimen contaminated with Co-60 and Cs-137 radionucleides. The gel adheres to the surface of the specimen and operates by dissolving the 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 sample, expressed by the decontamination factor (DF) were calculated by measuring the radioactivity concentration of Co and Cs radionuclides using MCA (Canberra, 2025).

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Thixotropy, defined as a decrease in 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

The shear rates before and after gel injection were simulated at 500/s and 1/s and the rheological properties were measured in accordance with the concentration of the viscosifier, as shown in Fig. 1. The viscosity increased with an increase in concentration, regardless of the type of viscosifier used.

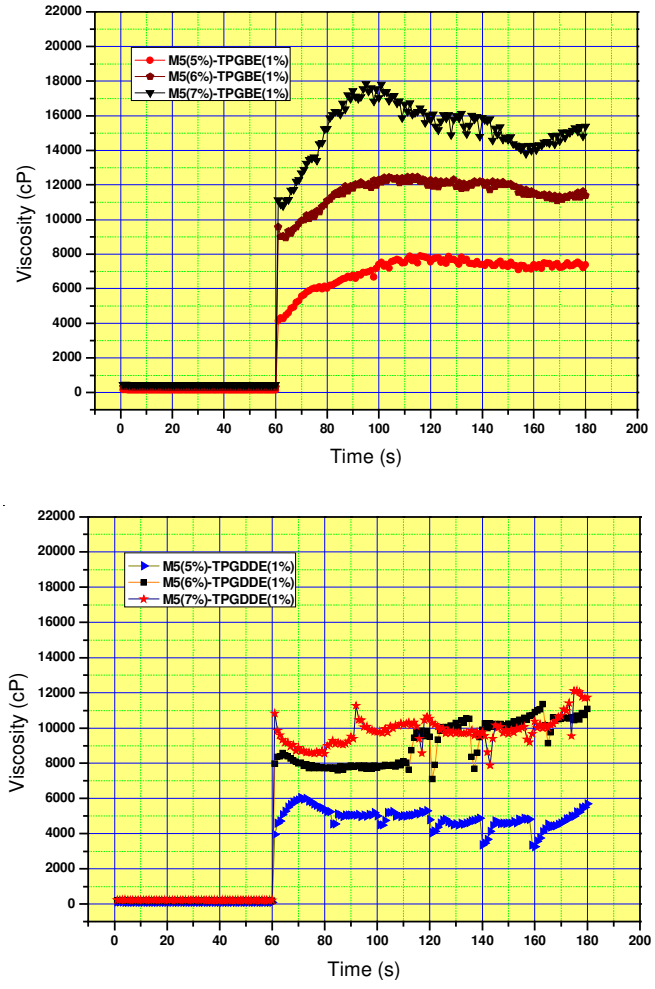

Fig. 1. Schematic rheograms of various gels

A low viscosity of less than 500 cP at a high shear rate of 500/s results in effective spraying to the surface, while a high viscosity of 7,000 cP (TPGBE), 4,000 cP (TPGDDE) or more showed the proper surface adhesion characteristics at low shear rates of 1/s. Thixotropy, which is one of the main characteristics in chemical gels, is characterized by a rebuild time when the gel structure evolves from a breakdown into a recovery state. As shown in Fig. 1, a rebuild time of less than around 3 s was achieved for the various gels tested and the effective thixotropic behaviour was observed.

It is known that the addition of a coviscosifier makes it possible to augment the viscosity recovery of the gel and also allows a better control of the cracking phenomena to increase the homogeneity of the size of the solid residues. The drying characteristics of the gel containing radioactive nuclides after decontamination were investigated using various concentrations of viscosifier and coviscosifier and are shown in Fig. 2. Gel containing radionuclides after decontamination were completely dried within 6 h, regardless of the viscosifier and coviscosifier concentration.

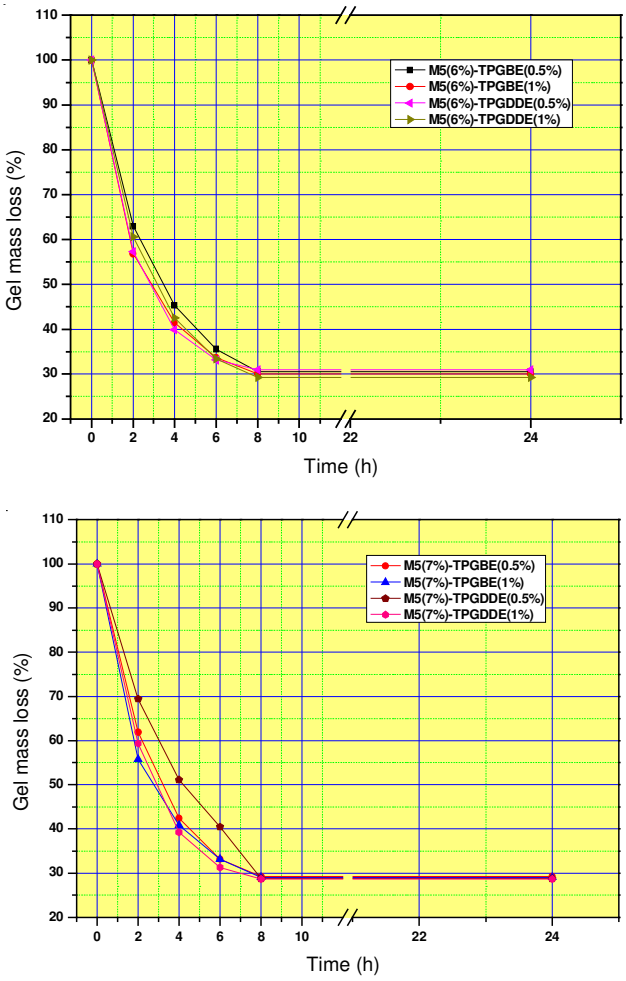

Fig. 2. Drying characteristice of various gels

The detachment characteristics of the gels after drying were also investigated with various gel formulations. As shown in Fig. 3, the percentage of detachability of the used gel increased in proportion to the concentrations of viscosifier and coviscosifier, both gels containing TPGBE and TPGDDE. The percentage of detachability of the gel containing TPGDDE was more effective than that of TPGBE. These results seem to be a relationship with the crack generated on the surface of SUS 304.

Fig. 4 shows the surface of the SUS 304 specimens before and after detachment, revealing that the gel products have been effectively removed from the surface.

The addition of a coviscosifier unexpectedly makes it possible to increase the ability of the solid gel residues obtained after drying to be detached from the support. However, the

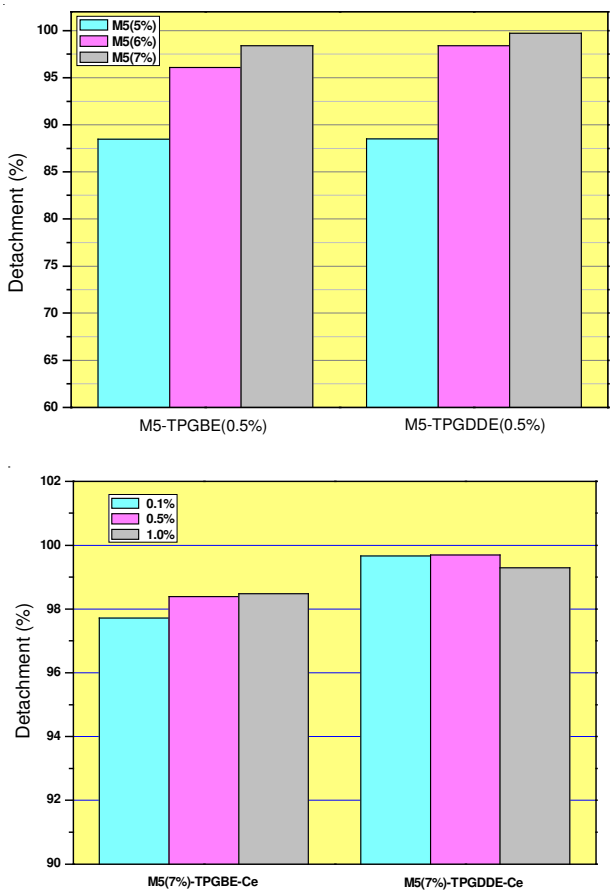

Fig. 3. Detachability of various gels

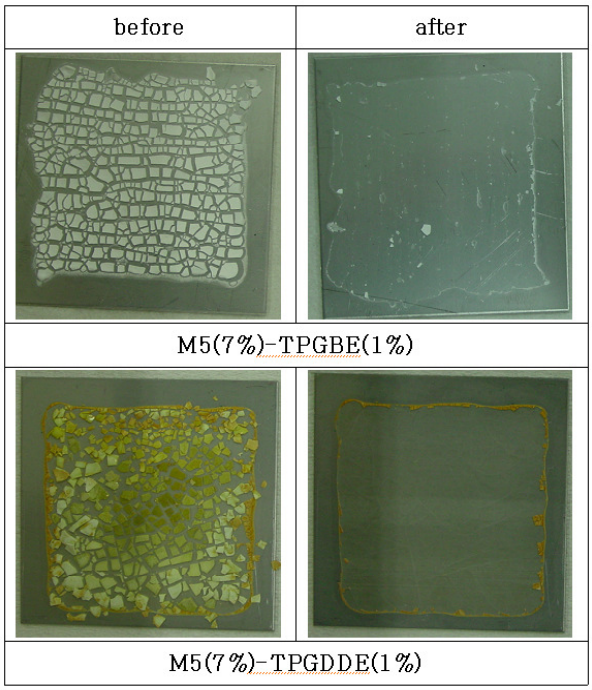

Fig. 4. Photographs of SUS 304 surface before and after detachment operation

surface of SUS 304 obtained from detachment of the gel containing TPGDDE was clearer than that of TPGBE. This seems to be due to the formation of uniform (2.5-3.0 mm) and larger sized cracks on the surface applied with gel containing TPGDDE.

The decontamination factor for Co and Cs (Fig. 5) shows that decontamination factor increased for both radionuclides depending on the viscosifier concentration, with a high decontamination effect within a decontamination factor range of 100-800.

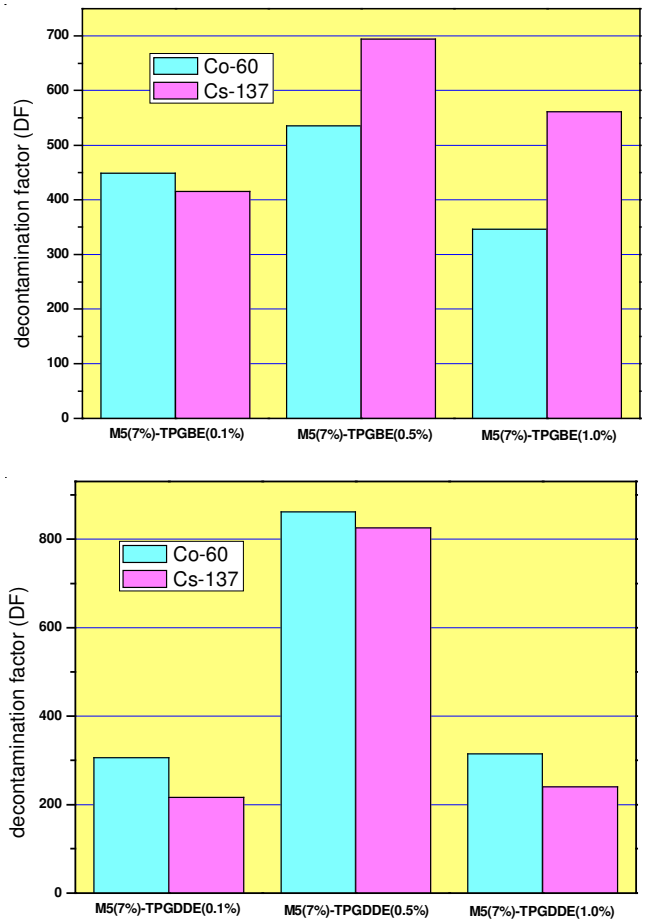

Fig. 5. Decontamination factor with variation in gel formulation

Using a gel formulation containing TPGBE as a coviscosifier, the decontamination factor for Co and Cs radionuclides were in the range of 200-520 and 180-700, respectively. The decontamination factor for Co and Cs radionuclides using a TPGDDE or TPGBE coviscosifier increased depending on the concentration of the viscosifier. In particular the decontamination factor increased greatly at a viscosifier concentration of 6 and 7 wt. % and ranged from 810-820 for Co and from 700-810 for Cs, respectively.

Conclusion

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 and the reduction of the amount of final solid waste. A pyro Si-based chemical decontamination gel was prepared by adding gelling agents composed of a pyro Si viscosifier and PEG-based non-ionic coviscosifier (TPGBE and TPGDDE) into a Ce(IV) solution in concentrated nitric acid and the decontamination and rheological behaviours, along with the drying behaviours of a chemical gel for SUS 304 metallic surfaces contaminated with Co and Cs radionuclides were

investigated. A chemical gel containing a 0.5 wt % TPGDDE coviscosifier was more effective in terms of the rheological and drying-detachment properties, as well as the radionuclide decon-tamination effectiveness in particular, compared to TPGBE.

ACKNOWLEDGEMENTS

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