

Chemical Gel for Decontamination of Cs Surrogate on Stainless Steel Surface - 11257

Chong Hun Jung, Jei Kwon Moon, Hui Jun Won, Kune Woo Lee and Chang Ki Kim
Korea Atomic Energy Research Institute
P.O.BOX 105, Yuseong-gu, Daejeon, 305-600
Korea

ABSTRACT

This work investigates the decontamination behaviors of an organic-based chemical gel for SS 304 metallic surfaces contaminated with cesium surrogate materials. Chemical gels can be formulated by adding gelling agents composed of a polymeric viscosifier and surfactant to chemicals used for traditional chemical decontamination processes. Polyvinyl alcohol (PVA) was selected as the viscosifier. Diethylenetriaminepentaacetic acid (DTPA) was chosen as a decontaminating chemical based on its chelating efficiency. Anionic surfactant (sodium dodecyl sulfate, SDS) with a concentration of 1.5 wt% - 3.0 wt% and DTPA were added to the PVA mixture. The removal % of Cs was investigated with variation in PVA and surfactant concentration at a Cs concentrations of 0.01 g/cm² and 0.0025 g/cm².

More than 99 % removal was obtained under the conditions of PVA > 12.5 wt.% and SDS > 2 wt.%. Increasing the viscosity of about 3,500 cP and an achievement in the rebuild time of less than around 4 seconds, demonstrated a relatively effective thixotropic behavior. Approximately 70 % of the amount of initial chemical gel remained after adhesion under the conditions of 15 % PVA, 2 % SDS, and 2% DTPA. The chemical gel used was effectively dried within 24 hours and was able to be detached as a sheet of film.

The appropriate combination of viscosifier and surfactant is a very important factor in the control of viscosity and adhesion properties of chemical gels.

INTRODUCTION

Many chemical decontamination processes operate by immersing components in aggressive chemical solutions. In these applications, chemical decontamination techniques produce large amounts of radioactive liquid waste [1-2]. Therefore, to avoid the well-known disadvantages of chemical decontamination techniques while retaining their high efficiency, it is necessary to develop processes using chemical gels instead of chemical solutions [3]. Chemical gels are used as carriers of chemical decontamination agents. This method is effective in situations where long contact times are required, together with the need to minimize waste.

A chemical gel decontamination process consists of applying a gel by spraying it onto the surface of large area components (floors, walls, etc) to be decontaminated, as shown in Figure 1. The gel adheres to any vertical or complex 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.

Only small quantities of gel are required, and the gels demonstrate thixotropic properties i.e., they are liquid when sprayed, and solid when stationary, allowing for strong adherence to surfaces.

This work investigates the decontamination behaviors of an organic-based chemical gel for SS 304 metallic surfaces contaminated with radioactive materials.

EXPERIMENTAL

Gel preparation

Chemical gels can be formulated by adding gelling agents, composed of a polymeric viscosifier and surfactant, to chemicals used for traditional decontamination processes. Polyvinyl alcohol (PVA) was selected as a viscosifier that can form synergistic emulsifying combinations with a surfactant. To obtain the tensile strength for peelability, PVA with a molecular weight higher than 20000g/mol was purchased from Sigma. Sodium dodecyl sulfate (SDS) was used as an anionic surfactant in order to further lower the surface tension for easy application, and to stabilize the chelating agent used as a decontamination chemical in the gel for a longer life. Among the most frequently used chelating agents, diethylene-triaminepentaacetic acid (DTPA) was chosen based on its chelating efficiency and non-harmful environmental effects. 12.5 % to 17.5 % of PVA relative to the total weight of the gel was mixed with demineralized water and heated in a silicone oil bath to about 90°C until the PVA was fully dissolved. The mixture was then cooled to room temperature. A surfactant with a concentration of 1.5 wt% to 3.0 wt% and DTPA(1.0 wt% - 2.0 wt%) were added to the mixture. The gel was then neutralized with a 10 wt% KOH solution to around a pH of 7.0.

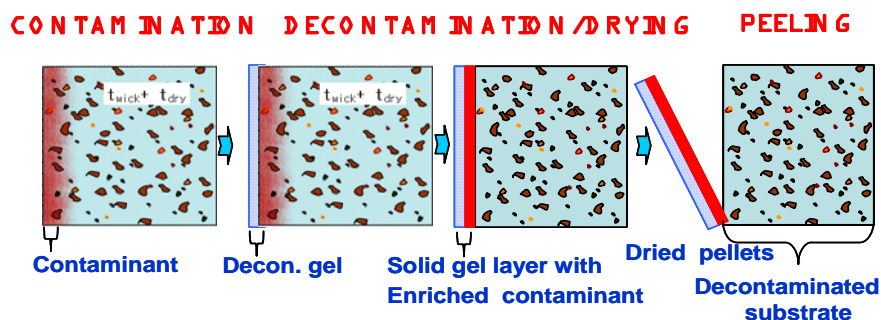


Fig. 1. Surface decontamination using chemical gel.

Surface decontamination and rheology

To examine the decontamination properties of the chemical gel, a non-radioactive surrogate compound, cesium chloride, was chosen as the surrogate of radionuclide Cs137. An aqueous stock solution prepared at a concentration of 10wt% was uniformly distributed onto the SS 304 plate with a dimension of 7cm X 7cm. After the solution had evaporated, a chemical gel containing DTPA was applied to the SS plate. After 3hrs, the dehydrated gel was peeled off. The SS plate was rinsed with distilled water to dissolve any residual salt. The rinsed solution was dried in an oven at 80C for 48hrs. The removal efficiency was determined by weighing the residual salt.

Thixotropy is a rheological phenomenon defined as a decrease in apparent viscosity under stress, followed by gradual recovery at rest. The rheogram curves at various shear rates according to the gel formulations were obtained using a viscometer (Brookfield Eng. & Lab. Inc., RVDV-□U) and Rheocalc 32 software.

RESULTS AND DISCUSSION

Rheology

The main characteristic in chemical gels lies in their thixotropic properties. Thixotropy is characterized by

a re-build time when the gel structure evolves from breakdown to recovery states.

Figure 2 shows schematic rheograms of various PVA-based gels and represents the effects of the formulation on the rheological properties. After shearing for 120 s at a shear rate of 15 s^{-1} , a consecutive shear rate of 1.7 s^{-1} representing a stationary states, was imposed for 120 s. It was reported that the equilibrium viscosity has an effect on the amount of viscosifier, chelating agent concentration, type and concentration of the surfactant, and the salinity and electrolyte properties of the medium [4].

The equilibrium viscosity at 1.7 s^{-1} ranged from 8000 to 9500 cP depending on the gel formulations. A rebuild time of about 4 seconds was achieved for various gels tested. When a surfactant and DTPA chemicals are added, the viscosity and rebuild time attain satisfactory values. However, the viscosity at a shear rate of 15 s^{-1} ranged from 5000 to 6000 cP, and was too high for spraying.

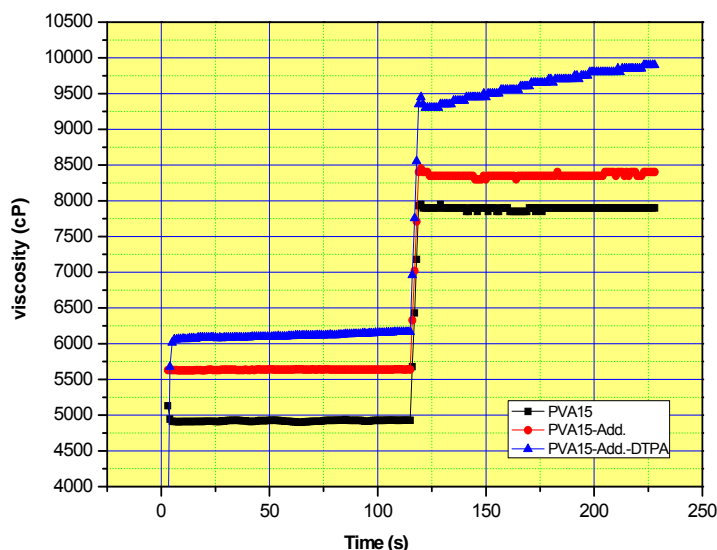


Fig. 2. Schematic rheograms of various chemical gels.

Adhesion

To test the adhesive force of a gel to a metal surface by viscosity control, the residual amount of chemical gel with the elapsed time was measured by keeping the metal sample perpendicular after coating the metal surface.

The residual amounts of chemical gel with variations in PVA concentration are shown in Figure 3 for a fixed concentration of 2 % surfactant and 2 % DTPA. The residual amount of gel increased with an increase of PVA concentration. 100% of the initial coated gel remained for 3 hours at a PVA concentration of 17.5 %, but 67 % and 34 % remained at PVA concentrations of 15 % and 12.5 %, respectively. The equilibrium residual time was 30 - 40 minutes.

At a fixed concentration of 15 % PVA and 2 % DTPA, the residual amounts of chemical gel with variations in surfactant concentration are shown in Figure 4. The residual amounts of a gel were 67 % and 84 % at surfactant concentrations of 2 % and 3 %, respectively.

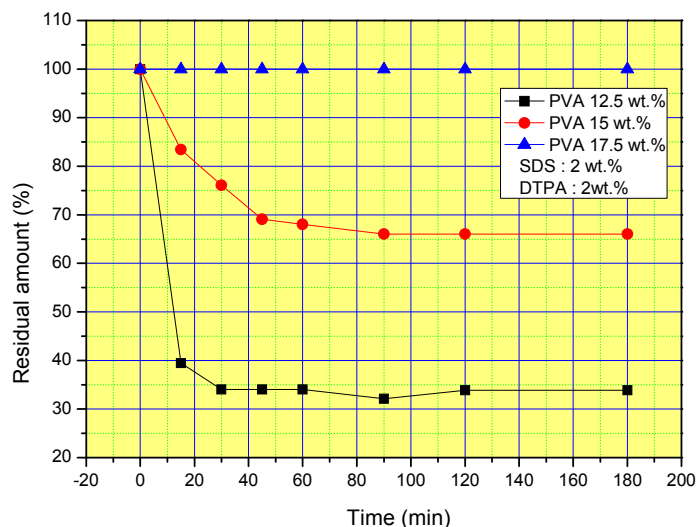


Fig. 3. Residual amounts of chemical gel with variations in PVA concentration.

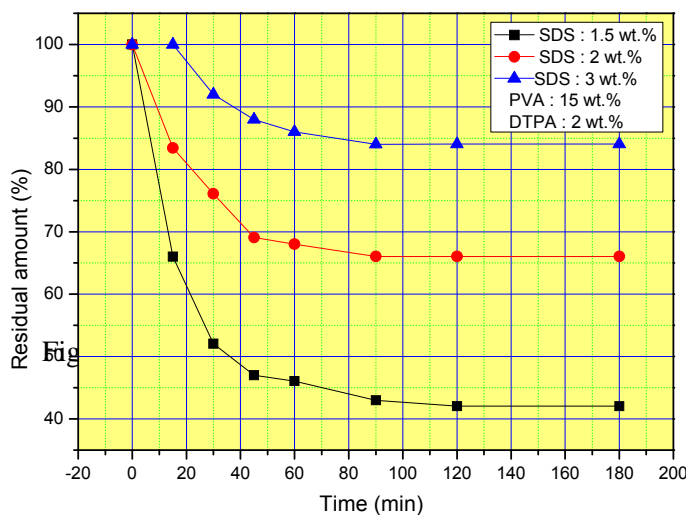


Fig. 4. Residual amounts of chemical gel with variations in surfactant concentration.

Cs removal

Figure 5 shows the removal of Cs with variation in CsCl & PVA concentration at a constant surfactant concentration of 2 %. The removal % of Cs was about 99.5 % and 99.0 % based on the variations in PVA concentration (12.5 - 17.5 %) at Cs salt concentrations of 0.01 g/cm² and 0.0025 g/cm², respectively. The chelating gel containing DTPA was effective in removing cesium.

At a PVA concentration of 15 %, Cs removal was examined by varying the surfactant concentrations of two samples with different contamination levels, the result of which are shown in Figure 6. The removal efficiency of Cs increased depending on the surfactant concentration, but the increment of removal % was less than 1 %. Cs removal % also increased by increasing the Cs contamination levels. The average

removal % was 99.5 % at a Cs contamination level of 0.01 g/cm², whereas Cs removal % ranged from 98.2 to 99 % at 0.025 g/cm².

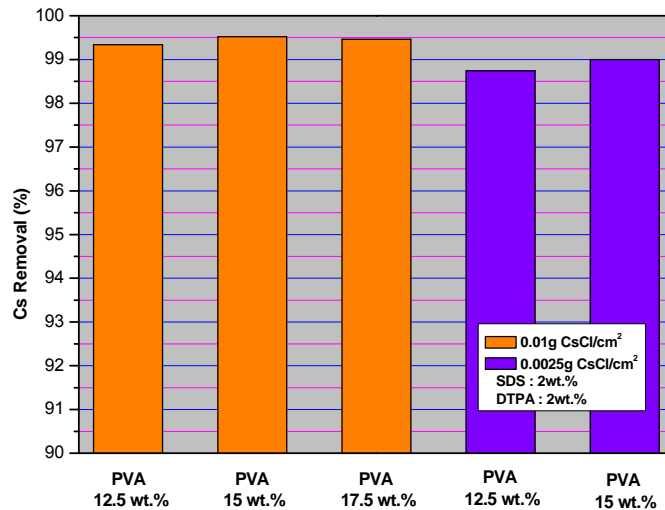


Fig. 5. Cesium removal % with variations in PVA concentration.

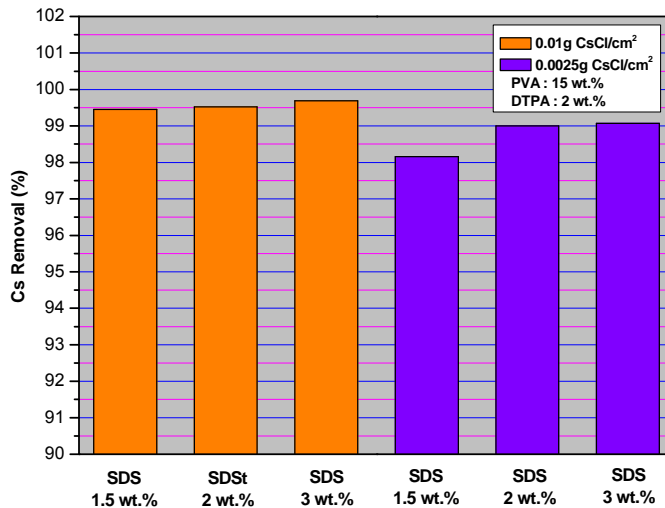


Fig. 6. Cesium removal % with variations in surfactant concentration.

Figure 7 shows Cs removal % using two types of polished and non-polished SUS 304 samples. Cs removal % on the surface of the polished SUS 304 sample was higher than that of the non-polished sample by 0.5 to 2.5 %. This is considered to be due to the difference of surface contact angle between the two samples, since the contact angle on polished SUS 304 (54 to 59 degrees) was lower than that of the non-polished SUS 304 (59 to 62 degrees). Several kinds of variables in the contact angles exist, but in general, the wettability and adhesiveness improve as the contact angle is reduced.

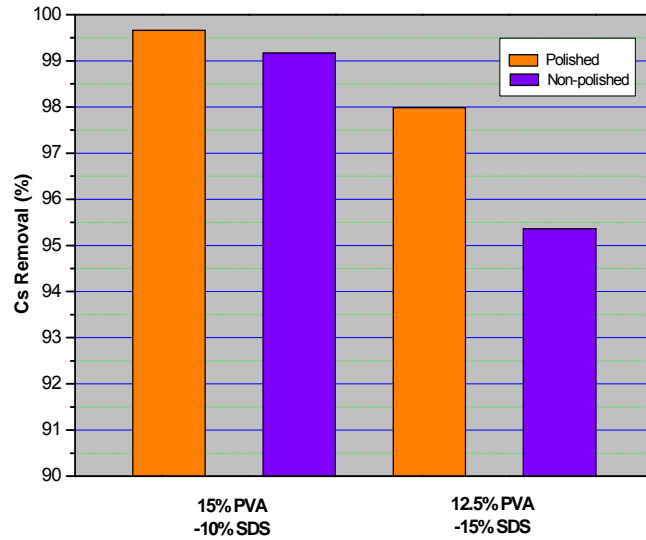


Fig. 7. Cesium removal % for polished and non-polished SUS 304 plates.

Drying and detachability

A cesium solution was uniformly distributed onto the surface of SUS 304 plate. After the solution was evaporated at ambient temperature, the organic decontaminating gel was applied to the SUS 304 plate and the coated plate remained in a fume hood for effective evaporation. After 3hrs, the gel was completely dried and could be peeled off as a single continuous sheet as shown in Fig. 8.

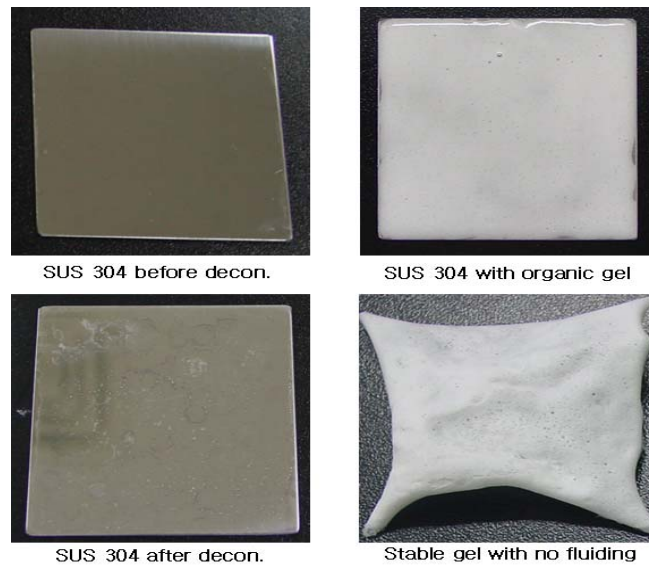


Fig. 8. Photographs of the removal of cesium using a chemical gel.

CONCLUSIONS

The decontamination behaviors of an organic-based chemical gel for SS 304 metallic surfaces contaminated with cesium surrogate materials was investigated. Chemical gels can be formulated by adding gelling agents composed of a polymeric viscosifier (PVA) and surfactant (SDS) to chemical (DTPA) used for traditional chemical decontamination processes.

The appropriate combination of viscosifier and surfactant is a very important factor in the control of viscosity and adhesion properties of chemical gels.

More than 99 % removal was obtained under the conditions of PVA > 12.5 wt.% and SDS > 2 wt.%. Increasing the viscosity of about 3,500 cP and an achievement in the rebuild time of less than around 4 seconds, demonstrated a relatively effective thixotropic behavior. With an increasing concentration of viscosifier, surfactant, and DTPA, the remaining portion of chemical gel was increased. Approximately 70 % of the amount of initial chemical gel remained after adhesion under the conditions of 15 % PVA, 2 % SDS, and 2% DTPA. The chemical gel used was effectively dried within 24 hours and was able to detached as a sheet of film. Cs removal % on the polished SS 304 sample was higher than that of the non-polished sample by 0.5 to 2.5 %, owing to the difference of surface contact angle between the two samples

ACKNOWLEDGMENT

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