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Development of Self-fracturing Atomizing Fixative for Radioactive Aerosol Control

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Abstract. This paper focuses on the need to control aerosols generated during pipeline cutting in nuclear facility decommissioning. A self-fracturing aerosol atomization fixing agent has been developed. Styrene (St) is used as the hard monomer, and methyl methacrylate (MMA) and methacrylic acid (MAA) are used as functional monomers. The fixing agent lotion and the properties of the formed film, as well as their impact on the capture performance of titanium dioxide aerosols, are investigated through emulsion polymerization synthesis. The results show that the fixing agent achieves a 93.17% capture rate for titanium dioxide aerosols within 6 hours. After being fixed by the fixing agent, the aerosol resuspension rate is only 5.10%. Moreover, the formed film can spontaneously fragment into pieces smaller than 4 cm^2 within 3 hours. This fixing agent possesses good capture efficiency and self-fracturing characteristics, providing an effective and easy-to-operate technical approach with minimal secondary pollution for the safe control of radioactive aerosols.

Keywords: radioactive aerosol control, self-fragmenting, polymer, atomization fixative

1. Introduction

A dispersion system formed by solid or liquid radioactive particulate matter suspended in air is defined as radioactive aerosol [1]. During the decommissioning of nuclear facilities and nuclear accident emergency responses, large quantities of radioactive aerosols are generated. These aerosols can cause external radiation damage to the skin, or internal radiation hazards to the human body when the radionuclides they carry enter the body [2]. Therefore, suppressing and removing radioactive aerosol particles in the air is of great significance for human health and environmental protection.

Current radioactive aerosol purification/suppression technologies and their working principles are shown in Table 1 [3,4].

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Table 1. Working principles of radioactive aerosol purification technologies

	Physical Filtration Method	Electrostatic Precipitation Method	Bubble Scrubbing Method	Pressure Dissolved Air Flotation Method	Oxidation Electric Field Capture Method	Atomizing Fixation Method
Kinetic energy reduction	√	√				√
Particle size enlargement					√	√
Utilization of chargeability		√			√	
Liquid-phase absorption			√	√		

Compared with other radioactive aerosol fixation technologies, the atomizing fixation technology has the advantages of simple operation, small amount of secondary waste, and adaptability to various operating scenarios. After atomizing fixation treatment, the level of radioactive aerosols in the treated area can be significantly reduced, which greatly lowers the hazards to operators [5].

In terms of the types of atomizing fixatives for aerosols, various types have been developed so far, including saccharide-based, foam-based, water-soluble polymer-based, copolymer-based and modified resin-based fixatives [6-11]. Each type of fixative has distinct characteristics in environmental friendliness, safety, cost and service life, making them suitable for different working conditions. In summary, atomizing fixation technology can significantly reduce aerosol concentration and effectively control radiation risks, possessing good engineering application prospects. Meanwhile, improving and optimizing the formulation of aerosol atomizing fixatives is also of great importance for radioactive aerosol suppression.

In practical applications, it has been found that for radioactive aerosols generated during operations such as pipeline cutting, fixatives whose films can spontaneously fragment under natural conditions have the advantages of easy film collection and convenient subsequent disposal. Through literature research, it is known that Zhang Kun et al. [12] prepared a self-fracturing decontamination solution using modified silicone resin molecules as the main monomer. The average decontamination rate of this solution can reach over 85%, and it can dry and agglomerate into fragments within 6 hours.

Research findings indicate that there are relatively few studies on self-fracturing atomizing fixatives, and the research on the effects of different component contents on their working efficiency and self-fragmentation performance is quite limited. Referring to domestic and foreign literatures and patents on decontaminants and fixatives with published formulations [13-15], this study used styrene (St) as the hard monomer, and introduced modified monomers methyl methacrylate (MMA) and methacrylic acid (MAA) to reduce the contact angle of the emulsion, adjust its viscosity, and improve the properties of the formed films. Finally, aerosols composed mainly of micron-sized titanium dioxide powder (with a particle size range of 0.1-5 μm) were used as the experimental objects. The optimal fixative formulation was determined by testing the capture efficiency of different samples on these aerosols.

DOI: 10.54254/2755-2721/2026.31807

2. Materials and methods

2.1. Experimental reagents

Table 2. List of experimental reagents

No.	Reagent	Manufacturer	Grade
1	Styrene (St)	Tianjin Dama Chemical Reagent Factory	Analytical Grade
2	Methyl Methacrylate (MMA)	Tianjin Dama Chemical Reagent Factory	Analytical Grade
3	Methacrylic Acid (MAA)	Tianjin Dama Chemical Reagent Factory	Analytical Grade
4	Octylphenol Polyoxyethylene Ether (OP-10)	Tianjin Dingshengxin Chemical Co., Ltd.	Analytical Grade
5	Sodium Dodecyl Sulfate (SDS)	Aladdin Reagent (Shanghai) Co., Ltd.	Analytical Grade
6	Sodium Carbonate (Na_2CO_3)	Aladdin Reagent (Shanghai) Co., Ltd.	Analytical Grade
7	Potassium Persulfate (KPS)	Aladdin Reagent (Shanghai) Co., Ltd.	Analytical Grade
8	Deionized Water	Laboratory-made	-
9	Titanium Dioxide Powder	Hebei Ruihuang Metal Materials Co., Ltd.	Industrial Grade

In this study, titanium dioxide powder was used to generate aerosols to replace radioactive aerosols in actual scenarios. Titanium dioxide is a very stable, non-toxic inert oxide. It does not undergo chemical reactions at room temperature, does not absorb moisture, and is not easily decomposed, which can well avoid the interference of irrelevant variables on aerosol capture. Meanwhile, titanium dioxide has good processability and can be precisely fabricated into particles within a specific size range to simulate radioactive aerosols in actual scenarios.

2.2. Experimental instruments

Table 3. List of experimental instruments

No.	Instrument	Manufacturer
1	DF-101 Heat Collecting Constant Temperature Magnetic Stirrer	Zhengzhou Biochemical Instrument Co., Ltd.
2	Analytical Balance	Guangzhou Shenhua Biotechnology Co., Ltd.
3	DV2TLV Viscometer	Brookfield Engineering Laboratories, Inc., USA
4	Surface Tension-Contact Angle Meter	Kunshan Beidou Precision Instrument Co., Ltd.
5	PT-998 Brushless Sprayer	Wuxi Putian Special Spraying Equipment Co., Ltd.
6	Aerosol Control Performance Test Bench	Self-developed
7	SAG-410 Dust Generator	Topas GmbH, Germany
8	ELPI+ Particulate Analyzer	Dekati Ltd., Finland

2.3. Experimental procedures

The specific synthesis steps of the self-fracturing atomizing fixative are as follows:

1. According to the required synthesis amount and solid content of the fixative, add an appropriate amount of mixed emulsifier (sodium dodecyl sulfate (SDS) and OP-10) and deionized water into a four-necked flask equipped with a stirrer, a thermometer and a constant pressure dropping funnel. Heat the mixture to 72 ℃ in a water bath and stir at 180 r/min.

2. Add appropriate amounts of polymerizable monomers (styrene (St), methyl methacrylate (MMA) and methacrylic acid (MAA)) into a beaker to prepare a mixed monomer solution.

DOI: 10.54254/2755-2721/2026.31807

3. Add 1/5 of the total mass of the mixed monomer solution and 1/3 of the total mass of the initiator potassium persulfate (KPS) into the four-necked flask, respectively. Transfer the remaining mixed monomer solution into the constant pressure dropping funnel for later use.

4. When the solution in the four-necked flask turns light blue, slowly add the remaining mixed monomer solution dropwise into the flask, and simultaneously add the remaining initiator and an appropriate amount of Na_2CO_3. Raise the temperature to 75 ℃.

5. Continue to slowly add the remaining mixed monomer solution dropwise into the four-necked flask, ensuring that the dropping process is completed at the same time as the previous step. After the dropping is finished, maintain the temperature for 1 hour and then discharge the product.

The specific experimental flow is shown in Figure 1.

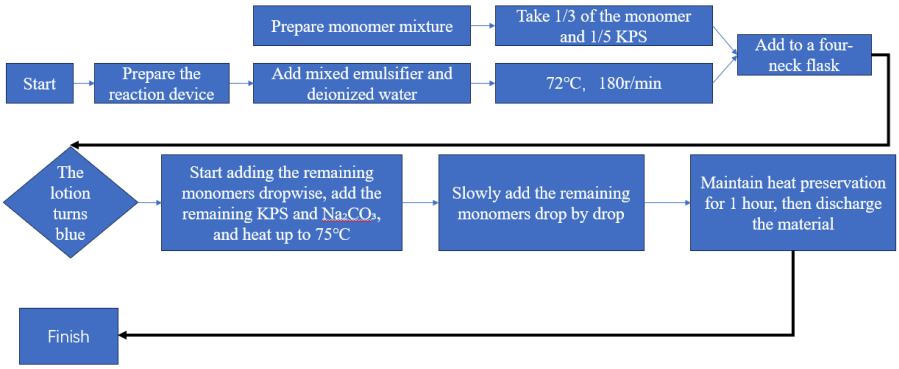


Figure 1. Synthesis process flowchart of self-fracturing radioactive aerosol atomizing fixative

Based on the above steps, three types of self-fracturing atomizing fixatives with different monomer contents were synthesized, named Sample 1, Sample 2 and Sample 3 respectively.

2.4. Performance tests

2.4.1. Natural sedimentation experiment of titanium dioxide aerosols

Use the SAG-410 dust generator to generate a certain concentration of titanium dioxide aerosols inside the aerosol control performance test bench. Turn off the SAG-410 dust generator after 20 minutes. Wait until the number concentration of the aerosols stabilizes, then use the ELPI+ particulate analyzer to sample and measure the central area of the test bench for 15 seconds. Record this moment as Time 0. Repeat the above sampling steps at 30 min, 60 min, 120 min, 180 min, 240 min, 300 min and 360 min respectively, and summarize the natural sedimentation law of the aerosols.

2.4.2. Determination of capture rate of water and fixatives

Use the SAG-410 dust generator to generate a certain concentration of titanium dioxide aerosols inside the aerosol control performance test bench. Turn off the SAG-410 dust generator after 20 minutes. Wait until the number concentration of the aerosols stabilizes, then use the ELPI+ particulate analyzer to sample and measure the central area of the test bench for 15 seconds. Record this moment as Time 0. Atomize water or the fixative samples into the test bench using an atomization device. Repeat the above sampling steps at 30 min, 60 min, 120 min, 180 min, 240 min,

DOI: 10.54254/2755-2721/2026.31807

300 min and 360 min respectively. The capture rate data at each time point is calculated using Formula (1) [16]:

$$\eta t = \frac{(c0-ct)}{c0} \times 100\% \quad (1)$$

Where: η_t is the aerosol capture rate at time t; c_0 is the aerosol concentration at Time 0, cm^{-3}; c_t is the aerosol concentration at time t, cm^{-3}.

The specific experimental setup is shown in Figure 2.

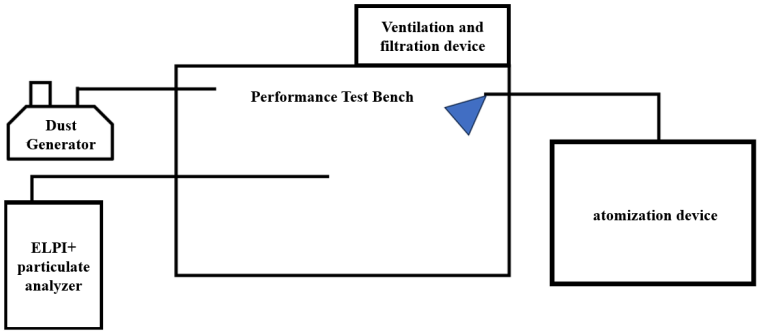


Figure 2. Schematic diagram of the experimental setup

2.4.3. Determination of resuspension rate

After completing the natural sedimentation experiment and the capture rate determination experiment of each fixative sample, let the test bench stand for 24 hours. After measuring the aerosol concentration, researchers enter the test bench to disturb the fixed aerosols to promote their resuspension. Sample and measure the aerosols every 30 seconds for a total duration of 300 seconds. The resuspension rate of each sample is calculated using Formula (2) [17]:

$$\varphi i = \frac{(ci2-ci1)}{c0} \times 100\% \quad (2)$$

Where: φ_i is the resuspension rate of Sample i; c_{i1} is the aerosol concentration of Sample i before disturbance, cm^{-3}; c_{i2} is the aerosol concentration of Sample i after disturbance, cm^{-3}.

2.4.4. Determination of film-forming time and film self-fracturing performance

Take 10 ml of fixative samples and evenly drop them into petri dishes with a diameter of 10 cm at different temperatures. Record the fluidity of the fixatives, the film-forming time and the self-fragmentation degree of the dried films, so as to evaluate the working performance of the fixatives.

3. Results and discussion

3.1. Natural sedimentation experiment

Figure 3 shows the natural sedimentation law of titanium dioxide aerosols. Data analysis reveals that the natural sedimentation of titanium dioxide aerosols exhibits a trend of being fast at first and then slow. The natural sedimentation rate reaches 25.01% within 1 hour, 38.08% within 3 hours, and

DOI: 10.54254/2755-2721/2026.31807

53.82% within 6 hours. These experimental data indicate that under natural sedimentation conditions, the sedimentation rate of titanium dioxide aerosols is relatively low within a short period of time. Therefore, it is necessary to use aerosol fixatives for the rapid suppression of aerosols.

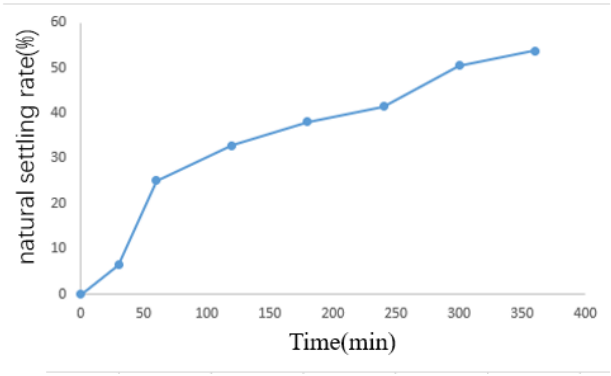


Figure 3. Natural sedimentation behavior of titanium dioxide aerosol

3.2. Determination of capture rate

Figure 4 shows the variation of capture rate with time in the experiment of water and the three fixative samples capturing titanium dioxide aerosols. Calculations show that within 1 hour, the capture rates of water, Sample 1, Sample 2 and Sample 3 are 52.1%, 44.23%, 74.85% and 59.43% respectively. Within 3 hours, the capture rates are 58.99%, 63.45%, 82.92% and 74.61% respectively. Within 6 hours, the capture rates are 66.44%, 84.83%, 93.17% and 89.25% respectively. The above data indicate that within a certain range, the higher the addition amount of St, the better the capture ability of the synthesized radioactive aerosol atomizing fixative for titanium dioxide aerosols. The main reason for this is the differences in viscosity and contact angle. However, with the continuous increase of St addition amount, the viscosity of the emulsion continues to rise, making it more difficult to atomize the emulsion, thus leading to a decrease in capture rate. Meanwhile, the contact angle of Sample 2 is lower than that of the other two samples, which means it is easier to wet the titanium dioxide aerosol particles upon collision, resulting in the highest capture rate [18].

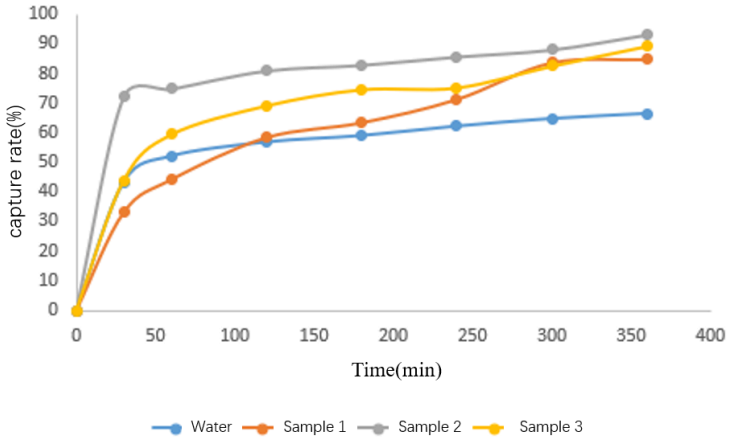


Figure 4. Capture effectiveness of water, sample 1, sample 2 and sample 3 on titanium dioxide aerosol

DOI: 10.54254/2755-2721/2026.31807

Compared with the self-fracturing decontamination solution reported in the literature [12], the capture rate of this sample is increased from 85% to over 93%. In comparison with other traditional aerosol atomizing fixatives, this sample can also reach an average capture rate of 90%.

3.3. Determination of resuspension rate

Figure 5 shows the variation of the number of aerosol particles in the test bench after disturbance in the resuspension experiment of Sample 2. After treatment with the fixative, the titanium dioxide aerosol particles are effectively fixed on the ground. The maximum resuspension rate caused by disturbances such as personnel movement and exhaust ventilation is only 5.10%, indicating that this sample has excellent fixation ability.

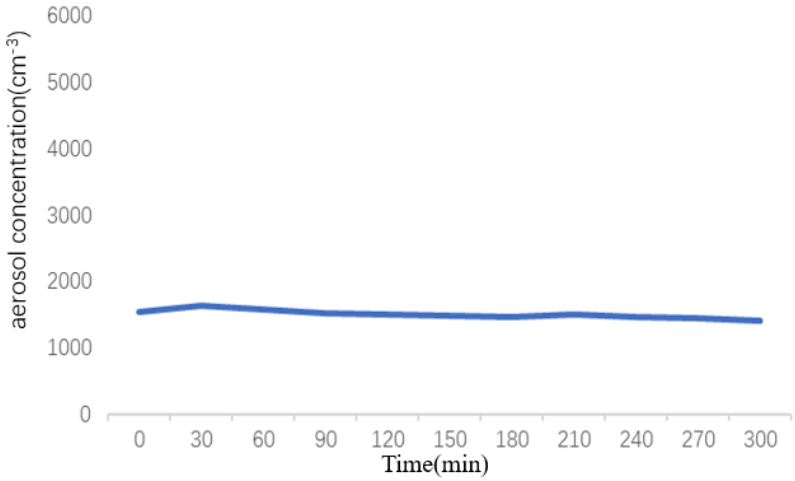


Figure 5. Aerosol resuspension after disturbance in the resuspension experiment of sample 2

Compared with the water-soluble synthetic resin "Clear Court C720Green" atomizing fixative applied in the Fukushima nuclear accident [11], both can significantly reduce the resuspension rate of aerosols, keeping it at a low level, and achieve a certain fixation effect on the controlled aerosols.

3.4. Film-forming time and film self-fracturing performance

Table 4 shows the self-fragmentation time of Sample 2 fixative at different temperatures.

Table 4. Self-Fragmentation time of fixative sample at different temperatures

Temperature/°C	Film-Forming Time
0	Over 72 h
10	Within 12 h
20	Within 3 h
30	Within 1 h

Experiments have proven that the synthesized emulsion can form films naturally within 3 hours at room temperature, and the films possess self-fracturing properties, with the area of the fragmented pieces smaller than 4 cm^2. The specific self-fracturing situation of the films and the size of the fragments are shown in Figure 6 and Figure 7.

DOI: 10.54254/2755-2721/2026.31807

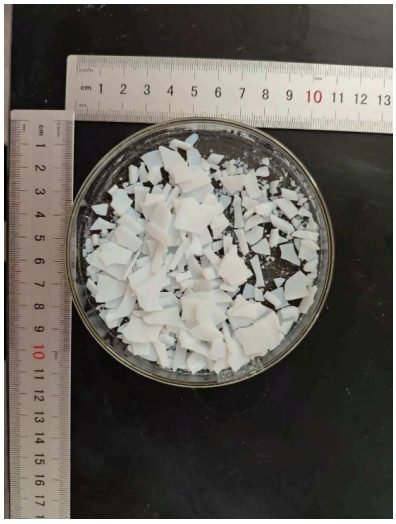


Figure 6. Schematic diagram of film self-fragmentation



Figure 7. Schematic diagram of fragment size

4. Conclusions

In this study, styrene (St) was used as the matrix monomer, methacrylic acid (MAA) and methyl methacrylate (MMA) as the functional monomers, potassium persulfate (KPS) as the initiator, and a mixture of sodium dodecyl sulfate (SDS) and OP-10 as the emulsification system. Fixative samples with different monomer ratios were prepared via emulsion polymerization. Subsequently, the fixation effects of the three samples on titanium dioxide aerosols were evaluated through aerosol capture experiments. The results showed that Sample 2 exhibited the optimal capture performance, with a maximum capture rate of 93.17% within 6 hours. After atomization fixation with this sample, the resuspension of aerosol particles caused by disturbance was significantly suppressed, with the resuspension rate only 5.10%.

Compared with existing products, the self-fracturing atomizing fixative developed in this study shows advantages in multiple performance aspects. Compared with the self-fracturing decontamination solution reported in the literature, this study has achieved an improvement in capture efficiency. Meanwhile, in comparison with traditional saccharide-based, foam-based or

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water-soluble polymer-based fixatives, this fixative not only maintains high capture efficiency, but also possesses the film self-fracturing property, which greatly simplifies the subsequent cleaning process, reduces the burden of secondary waste disposal, and is more suitable for applications in operations such as pipeline cutting. In addition, its raw materials are common and the synthesis process is simple, showing good engineering application prospects and environmental friendliness. While effectively controlling the hazards of radioactive aerosols, it focuses on solving the key problem of waste minimization, providing an effective solution to related issues.

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