A novel self-embrittling strippable coating for radioactive decontamination based on silicone modified styrene-acrylic emulsion

Jing Wang¹ ², Jianhui Wang¹ ², Li Zheng¹ ², Jian Li¹, Can Cui¹ and Linmei Lv¹

¹Luoyang Hydraulic Engineering Technique Institute, Xishan Road, Luoyang, China
²Engineering Lab of Henan Province for Radioactive Decontamination Material and Equipment Technology, Wangcheng Street, Luoyang 471023, China

e-mail: jwang07@163.com

Abstract. Silicone modified styrene-acrylic emulsion and butyl acrylate were used as a main film-forming agent and an additive respectively to synthesize a self-embrittling strippable coating. The doping mass-ratio of butyl acrylate was adjusted at 0, 5%, 10%, 15%, 20%, and the results indicated the optimized doping ratio was 10%. Ca(OH)₂ was used to promote the coating film self-embrittling at a moderate doping mass-ratio of 20%. The synthesized coating’s coefficients of α and β decontamination on concrete, marble, glass and stainless steel surfaces were both greater than 85%, which indicated the synthesized coating is a promising cleaner for radioactive decontamination.

1. Introduction
Strippable film coatings were highly focused in the last few decades for the application of radioactive decontamination. Numerous studies have shown that strippable film coatings were suitable for radioactive decontamination on flat surfaces [1-6]. However, continuously strippable films were more difficult for decontamination on complex geometrical surfaces or narrow space surfaces. Research have shown that self-embrittling strippable coating could be used for decontamination on those complex geometrical surfaces [7, 8]. This provided a new idea for the radioactive decontamination on the complex geometrical surfaces. The phenomenon that some silicone modified styrene-acrylic emulsion coatings presented embrittling glassy state after solidification was discovered in our previous studies. The cracking was irregular, nevertheless, the silicone modified styrene-acrylic emulsion was likely to become a new type of self-embrittling strippable coating for radioactive decontamination. Butyl acrylate (BA) was widely used as a typical soft functional monomer [9-11], which could regulate the flexibility and weaken the glassy state of coating films. The aim of this work was to prepare a novel self-embrittling strippable coating for radioactive decontamination based on silicone modified styrene-acrylic emulsion. In this paper, the formula of coating was optimized by evaluating its self-embrittling and strippable performance.

2. Experimental

2.1. Raw materials
Silicone modified styrene-acrylic emulsion was used for film-forming main agent, which was provided by Beijing Tongzhou Huyi Chemical Factory, and the material designation was HB-C. The
main performance parameters of silicone modified styrene-acrylic emulsion were listed in Table 1. Butyl acrylate, provided by Hubei Qirui Chemical Factory, was used as an additive to regulate the flexibility and strippable performance of coating, and the material designation was BA-163. Calcium hydroxide was used to improve the self-fragmentation property of the coating.

**Table 1. Performance parameters of silicone modified styrene-acrylic emulsion.**

| Property  | Solid content/% | Viscosity/ mPa·S | PH   | Glass transition temperature/°C |
|-----------|-----------------|-----------------|------|-------------------------------|
| Parameter | 48 ± 1          | 1400~1600       | 7.5~8.5 | 25                            |

2.2. **Procedure**

Different proportions of silicone modified styrene-acrylic emulsion and butyl acrylate as shown in table 2 were placed in a flask and stirred at 300r/min. Then calcium hydroxide was added and stirred at 1000r/min. The mixture was stirred for about 20 minutes. After the suspension was dispersed evenly, the thickener was added and the resulting mixture was stirred at 500r/min for about 20 minutes. Finally, the mixture was filtered to obtain a homogeneous coating sample.

**Table 2. Formulae of coatings.**

| No. of coating | Mass of silicone modified styrene-acrylic /g | Mass of butyl acrylate /g | Mass of Ca(OH)$_2$/g |
|----------------|---------------------------------------------|---------------------------|---------------------|
| C1             | 100                                         | 0                         | 0                   |
| C2             | 100                                         | 5                         | 0                   |
| C3             | 100                                         | 10                        | 0                   |
| C4             | 100                                         | 15                        | 0                   |
| C5             | 100                                         | 20                        | 0                   |
| C6             | 100                                         | 10                        | 10                  |
| C7             | 100                                         | 10                        | 20                  |
| C8             | 100                                         | 10                        | 30                  |

3. **Results and discussion**

3.1. **Content optimization of butyl acrylate**

The mass-contents of BA were varied from 0 to 20% in the coating samples of C1–C5. The self-embrittling and strippable performances of these coating film were listed in table 3.

**Table 3. Properties of coating films with different contents of BA.**

| No. of coating | Mass-content of BA/% | Self-embrittling performance | Strippable performance |
|----------------|----------------------|------------------------------|------------------------|
| C1             | 0                    | self-embrittling, hard fragment, uniform in fragment size | difficult to strip     |
| C2             | 5                    | self-embrittling, hard fragment, uniform in fragment size | difficult to strip     |
| C3             | 10                   | self-embrittling, soft fragment, uniform in fragment size | strippable            |
| C4             | 15                   | self-embrittling, soft fragment, non-uniform in fragment size | strippable            |
| C5             | 20                   | difficult to embrittle, soft coating film | strippable            |

As can be seen in table 3, silicone modified styrene-acrylic emulsion coating film could self-embrittling with hard fragment. But the adhesion to concrete surface was too strong to strip. After the introduction of BA, the coating film gradually became softer and more strippable. When the content of BA increased to 15%, the coating film of C4 represented a non-uniform fragment. And even when the content of BA increased to 20%, the coating film of C5 was difficult to embrittle. As a classical soft
functional monomer, BA could decrease the hardness of coating film effectively. And also could modify the adhesion to concrete surface, which made the coating film strippable. However, excessive BA could make coating film too soft to self-embrittling. As can be seen from the formula experiment, the most appropriate doping mass-content of BA was 10%.

3.2. Content optimization of Ca(OH)$_2$

Addition of varied mass contents of Ca(OH)$_2$ from 10~30% has been achieved in the samples of C6–C8, and their properties and embrittling photos of the coating films were shown in table 4 and figure 1, respectively. The coating C3 that with no additional Ca(OH)$_2$ was also included as comparison.

Table 4. Properties of coating film with different contents of Ca(OH)$_2$.

| No. of coating | Mass content of Ca(OH)$_2$/% | Fragment size/mm | Strippable performance |
|---------------|-----------------------------|-------------------|------------------------|
| C3            | 0                           | 20~30             | strippable             |
| C6            | 10                          | 10~20             | strippable             |
| C7            | 20                          | 5~10              | strippable             |
| C8            | 30                          | 20~30             | difficult to strip     |

![Figure 1. Embrittling photos of coatings with different Ca(OH)$_2$ contents.](image1)

As can be seen in table 4 and figure 1, moderate doping of Ca(OH)$_2$ could promote the coating films to self-embrittle, resulting in the decreased fragment size of coating films. When the mass-content of Ca(OH)$_2$ was 20%, the coating C7 self-embrittled well with a fragment size of 5~10mm. But when the doping content of Ca(OH)$_2$ increased to 30%, the coating film of C8 became hard and difficult to be stripped. Excessive doping of Ca(OH)$_2$ made the coating film harder and the adhesion to concrete surface stronger, mainly due to the transformation of Ca$^{2+}$ to CaCO$_3$ during solidification. As can be seen from the formula experiments, the suitable doping mass-content of Ca(OH)$_2$ was 20%. Consequently, the optimized formula was obtained as coating C7 through formula experiments.

3.3. Viscosity of coating

Viscosity affected the application mode of the coating. Figure 2 was the viscosity-temperature chart between -10~30°C of the coating C7. It showed that the coating had obvious viscosity-temperature characteristic. The coating’s viscosity was below 2000 mPa·S when the temperature was above 5°C. Under this condition, the coating could be quickly sprayed by spraying machine. When the
temperature was below 5℃, the coating was too adhesive to spray. Under this condition, it was just suitable for using as a brush-applied coating.

![Figure 2. Viscosity-temperature chart of coating C7.](image)

### 3.4. Coefficients of decontamination

Coefficients of decontamination on different surfaces of the coating were measured according to a previous literature [12], and the result was listed in table 5. The coefficients of α and β decontamination on glass and stainless steel surface were both above 90%, which were higher than those on concrete and marble surface. The above difference was ascribed to the porosity of different surfaces. Concrete had porous surface, therefore the coefficient of decontamination was smaller than those on other non-porous surface. To sum up, coefficients of α and β decontamination on four kinds of surface were both greater than 85%. It indicated the prepared coating was a promising cleaner for radioactive decontamination.

| Surface    | Coefficient of α decontamination/% | Coefficient of β decontamination/% |
|------------|------------------------------------|------------------------------------|
| Concrete   | 85.1                               | 86.3                               |
| Marble     | 92.2                               | 87.5                               |
| Glass      | 98.4                               | 92.1                               |
| Stainless steel | 97.6                       | 91.3                               |

### 4. Conclusions

In order to obtain a high performance coating for radioactive decontamination, the ratio of different components was optimized. The viscosity and coefficient of decontamination were also characterized. The conclusions can be drawn as follows.

1. The ratio of the main film-forming agent and the additive was optimized. Butyl acrylate used as an additive could decrease the hardness of coating film effectively, and an optimized doping mass-ratio of BA was 10%.

2. Ca(OH)2 used as a filler could promote the coating film to self-embrittlement at a moderate doping mass-ratio of 20%.

3. Coefficients of α and β decontamination on concrete, marble, glass and stainless steel surfaces were both greater than 85%. It indicated that the prepared coating is a promising cleaner for radioactive decontamination.
Acknowledgements
This work was financially supported by the Outstanding Talents Innovation Foundation of Henan Province (No. 154200510029).

References
[1] Bernaola O A, Filevich A 1970 Health Physics 19 685
[2] Tan M, Whitaker J D and Schwartz D T 2009 J Hazard Mater. 162 1111
[3] Guidi G, Cumo F, Santoli L D 2010 Clean Techn Environ Policy 12 283
[4] Kohli R 2015 Particle Adhesion and Removal (Hoboken: John Wiley & Sons, Inc.)
[5] Moore R C, Tucker M D and Jones J A 2009 U.S. Patent 7,514,493
[6] Andre S, Andrieu X and Melin G 2008 U.S. Patent 0,159,706
[7] Liu R L, Li Y T and Zhou Y L 2016 Chemistry Letters 45 1
[8] Wang T Y and Li S 2014 C.N. Patent 103,695,205
[9] Sebastian G R and Axel H E M 1999 Macromolecules 32 8331
[10] Wang J C, Chen Y H and Wang J H 2016 e-Polymers 9 1079
[11] Bose R K, Hohlbein N and Garcia S J 2015 Phys Chem Chem Phys 17 1697
[12] Wang X D, Wang X M and Xing G P 2001 Chin J Radiol Health 10 167