Research on synthetic formula of a quick-drying type of radioactive contamination detergent

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Abstract. To synthesize a quick-drying type of radioactive contamination detergent, a solvent type of detergent based on polyurethane was designed. And the synthetic formula of the detergent was studied by theoretical calculation and experiment. Through theoretical calculation, the basic molar ratio of toluene diisocyanate, chain extender, and polyether polyol were 1.73:0.73:1. The effects of the type of chain extenders and the content of catalysts on the performance of decontamination material were studied by formula experiment. The optimized formulation of detergent was obtained and a quick-drying type of radioactive contamination detergent was synthesized by prepolymer process. The decontamination effect of the detergent on uranium dust was verified on different surfaces, and both decontamination rates exceeded 90%.

1. Introduction

In recent years, the number of nuclear power plants all over the world has increased due to the clean nature of nuclear energy [1]. However, since the application of nuclear energy, a large number of nuclear leakage accidents have occurred around the world [2, 3]. If the radioactive contamination from these accidents was not removed in time, it would pose a serious threat to the environment and human health[4]. At present, the commonly used methods of removing radioactive contamination include high-pressure water flushing method, soil shoveling and strippable coating method[5-7]. The strippable coating decontamination technology has become a research hotspot due to its advantages of no secondary pollution and high single decontamination rate[8]. Nowadays the strippable coating materials are divided into water-soluble and organic solvent according to the type of solvents. Although water-soluble materials are environment-friendly, the drying time is long. In contrast, the solvent type of detergent has a shorter drying time, which is due to the use of volatile solvents accelerates their reaction rate to achieve the purpose of timely removal of radioactive contamination.

In order to shorten the drying time and improve the timeliness of decontamination, a solvent type of detergent based on polyurethane was designed and synthesized. In this experiment, toluene diisocyanate, polyether polyol, and chain extender were used as main film-forming agents, and the basic proportion was determined by theoretical calculation. And then, the appropriate chain extender was selected and the amount of catalyst was determined by experiment. In addition, the decontamination rates of this detergent on different surfaces were tested.
2. Experiment

2.1. Raw materials and instruments
Toluene diisocyanate (TDI-80) was obtained from Hubei Xinmingtai Chemical Co. Ltd. Polyether polyol was purchased from Guangzhou Wenlong Chemical Co. Ltd. Glycerin (GLY) was received from Jinan Kanjun Chemical Co. Ltd. 4, 4′-Methylene bis(2-chloroaniline) (MOCA) was bought from Jinan Dahui Chemical Technology Co. Ltd. Xylene was provided by Jinan Jinghao Chemical Co. Ltd. The values of decontamination effect were received by PAM-170E surface contamination detector (VF Nuclear Instrument Co. Ltd, Czech Republic).

2.2. Preparation of contamination detergent
The preparation process of the contamination detergent is shown in Fig. 1. Firstly, polyether polyol and TDI were weighed at a predetermined scale. The polyether polyol was heated and dehydrated at a heating temperature of 100-110℃ for 3h. In the second step, the TDI was added in batches, and the stirring temperature was raised to 70-90℃ for 1-2h. Finally, the solution was cooled to room temperature. The component A was obtained and the viscosity was 2500 mPa-s. The component A was 10kg polyether polyol and 7.5kg TDI. In addition, the component B was prepared by dissolving 12kg polyether polyol and 4.8kg MOCA in 1L xylene for 2h. The component B was obtained and the viscosity was 2500 mPa-s. The contamination detergent was obtained by stirring the component A and component B for 2 min.

![Diagram of contamination detergent preparation](image)

**Fig. 1 The preparation process diagram of the contamination detergent**

3. Result and discussion

3.1. The theoretical calculation of main agent formula
In this study, TDI and chain extender were selected as hard segment material, polyether polyol was regarded as soft segment material. The basic proportion of main agent content was determined by theoretical calculation. The terminal isocyanate prepolymer was obtained by the reaction of TDI and polyether polyol, the reaction equation was shown in Equation 1.

\[
\begin{align*}
    n \text{ HO-R-OH} + (n+1) \text{OCN-R-NCO} & \rightarrow \text{OCN} \left[ \text{R-N-C} - \text{O-R-C} - \text{N-H}_n \right] \text{R'-NCO}
\end{align*}
\]

**Equation 1 The reaction equation of TDI and polyether polyol**

According to the hardness requirement of the strippable coating film, the hard segment content was 36.8%.

\[
\begin{align*}
    \omega_h &= \frac{m_1 + m_2}{m_1 + m_2 + m_3} \tag{1} \\
    m_1 &= \frac{m_2}{M_2} + \frac{m_3}{M_3} \tag{2}
\end{align*}
\]

Where, \( \omega_h \) represents the content of hard segment. \( m_1 \), \( m_2 \) and \( m_3 \) indicate the mass of TDI, chain extender and polyether polyol, respectively. \( M_1 \), \( M_2 \) and \( M_3 \) express the molecular weight of TDI, chain extender and
polyether polyol, respectively. The molecular weight of TDI, chain extender and polyether polyol were $M_1=174.2$, $M_2=267.2$ and $M_3=849$, respectively. Substitute the data into Equation (1) and (2):

$$m_2=0.2282m_3$$
$$m_1=0.3540m_3$$

Then, when the moles number of polyether polyol is set to 1, the ratio of moles number of the TDI, the chain extender, and the polyether polyol was calculated to be 1.73:0.73:1.

### 3.2. Optimization of chain extender

In order to synthesize TDI and polyether polyol into strippable film, it is necessary to add a chain extender into the solution. In this study, the initial setting time and strippable time of contamination detergent with GLY or MOCA as extender were tested respectively. The results were shown in Fig. 2. For GLY, the initial setting time was less than 1 min. The short initial time of contamination detergent made the solution easier to solidify in the stirring process, which made it difficult to spray. For MOCA, the initial setting time was about 12 min, which was plenty to stir and spray. Therefore, MOCA was selected as appropriate chain extender in this study.

![Fig. 2 The initial setting time and strippable time of contamination detergent that the GLY or MOCA as chain extender](image)

### 3.3. Optimization of catalyst addition

As seen in Fig. 2, the strippable time of contamination detergent was exceeded 2h, which could not achieve the effect of rapid decontamination. In order to shorten the strippable time, an appropriate catalyst was added into the formula. Dibutyltin dilaurate has good catalytic efficiency and stability in polyether polyol\[9,10\]. In the experiment, the additive amount of dibutyltin dilaurate was optimized. The result was shown in Fig. 3. The strippable time decreased with the amount of catalyst increased from 0.2 wt% to 1.0 wt%. The strippable time tended to be stable when the addition of catalyst exceeded 1.0 wt%. The reason is that the amount of catalyst reached saturation when the addition of catalyst is 1.0 wt%. Therefore, it was confirmed that the optimal addition amount of dibutyltin dilaurate was 1.0 wt%, and the strippable time of contamination detergent was about 1.5h.

![Fig. 3 The relation of catalyst addition and strippable time](image)
3.4. Performance test of contamination detergent
The main performance including tensile strength and decontamination rate of the contamination detergent were tested. The tensile strength tests of film materials was carried out in accordance with GB/T 1040-1992 at a test speed of 50mm/min. The results of tensile strength were recorded in Table 1. The result indicated that the strippable coating film has good mechanical properties. The decontamination rates of contamination detergent for uranium dust were measured on the surface of concrete and terrazzo. The results were shown in Table 2 and Table 3, the photographs were shown in Fig. 4. The decontamination rates of contamination detergent for uranium dust on the two surfaces were exceeded 90%. Therefore, it is suggested that the contamination detergent can be used for uranium dust decontamination on the surface of concrete and terrazzo.

Table 1. The tensile strength of strippable coating film at a speed of 50mm/min

| Test number | Tensile strength/MPa |
|-------------|----------------------|
| 1           | 2.74                 |
| 2           | 2.55                 |
| 3           | 2.70                 |
| 4           | 2.73                 |
| 5           | 2.26                 |

Table 2. The decontamination efficiency of contamination detergent for uranium dust on the surface of concrete

| Test number | Before decontamination/cpm | After decontamination/cpm | Decontamination efficiency/% | Average decontamination efficiency/% |
|-------------|----------------------------|---------------------------|-----------------------------|-------------------------------------|
| 1           | 734                        | 82                        | 88.83                       | 90.54                               |
| 2           | 689                        | 61                        | 91.15                       |                                     |
| 3           | 693                        | 65                        | 90.62                       |                                     |
| 4           | 695                        | 62                        | 91.08                       |                                     |
| 5           | 670                        | 60                        | 91.04                       |                                     |

Table 3. The decontamination efficiency of contamination detergent for uranium dust on the surface of terrazzo

| Test number | Before decontamination/cpm | After decontamination/cpm | Decontamination efficiency/% | Average decontamination efficiency/% |
|-------------|----------------------------|---------------------------|-----------------------------|-------------------------------------|
| 1           | 609                        | 40                        | 93.43                       |                                     |
| 2           | 680                        | 81                        | 88.09                       |                                     |
| 3           | 632                        | 60                        | 90.51                       | 90.68                               |
| 4           | 610                        | 54                        | 91.15                       |                                     |
| 5           | 625                        | 61                        | 90.24                       |                                     |

Fig. 4 The photographs of decontaminate process on the surface of concrete (a) and terrazzo (b).
4. Conclusion
In this study, a synthetic formula of a quick-drying type of radioactive contamination detergent based on polyurethane was obtained by theoretical calculation and experiment. MOCA was selected as a chain extender by screening experiment. The basic molar ratio of toluene diisocyanate, MOCA and polyether polyol was 1.73:0.73:1. Dibutyltin dilaurate was selected as the catalyst, and the addition amount was 1.0 wt%. What’s more, under the optimal condition, the single removal rate of the contamination detergent on concrete and terrazzo surface were both over 90%. In short, the formula of contamination detergent provided a new method for the removal of uranium dust.

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