

Preparation and Performance of a Color-indicating Water-based Peelable Decontamination Coating

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Abstract

In nuclear facility decommissioning and nuclear accident emergency response, most existing decontamination materials lack real-time monitoring capabilities for radioactive nuclides such as uranium, making it difficult to precisely control the decontamination process and immediately evaluate decontamination effectiveness. To address the limitations of current materials, this study prepared a peelable decontamination material with uranium colorimetric tracing functionality. By introducing specific chemical groups, this material can undergo significant color changes during the adsorption of uranium contamination, thereby enabling visual monitoring of uranium pollution. This paper discusses relevant factors affecting the decontamination and colorimetric performance processes through testing of the decontamination agent's stress-strain behavior, acid-base environment, wettability, and surface spreading degree. Systematic studies were conducted on the material's film-forming performance, decontamination efficiency, colorimetric sensitivity, and environmental stability to verify its feasibility and advantages in practical applications.

Full Text

Preparation and Performance Study of Color-Indicating Traceable Water-Based Strippable Decontamination Coating

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Abstract

In nuclear facility decommissioning and nuclear accident emergency response, existing decontamination materials mostly lack real-time monitoring capabilities for radioactive nuclides such as uranium, making it difficult to precisely control the decontamination process and immediately evaluate its effectiveness. To address this limitation, this study prepared a strippable decontamination material with a “uranium” colorimetric tracing function. By introducing specific chemical groups, this material undergoes significant color changes during uranium adsorption, enabling visual monitoring of uranium contamination. This paper systematically investigated the material’s film-forming performance, decontamination efficiency, colorimetric sensitivity, and environmental stability through stress-strain tests, pH environment analysis, wettability measurements, and surface spreading assessments. The factors affecting decontamination and colorimetric performance were discussed to verify the feasibility and advantages of this material in practical applications.

Keywords: radioactive decontamination; strippable decontamination material; block copolymer; colorimetric tracing

Introduction

Nuclear facility decommissioning is a complex systematic engineering project with significant variations in strategies, technologies, and required timelines. Particularly for large nuclear facilities such as reactors and reprocessing plants, decommissioning is typically conducted in phases over extended periods. Therefore, scientific and rational zoning of decommissioning areas is crucial. Generally, nuclear facility decommissioning zones are divided into: (1) the decommissioning operation area for core activities such as nuclear facility and radioactive material cleanup, cutting, excavation, and decontamination; (2) the radioactive waste classification area; and (3) the monitoring area requiring continuous surveillance. To meet operational requirements, the decommissioning operation area is further subdivided into controlled areas, supervised areas, and unrestricted areas [1] (shown in [Figure 1: see original paper]). When selecting decontamination methods, multiple factors must be considered, including expected worker radiation dose, target decontamination levels and their measurability, feasibility of achieving target levels with existing technologies, contamination types, and source terms. Consequently, radioactive decontamination materials, as the core means for removing radionuclides (such as uranium, cesium, strontium, cobalt, etc.), are critical for environmental protection, human health, and sustainable nuclear industry development.

Compared with other decontamination methods, the strippable decontamination approach offers advantages of simple operation, excellent decontamination effectiveness, low liquid waste generation, and non-corrosiveness to nuclear facilities and equipment, making it an economical and practical method widely used in nuclear facility decommissioning [2-5]. Among these, radioactive strippable

decontamination agents with colorimetric tracing capabilities enable construction personnel to intuitively assess on-site nuclide contamination conditions during decontamination, allowing timely adjustment of controlled area boundaries. Unlike “point” detection by detectors and test kits, such decontamination agents provide “area” detection applicable to large spaces for nuclide range, level, and type identification. In 2001, researchers at the University of Texas, including Neil Gray [6], prepared and investigated decontamination agents with different polymer compositions, innovatively incorporating the color-responsive indicator 2-(5-bromo-2-pyridylazo)-5-diethylaminophenol (Br-PADAP) to enable different color changes for uranium and plutonium contamination, thereby expanding the performance and applications of strippable films. However, currently used indicators suffer from poor dispersibility and stability, tending to agglomerate in environmentally friendly water-based strippable decontamination agents, which affects colorimetric and decontamination effectiveness. Furthermore, no further investigation has been conducted on the minimum detection limits and decontamination applications of such colorimetric traceable strippable decontamination materials. Based on these considerations, the core objective of this study is to prepare a strippable decontamination material with both high decontamination efficiency and uranium colorimetric tracing function to meet the need for timely monitoring of contaminants and contamination levels during radioactive decommissioning. This material should not only efficiently enrich uranium contamination but also visually reflect the decontamination process and effectiveness through color changes after uranium adsorption.

2. Instruments and Reagents

2.1 Materials

- PEA-b-PAA block copolymer
- Polyvinyl alcohol (1788)
- Polyvinylpyrrolidone
- Ethylenediaminetetraacetic acid (EDTA)
- 2-(5-bromo-2-pyridylazo)-5-(diethylamino)phenol (Br-PADAP)

2.2 Experimental Instruments

- Electric thermostatic water bath
- Electric stirrer
- Vacuum drying oven
- Universal testing machine
- Contact angle measuring instrument

2.3 Preparation of the “Uranium” Colorimetric Traceable Strippable Decontamination Material

The preparation procedure was as follows: (1) A quantity of distilled water was mechanically stirred in a beaker and heated to 90°C, after which PVA (10

wt%) was gradually added and thoroughly mixed. (2) Following complete PVA dissolution, PVP (6 wt%) was added and thoroughly mixed, and the mixture was stirred for 20 minutes to ensure dissolution. (3) Subsequently, a certain amount (3–4 g) of glycerol, 0.4 wt% EDTA, 3×10^{-3} wt% Br-PADAP, and a quantity of the block copolymer PEA-b-PAA were added, followed by mechanical stirring for 30 minutes.

2.4 Characterization and Test Methods

- (1) **Contact Angle Measurement:** An appropriate amount of decontamination material was dropped onto a glass slide surface, and the contact angle between the droplet and the glass surface was recorded to evaluate wetting performance.
- (2) **Tensile Strength Testing:** Test specimens were prepared according to GB/T 1040.1-20 standard and tensile tests were conducted on a universal testing machine.
- (3) **Inductively Coupled Plasma (ICP) Spectroscopy:** This analysis determined the concentration of residual simulated contaminants after decontamination as part of the decontamination testing protocol.
- (4) **Color Difference Measurement:** Color changes of the cured strippable film before and after decontamination were analyzed using a colorimeter, with color difference values calculated through the Lab system.

3. Results and Discussion

3.1 Decontaminant Performance

2-(2-pyridylazo)-5-diethylaminophenol (PADAP) possesses a tridentate coordination site of [pyridine N]-[azo N]-[phenol hydroxyl O] that can strongly coordinate with uranium and cause a redshift in the maximum absorption of uranium complexes, thereby deepening the solution color. The aforementioned Br-PADAP introduces an electron-withdrawing bromine group at the fifth position of the pyridine ring. However, this indicator currently suffers from poor dispersibility and stability, tending to agglomerate in environmentally friendly water-based strippable decontamination materials and affecting colorimetric and decontamination effectiveness. This study selected an amphiphilic diblock copolymer PEA-b-PAA, prepared through RAFT one-pot polymerization of ethyl acrylate and acrylic acid monomers, as a dispersing aid for the indicator to solve the agglomeration problem.

To investigate the influence of the functional additive amphiphilic copolymer on material properties, dispersants prepared with different monomer ratios were added at 1% mass ratio to determine the optimal ratio for performance. Since the mechanical properties of the decontaminant during curing and drying fundamentally affect its usability, the optimal monomer ratio was identified through comprehensive comparison and analysis of the strippable coating's mechanical performance. Block copolymers with monomer ratios of 2:1, 1.5:1, and 1:1 were initially selected for 1 wt% addition, and the prepared strippable films were

subjected to tensile testing. As shown in Figure 2: see original paper, the experimental groups with indicator addition generally exhibited improved elongation at break and tensile strength compared to coatings without copolymer addition, with the 1.5:1 monomer ratio copolymer showing the greatest enhancement. Structural analysis of the synthesized indicator revealed that its macromolecular chains formed an appropriate crosslinking network with the substrate during film formation.

Temperature represents another critical factor affecting material performance during application. Therefore, the thermodynamic properties of the decontaminant were analyzed. [Figure 4: see original paper] presents the thermogravimetric curves of indicators with different monomer ratios. As shown in Figure 2: see original paper, the thermogravimetric curves showed no significant changes overall. However, due to the addition of copolymer resulting in trace ethanol in the coating, weight loss began below 100°C. The first mass loss step (50–180°C) corresponded to the removal of water, structural water, and inorganic solvents. Because these polymers are highly hydrophilic, broad melting peaks were observed. Following the melting peaks, two additional main peaks were associated with weight loss: the second stage (200–400°C) and third stage (400–450°C) represented the degradation of side groups (–OH) and structural decomposition of oligomers, respectively. The final weight loss in the 460–780°C range corresponded to polymer structure decomposition, consistent with the designed structure.

Based on the above characterization, the dispersant with a 1.5:1 monomer ratio was selected as the additive. To determine the optimal addition amount, its influence on comprehensive performance metrics including tensile strength, peel strength, and viscosity was investigated. Peel strength is crucial for ensuring effective contact between the cured decontaminant and object surface while enabling rapid peeling during removal. The value should not be too high, as excessive adhesion would prevent complete stripping, nor too low, as insufficient adhesion would make the coating susceptible to premature detachment. Therefore, based on the experimental data, addition levels of 2%, 4%, and 6% emerged as viable candidates. Lower viscosity facilitates decontaminant flow on object surfaces, enabling contamination zone wetting and pollutant encapsulation, which benefits both decontamination and pollutant response. However, excessively low viscosity hinders retention of active components on surfaces, affecting film thickness and consequently decontamination effectiveness. Thus, 2% and 4% addition levels were identified as candidate ratios. As shown in [Figure 4: see original paper], which displays tensile properties at different addition levels, the 4% addition level provided significant improvement to tensile performance while maintaining viscosity and peel strength within reasonable ranges compared to other formulations. Therefore, considering all factors, the optimal addition amount was determined to be 4%.

3.2 Investigation of Factors Affecting Decontaminant Stability

The prepared decontaminant primarily characterizes and marks pollutants through color changes resulting from complexation reactions with metal ions. During this process, the pH value of the environment affects the complexation reaction and consequently influences the indicator's colorimetric performance. The indicator exhibits significant color differences under acidic and alkaline conditions, primarily attributable to the acid effect. The acid effect refers to the phenomenon where the ability of ligands to participate in the main reaction decreases due to the presence of hydrogen ions in solution. In this case, both the phenolic hydroxyl coordination site of the indicator and the hydroxyl coordination sites of EDTA are affected by the acid effect. Specifically, in acidic environments, hydrogen ions interfere with complexation reactions between ligands and target ions, thereby affecting the indicator's colorimetric behavior. Therefore, the colorimetric performance was analyzed by examining its response under different pH values.

To avoid interference from other metal ions, ammonia solution was used to adjust the pH of the system. Experimental results demonstrated that upon pollutant addition, the solution color changed dramatically. Specifically, within the pH range of 6–12, the solution color changed rapidly, indicating immediate and strong impact on the acid-base balance. After 30 seconds of mixing, different pH ranges exhibited distinct color change characteristics. Under strongly acidic conditions (pH 1–2), no significant color change occurred. In solutions with pH 3–5, the orange color lightened to yellow-orange. When pH was between 6–10, the color changed from orange to purple-red. Under strongly alkaline conditions (pH 12), the color shifted from bright orange to deep purple. These pH-induced changes essentially reflect alterations in hydrogen ion concentration and solution acidity, which affect indicator dissociation through the acid effect.

As pH changes modify ionic equilibrium in solution, the complexation process between indicator and pollutants is affected. Strongly acidic environments inhibit the indicator's colorimetric reaction. In strongly alkaline conditions, complexation stability is impacted. Using trivalent cerium ions (Ce^{3+}) as an example, when Ce^{3+} from pollutants initially complexes with the indicator, color development occurs. Subsequently, Ce^{3+} reacts further with ammonia solution, eventually forming cerium hydroxide ($\text{Ce}(\text{OH})_3$) precipitate. This demonstrates that under strongly alkaline conditions, the indicator-pollutant complexation reaction is unstable, with final products precipitating from solution, resulting in progressively noticeable color changes.

3.3 Decontamination Performance Testing

A colorimetric decontaminant requires comprehensive evaluation of both decontamination and colorimetric performance. Therefore, based on colorimetric performance investigation, the decontamination performance under different experimental conditions was examined.

3.3.1 Decontamination Effectiveness on Different Object Surfaces

During nuclear facility decommissioning, most radioactive contaminants exist on floors and walls within structures. To investigate the decontamination performance of this strippable material in such scenarios, experiments were designed to assess its effectiveness on different object surfaces and roughness conditions.

Experiment 1 examined large-area decontamination on wood flooring, steel plate, and ceramic tile surfaces. The decontamination area for each substrate was set at 50 cm × 50 cm, with mixed large-particle sand used as contaminant particles. Decontamination effectiveness was evaluated by comparing weight changes before and after treatment. As shown in [Figure 5: see original paper], after curing, the decontaminant exhibited color differences between contaminated and uncontaminated regions, enabling characterization of contamination composition and scope through color changes. The decontaminant formed continuous films on all surfaces and could be completely stripped in one piece after curing, with no obvious particle residues remaining on the surfaces, demonstrating effective fixation of large-particle contaminants. [Figure 6: see original paper] shows that decontamination rates on all surfaces exceeded 99%, indicating excellent performance.

Experiment 2 investigated decontamination effectiveness on surfaces with different roughness levels. Different roughness surfaces were prepared by polishing glass with sandpaper of varying grits. The study examined decontaminant performance, colorimetric capabilities, and decontamination effectiveness on these surfaces. As shown in [Figure 7: see original paper], the decontaminant formed complete films on all roughness levels and could characterize surface contamination through color changes. Furthermore, [Figure 8: see original paper] demonstrates that decontamination rates for simulated pollutants exceeded 95% on all roughness levels. These results indicate that the decontaminant can efficiently remove loose contaminants and exhibits good adaptability across different material surfaces.

3.3.2 Simulated Radionuclide Decontamination ICP spectroscopy was used to measure potassium ion concentrations in: (C1) gauze soak solution after wiping contaminated surfaces, (C0) gauze soak solution after wiping uncontaminated blank surfaces, (C_{ash}) fallout ash soak solution, and (C_{water}) distilled water. Decontamination effectiveness was quantitatively assessed using these data with the following formula:

Decontamination rate () calculation formula:

$$K^+ = 1 - \eta = 1 - \frac{C1 - C0 - C \times 0.2L}{3.0mg \times 0.2L \times (M0 - M1) \times 1000}$$

Where: - M_K⁺ after decontamination = (C1 - C0 - C_{water}) × V (mass of K⁺ on sample after decontamination, in g) - M_K⁺ in fallout = (C_{ash} -

$C_{\text{water}} \times V$ (mass of K^+ applied to sample, in g) - V is soak solution volume (L) - C_1 is K^+ concentration in gauze soak solution after wiping contaminated surface (mg/L) - C_0 is K^+ concentration in gauze soak solution after wiping blank surface (mg/L) - C_{water} is K^+ concentration in distilled water (mg/L) - C_{ash} is K^+ concentration in fallout ash soak solution (mg/L) - M_1 is filter cloth mass after fallout application (g) - M_0 is filter cloth mass before fallout application (g)

[Figure 9: see original paper] and [Figure 10: see original paper] show the decontamination process and rates on different surfaces. The decontaminant achieved decontamination rates above 90% across all surfaces in three comparative experiments, demonstrating its capability for effective decontamination on various substrates. To further investigate performance, optical microscopy was used to analyze pollutant encapsulation and film formation on different surfaces. Microscopic images revealed that after curing, the decontaminant firmly fixed dust particles and formed relatively smooth films with uniform overall height. Maximum surface peak values among different components were approximately 500 nm. Color changes in the optical microscopy images corresponded to height variations, with continuous color transitions in depth maps indicating that the protruding regions corresponded to surface pollutants. The absence of abrupt color changes demonstrated continuous film formation with uniform thickness, ensuring uniform stress distribution during peeling and preventing fracture, thereby enabling decontamination through film transfer.

3.4 Colorimetric Performance Testing

This experiment prepared a decontaminant responsive to nuclide-contaminated environments. To deeply analyze the colorimetric tracing performance of the strippable decontamination material, a series of orthogonal decontamination experiments were designed and conducted based on decontamination performance evaluation.

The study systematically investigated the effects of different indicator addition amounts and pollutant concentrations on color development. Using orthogonal experimental design, optimal combinations of indicator dosage and pollutant concentration were efficiently screened to optimize colorimetric performance. During the experiment, real-time observation of color changes after liquid pollutant (uranyl ion standard solution) addition was performed, with dynamic changes recorded over 5 minutes. As shown in [Figure 12: see original paper], the decontaminant produced obvious color indication within 1 minute of pollutant addition, with color changes reaching a threshold indistinguishable to the naked eye under indoor lighting after 5 minutes. The material showed no color response to other metal ions, providing sensitive visual feedback for real-time decontamination monitoring.

To quantitatively evaluate colorimetric effectiveness, Lab color difference values were introduced as an assessment metric based on the CIE Lab color space,

widely used in color measurement and quality control. According to colorimeter measurements, color difference values greater than 5 are considered visually perceptible. However, in practical applications, color changes with values below 10 are often difficult to accurately distinguish. Therefore, this study adopted a color difference value of 10 as the standard for visually perceptible differences. Results are shown in [Figure 14: see original paper], Table 3-5, and Table 3-6. The data revealed that most color differences achieved perceptible levels under various pollutant and indicator addition amounts. The indicator's discrimination of pollutants showed significant numerical differences with increasing pollutant content. For example, with a median indicator addition of 0.002, the three pollutant content groups exhibited color difference values of 8.06, 31.85, and 56.08 after decontamination—a difference of approximately 20 between groups. This demonstrates that the prepared decontaminant can effectively characterize contamination levels through color changes. Meanwhile, different indicator content groups showed small inter-group differences (mostly below 5) across various pollutant concentrations, indicating that the response is not biased by indicator content and can effectively characterize pollutants.

Finally, to investigate adaptability on different surfaces, colorimetric performance was studied on various material surfaces and roughness conditions. The study examined color changes before and after decontaminant application on different interfaces and on custom-scratched surfaces simulating various roughness levels. As shown in [Figure 15: see original paper], obvious color changes were observed as the decontaminant interacted with pollutants. These changes resulted from complexation reactions between the indicator and ionic pollutants, demonstrating that the material can detect surface contaminant presence through color changes.

3.5 Analysis of Decontamination Process

Based on the above experiments, the colorimetric decontamination process was investigated. As illustrated, when the colorimetric agent component in the decontaminant binds with pollutants, a visible color change occurs, enabling characterization of surface contamination levels. Therefore, characterization of contact between the decontaminant and object surface, as well as between the cured decontaminant and pollutants, was necessary.

To deeply understand the decontamination mechanism and microscopic processes, scanning electron microscopy (SEM) was used to observe microstructural changes during actual decontamination. As shown in [Figure 18: see original paper], during film curing, pollutants were effectively encapsulated within the formed film structure, creating a microscopic “encapsulation” that separates contaminants from the polluted surface. The images clearly show the cured film's microstructure—dense and uniform, firmly encapsulating pollutant particles and preventing their redispersion or detachment during subsequent processes.

Optical microscopy was also employed to observe changes during decontami-

nation. As shown in [Figure 18: see original paper], similar to standard soil pollutant decontamination, particulate pollutants were effectively encapsulated within the film structure. The images demonstrate dense, uniform film morphology that firmly encapsulates pollutant particles. [Figure 19: see original paper] shows optical micrographs of the film surface and cross-section after decontamination on wood flooring, steel, and ceramic tile surfaces. The decontamination material formed complete films after curing and could be stripped intact from glass surfaces in one piece, proving its high efficiency in removing loose and large-area particulate contaminants while offering simple operation and easy cleanup in practical applications.

4. Conclusion

This paper investigated the colorimetric tracing performance and decontamination effectiveness of the prepared strippable decontamination material under various conditions. A series of dispersants with different monomer ratios was synthesized to study the influence of functional additives on decontaminant performance. The effects of different interfaces on usability, decontamination performance, and curing properties were discussed. Using optical microscopy combined with analysis of wettability and contact angle changes, as well as pollutant encapsulation after curing, the colorimetric mechanism was elucidated. The flow process was discussed, Lab color difference values were introduced for quantitative colorimetric performance evaluation, and the indicator's minimum detection limit was studied, providing important reference for material sensitivity at extremely low pollutant concentrations. Experimental results demonstrate that this material not only exhibits high adsorption capacity for uranium contamination but also visually reflects the decontamination process and effectiveness through color changes after uranium adsorption.

Author Contributions

HE Zhiyu, RAN Ke: Research design, data analysis/interpretation, and manuscript writing

ZHANG Ziyuan, CHEN Hongzhi: Research implementation and data collection

LI Yintao: Research funding acquisition and administrative, technical, or material support

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