SURFACE DECONTAMINATION STUDIES OF $^{137}$Cs AND $^{85}$Sr USING POLYMER GEL

ID number "23rdWiN/5"

Pham Quynh Luong$^1$, Nguyen Van Chinh$^1$, Nguyen Dinh Lam$^2$

$^1$Institute for Technology of Radioactive and Rare Elements (ITRRE)
$^2$Nuclear Research Institute (NRI)
Vietnam Atomic Energy Institute (VINATOM)
Email: phamquynhluong@yahoo.com

I. ABSTRACT

Strippable polymer coating is one of the methods for effective surface decontamination. A gel solution of polyvinyl alcohol (PVA) and chelating agents were applied to remove radioisotopes of $^{137}$Cs and $^{85}$Sr from surface of stainless steel, mild steel, ceramic, PVC plastic. After cleaning was completed, the gel solution was dried (24 h), formed a strong thin film and it was easily peeled off from a contaminated surface with the radioactive isotopes and can be disposed off as radioactive solid waste. The infrared spectrophotometer has been conducted to study the interaction between PVA, polymer gel with cesium, strontium ions.

Key word: strippable coating. Polymer coating, decontamination by polymer

II. INTRODUCTION

Radioisotopes are widely used in industry, agriculture, scientific researches and education, especially in nuclear industry... While handling and storing radioisotopes, surface of machines, tools, laboratory desks, floors... may get contaminated with radioactive materials. If contaminated surfaces haven't decontaminated in time, the radioactive materials can transfer to the environment and staffs working around. Radioisotopes may be absorbed by the human body and caused internal doses. Decontamination is a process of remove radioisotopes from contaminated surfaces. Normally, diluted acids or detergents could be used for surface decontamination, however, these techniques have a disadvantage: it'll release large amount of secondary liquid radioactive waste, which needs to treat before storage and final disposed.

Using strippable polymer coating is thought to be an effective and simple technique to remove radioisotopes from contaminated surfaces. The secondary wastes from decontamination process exist in solid form, small volumes. It's also compressible and combustible. This enables the management of radioactive waste to be more effective and economy.
III. EXPERIMENTS

1. Polymer coating preparation.

Dissolve 140 g polyvinyl alcohol (14% wt) in 775 g distilled water
stir well for 30 min at $T^0 = 50 – 60^\circ$C

Add 75 g glycerine (7.5% wt) glycerine, 5 g citric acid in
5 ml distilled water (0.5 %wt)
stir well for 30 min at $T^0 = 50 – 60^\circ$C

Stay gel solution for overnight at the room temperature
without stirring for defoaming

Transfer this gel mixture is in to a sealed clean
polyethylene bottle for storage
(Solution sticky, gel-form and transparent)

Fig. 1. Polymer coating preparation.

2. Influence of various agents to decontamination factor

The aqueous PVA/H$_2$O blend was prepared as described. Some decontamination agents were studied: EDTA, DTPA, oxalic acid, citric acid and PVA. 1 ml solution of radioisotope Cs$^{137}$ and Sr$^{85}$ (activity 0.0377 mCi/ml and 0.0145 mCi/ml respectively) is spread on plates of glass, Activity on the surfaces is measured by MED-CoMo170 survey meter before and after decontamination. Decontamination efficiency of polymer coating is represented by K (%) or decontamination factor DF and calculated by following formula:

$$K(\%) = \frac{[A_i - A_f]}{A_i} \times 100 \quad (1)$$

$$DF = \frac{A_i}{A_f} \quad (2)$$

Where: $A_i$ is the surface activity before decontamination
$A_f$ is the surface activity after decontamination

3. Influence of polymer coating thickness to DF

About 1 ml $^{137}$Cs and $^{85}$Sr solutions is spread on a 10 cm x 10 cm of stainless steel plates, then dry out and cover contaminated area with various amount of gel 0.7 g; 1.0 g; 2.0 g; 3.0 g, equivalent to various thickness: 0.09 mm; 0.11 mm; 0.22 mm; 0.34 mm respectively. Coating thickness is measured by spline micrometer. Activity of the contaminated area were measured by MED-CoMo170.
4. Influence of activities to decontamination factor

On the surface glasses apply amount of different $^{137}\text{Cs}$ and $^{85}\text{Sr}$ solutions. The polymer coating is peeled off after drying. Radioactivity of each glass was measured before and after decontamination by MED-CoMo170. Decontamination factor related to each activity is calculated by formula (2)

5. DF of Polymer gel and Decongel 1101 on various surfaces

On the surface of glass, stainless steel, mild steel, ceramic, PVC plastic plates, apply 1.0 ml $^{137}\text{Cs}$ and $^{85}\text{Sr}$ solutions, dry out and cover contaminated area by 1.5 g Polymer gel and Decongel 1101 (supply by CBI Polymer US) separately. Activity of each plate was measured by MED-CoMo170. DF related to polymer gel and Decongel 1101 were calculated by formula (2)

6. Interaction of polyvinyl alcohol (PVA) and Polymer gel (Gel) with cesium and strontium ion

In order to study the interaction between the PVA, polymer gel with cesium, strontium ions, the infrared spectra of PVA, PVA-Cs, PVA-Sr, Gel, Gel-Cs and Gel-Sr were obtained apply 0.5 ml SrCl$_2$ and CsCl 1.0 mg/ml solution on 50 mm diameter pieces of smooth stainless steel. Dried out and covered it by PVA gel, polymer gel separately. The coatings were peeleed off after drying, they were analyzed by FTIR (pellet compressed with KBr in IMPACT-410 compressor, Nicolet-Carl Zeiss Jena – Germany)

IV. RESULTS AND DISCUSSION

1. Influence of various agents to decontamination factor

The DF of polymer gel with all the agents tested for $^{85}\text{Sr}$ was higher than that for $^{137}\text{Cs}$. It is possible that Sr ion interact with polymer gel better than Cs ions does. It was also revealed that citric acid was the most effective decontamination agent of this particular coating for $^{137}\text{Cs}$ and $^{85}\text{Sr}$, because in this case, citric acid not only acts as acid but also as a good chelating agent

![Decontamination factor DF of potential decontamination agents](image)

Fig. 2. Decontaminating factor DF of potential decontamination agents

2. Influence of polymer coating thickness to DF

In the Fig. 3 showed that DF get the highest value at coating thickness from 0.11 mm to 0.22 mm (equivalent to 1 – 2 g/20 cm$^2$). At thickness of 0.09 mm, there's too
little of gel, so, coating film can't absorb all radioisotopes. At thickness of 0.34 mm, the coating can't dry out completely in within 24 hours under the room temperature and leave a small amount gel with radioisotopes on the surface. As a result, the DF factor is decreased. The optimal polymer gel is about 0.8 – 1 ml/m² surface, equivalent to 0.15 mm thickness of coating film. With this amount, decontamination efficiency can archive 97% for $^{137}$Cs and 99% for $^{85}$Sr.

Fig. 3. Decontaminating factor DF of various coating thickness

3. Influence of activities to decontamination factor

Experiments showed that in the range of studied activities, the higher activity of $^{137}$Cs and $^{85}$Sr, the lower DF we can obtained, because at high activity, the density of Cs and Sr ion is very high and they will compete with each other in interaction with decontaminating agent or they possible need more agents for this removal process completely. The DF of $^{137}$Cs and $^{85}$Sr the glass plate are demonstrated in Fig. 4

Fig 4. DF of polymer gel for various activities of $^{137}$Cs and $^{85}$Sr

4. DF of polymer gel and Decongel1101 on various surfaces

DF of polymer gel for $^{137}$Cs and $^{85}$Sr has great difference on various surfaces, for both of $^{137}$Cs and $^{85}$Sr, the DF decreased in the order: glass > stainless steel>ceramic>plastic>mild steel. Results in table1 showed that DF great depends on porosity, smoothness of each surface. After decontamination, all activity of surfaces ($A_d$) are below 3.7 Bq/cm², satisfy the regulations of IAEA for β and γ contaminated surface. Results of experiment also showed in Fig. 5 that DF of Decongel 1101is higher than polymer gel on most of surfaces for both $^{137}$Cs and $^{85}$Sr but the difference isn't noticeable.
Table 1. DF of polymer gel for $^{137}$Cs and $^{85}$Sr on different surfaces.

| Surface       | Radioisotope | $A_i$ (Bq/cm$^2$) | $A_f$ (Bq/cm$^2$) | DF  | K (%) |
|---------------|--------------|------------------|------------------|-----|-------|
| Glass         | Cs-137       | 22.6             | 0.07             | 315 | 99.7  |
|               | Sr-85        | 155.4            | 0.21             | 723.7 | 99.9 |
| Stainless steel | Cs-137     | 24.2             | 0.64             | 38.1 | 97.4  |
|               | Sr-85        | 70.3             | 0.36             | 196.6 | 99.5 |
| Ceramic       | Cs-137       | 24.3             | 0.87             | 27.9 | 96.4  |
|               | Sr-85        | 77               | 0.78             | 98.3 | 99.0  |
| PVC plastic   | Cs-137       | 22.5             | 0.84             | 26.8 | 96.3  |
|               | Sr-85        | 77.7             | 0.99             | 78.2 | 98.7  |
| Mild steel    | Cs-137       | 27.9             | 1.31             | 21.3 | 95.3  |
|               | Sr-85        | 92.7             | 2.50             | 37.1 | 97.3  |

Fig.5. DF of Decongel 1101 and Polymer gel for $^{137}$Cs and $^{85}$Sr on various surfaces

5. Interaction of PVA, polymer gel with cesium, strontium ions

FTIR spectra of PVA, PVA-Cs, Gel, Gel-Cs are given in figure 6;7. There is a shift in wave number of (C=O) group: from 1710.11 cm$^{-1}$; 1652.00 cm$^{-1}$(PVA) to 1637.13 cm$^{-1}$(PVA-Cs) and from 1714.46; 1636.14 cm$^{-1}$(Gel) to 1695.06;1623.21 cm$^{-1}$(Gel-Cs), the carbonyl group (C=O) of polymer gel contributed mainly from citric acid. It is indicating that there is interaction between cesium ion and carbonyl group (C=O) in PVA and polymer gel

FTIR spectra of PVA- Cs, Gel-Cs and PVA- Sr Gel-Sr are given in fig. 6;7. There is a shift in wave number of (C=O) group: from 1710.1; 1652.00 cm$^{-1}$(PVA) to 1715.84; 1650.70cm$^{-1}$(PVA-Sr) and from 1714.46; 1636.14 cm$^{-1}$(Gel) to 1717.58; 1627.44 cm$^{-1}$(Gel-Sr).This indicated that there is a strong interaction between strontium ion and carbonyl group (C=O). The reason is citric acid in polymer gel. Citric acid has three carboxyl groups, capable of forming chelate complex with metals, especially with Cs, Sr ions in following equation:
\[
\begin{align*}
\text{Cs}^+ + \text{HL}^3- & \rightarrow \text{CsHL}^2- \quad \text{K} = 0.32 \quad \text{at } 25^\circ\text{C} \\
\text{Sr}^{2+} + \text{HL}^3- & \rightarrow \text{SrHL}^- \quad \text{K} = 3.05 \quad \text{at } 25^\circ\text{C}
\end{align*}
\]

It means that citric acid can form chelating complex with Sr ion better than with Cs ion, this is the same the results obtained from decontamination process. It showed that decontamination of strontium is better than that of cesium and polymer gel decontaminate strontium, cesium much better than PVA gel do.

![Fig.6. IR spectrum of PVA, PVA-Cs, PVA-Sr](image1)

![Fig.7. IR spectrum of polymer gel, gel - Cs, gel - Sr.](image2)

**IV. CONCLUSION**

The polymer coating that we have prepared meet actually the needs to high efficiency decontamination of radioisotopes $^{137}\text{Cs}$, $^{85}\text{Sr}$ up to 99 % on glass and stainless steel surfaces. In addition both of two gels can remove medical radioisotopes such as $^{131}\text{I}$, $^{32}\text{P}$, $^{99}\text{mTc}$ on the glass and stainless steel surface with decontamination efficiency (K%) reached 99 %.

Decontamination factor (DF) strongly depends on property, porosity and smoothness of the contaminated surface. The DF also depends on activity and coating thickness. Optimization of thickness is around 0.15mm. Decontamination factor of Polymer gel and Decongel 1101 on surfaces are not much different from each other. IR spectra studies indicated that Cs and Sr ions interacted with PVA and citric acid in Polymer gel through C=O group. Polymer gel could remove of Sr$^{85}$ and Cs$^{137}$ better than PVA gel does because of citric acid, which can form chelating complex with Cs and Sr ion. This decontamination technique can save costing for secondary waste processing that produce in decontamination.

We hope that this study will to develop the efficiency decontamination material that will be able apply in nuclear facilities or hospital in Viet Nam.
