Self-Healing Aroma-Schiff Base Functionalized Polyurethane for Scratch Resistant Flexible Fabrics

Conference: 13th AICLST, Kolkata, 6–8 March 2026

Paper reference: SP-02

Printed pages: 539–542

Source: *Leather 2050: Challenges & Opportunities*, ILTA (2026)

Self-Healing Aroma-Schiff Base Functionalized Polyurethane

for Scratch Resistant Flexible Fabrics

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Ref. No. SP-02

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Abstract

Developing aroma-infused, flexible, and self-healing polymers for durable and high-performance fabrics remains a complex and unmet challenge. In this study, we introduce a dynamic Schiff base-based aromatic self-healing system designed to improve the durability and mechanical resilience of flexible fabrics. The material is based on dynamic aromatic Schiff base linkages formed between cinnamaldehyde and an organosilane, integrated into a polyurethane matrix to produce a flexible functional coating. Self-healing arises from reversible imine bonds supported by hydrogen-bonding interactions between siloxane units and polyurethane chains. Under mild UV activation (365 nm), rapid healing occurs within 20 minutes while retaining a characteristic woody aroma. The film exhibited good mechanical performance, with 68% elongation in the undamaged state and an elongation recovery of 275% after healing, corresponding to a healing efficiency of 65%.

When applied by scalable dip and spray coating methods, the coating enhanced mechanical performance and damage resistance, with dip coating providing more uniform coverage and higher healing efficiency. Coated leather showed effective crack healing within 20 minutes, with 76% healing efficiency, and tear resistance of 39 N/mm after healing. Overall, this aroma-functionalized self-healing polyurethane demonstrates strong adhesion, rapid recovery, and

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sustained fragrance release making it a competent coating material for advanced textile and leather applications.

Keywords: Self-healing, synergistic healing, cinnamaldehyde, aroma-Schiff base, scratch resistance leather, self-healing fabrics.

1. Introduction

Self-healing materials have gained attention in recent years due to their ability to autonomously repair damage, significantly enhancing the durability and performance of coatings and substrates in a variety of application. These materials address common issues such as wear, tear, and microdamage that occur during regular use, ultimately extending the lifespan of products and reducing maintenance costs. By restoring mechanical and functional properties without external intervention, selfhealing materials represent a major milestone in material science, offering waste reduction benefits[1].

Self-healing processes are generally classified into two mechanisms: intrinsic and extrinsic [2,3]. Intrinsic selfhealing systems are governed by inherent material properties, where healing occurs through reversible chemical reactions or physical interactions within the polymer matrix. These mechanisms include hydrogen bonding, dynamic covalent bonds (e.g., disulfide and Schiff base), and reversible interactions that promote polymer chain mobility and restore structural integrity after damage

[4,5]. In contrast, extrinsic self-healing systems rely on external healing agents embedded in microcapsules, hollow fibers, or vascular networks, which are released upon damage to fill cracks or restore cohesion. Intrinsic systems are particularly suited for applications requiring flexibility and dynamic responsiveness, as they can autonomously heal under external stimuli such as temperature, light, pressure, and pH [6]. Among these, UV light–assisted intrinsic systems offer precise control over the healing process, making them highly suitable for protective coatings that require controlled and repeatable repair [7].

Polyurethanes (PURs) are among the most widely studied materials for self-healing, due to their versatile molecular structure and inherent phase-separated morphology. The soft/hard microphase separation in PURs contribute to their unique mechanical properties, making them adaptable for diverse applications[2]. Intrinsic self-healing in PURs is typically achieved by incorporating dynamic bonds, such as disulfide, Diels -Alder, hydrogen, or coordination bonds into the polymer network. PURs based on hydrogen bonds for self-healing, have attracted attention for their simple design and effective healing ability[3]. However, these systems often suffer from low mechanical strength, which limits their utility in fabric applications. Consequently, there is a growing need for dual-stimuli-responsive systems that combine multiple healing mechanisms to enhance both repairability and durability[4, 5]. Schiff bases, formed through dynamic imine bonds, exhibit reversible bond dissociation and reformation on exposure to UV light[6, 7]. These reversible bonds aid in the efficient and repeatable healing processes. However, existing Schiff base systems primarily operate as single-stimulusresponsive materials and lose their effectiveness in applications where the penetration of heat or light is restricted[8].

To address this, a dual-dynamic responsive system that combines hydrogen bonding with Schiff base imine bonding offers a more robust solution. In this system, hydrogen bonds provide immediate repair at the damaged site by

facilitating rapid chain mobility and cohesion. Subsequently, UV light activates the dynamic Schiff base bonds, enabling deeper structural restoration and mechanical reinforcement[9]. This combination addresses the challenge of achieving efficient healing while maintaining mechanical strength, making it suitable for fabric applications requiring both flexibility and durability. Although cinnamaldehyde has been studied for its use in self-healing materials, particularly in Diels-Alder-based systems, they are often limited by their slow healing rates, low efficiency, limited reusability, and dependence on high temperatures[10, 11]. Despite these challenges, cinnamaldehyde’s reactive aldehyde group makes it a suitable component for exploring Schiff base chemistry, which remains unexplored.

In this study, we develop a UV-responsive dynamic Schiff base-polyurethane system as an aromatic protective coating for fabric surfaces. By integrating cinnamaldehyde with APTES (3-aminopropyltriethoxysilane), we create an aroma-based Schiff base system that enables UVassisted self-healing. The silane functionality of APTES strengthens the organic-inorganic interface, enhancing adhesion and mechanical stability, while polyurethane provides a robust framework for crosslinking. The UVresponsive imine bonds facilitate reversible dissociation and reformation, enabling repeated healing cycles, overcoming the limitations of single- stimulus self-healing systems. Additionally, cinnamaldehyde imparts a lasting fragrance, enhancing the sensory appeal of the material. When applied as a protective fabric coating on leather and cotton fabrics, this system offers exceptional flexibility, durability, and mechanical strength, extending the lifespan of fabrics while maintaining aesthetic and functional integrity.

2. Experimental Methods

In ethanol, APTES and cinnamaldehyde were combined and stirred for 2 hours at 40OC until the formation of a Schiff base (SB). In tetrahydrofuran (THF), polyethylene glycol-300 (PEG-300) was added and the mixture was

maintained at 60OC, after which isophorone diisocyanate (IPDI) was introduced, followed by dibutyltin dilaurate (DBTDL). The reaction mixture was continuously stirred at 60OC for 5 hours until an increase in viscosity was observed. The resulting polyurethane prepolymer (PU) was then cooled to room temperature, and the Schiff base was gradually added. The mixture was further stirred for 1 hour at 40OC to promote the formation of functionalized polyurethane (PSB) containing dynamic covalent bonds.

3. Results and Discussion

This study presents a self-healing polyurethane based on a cinnamaldehyde–APTES aroma– Schiff base system, which enhances adhesion, mechanical properties, and fragrance. The Schiff base was incorporated into the polyurethane prepolymer through the reaction of free amine terminals with residual isocyanate groups, forming carbamate (urea) linkages and yielding Schiff base– functionalized polyurethane (PSB). The PSB was cast into a mold, solvent was evaporated at room temperature for 24 hours, and the material was subsequently cured at 80OC for 12 hours to achieve effective crosslinking.

Infrared spectroscopy confirmed Schiff base formation by the disappearance of the aldehyde C=O peak and the appearance of a new band at 1600–1650 cm-1, which was further supported by ^{13}C NMR analysis. Incorporation of the Schiff base into the polyurethane matrix was verified by ATR-IR, where the disappearance of the free isocyanate band at 2200–2300 cm-1 indicated complete consumption of isocyanate groups through reaction with Schiff base amines. Thermogravimetric analysis (TGA) of PSB revealed a two-step degradation process, indicating enhanced thermal resistance.

The self-healing behavior of the SB-functionalized polyurethane arises from the synergistic effects of hydrogen bonding and dynamic Schiff base imine bonds. Hydrogen bonding initiates healing immediately after damage through interactions between siloxane (Si–O–Si) groups and urea N–H groups, as well as between free

amine (Si–NH2) groups and polyurethane N–H groups, reinforcing structural integrity. Upon UV irradiation (365 nm), reversible dissociation and reformation of imine bonds occur, increasing molecular mobility and enabling polymer chain reorganization, which restores the network structure and facilitates efficient self-healing.

To evaluate the self-healing behavior of PSB, a crack was introduced on the surface of the PSB film and the sample was exposed to UV light (365 nm). Crack healing was monitored using an optical microscope at different time intervals, and 20 minutes of UV exposure resulted in significant crack reduction. The PSB film exhibited enhanced tensile strength and elongation at break compared to bare PU. After healing, the PSB-h film showed a remarkably improved elongation at break, attributed to effective crosslinking between neighboring polymer chains through imine bond realignment and network recovery. The healed PSB-h film achieved a healing efficiency of 76% within 20 minutes of UV exposure, which was further supported by SEM analysis.

PSB was also applied to resin-finished leather via a spraycoating technique. SEM images confirmed a smooth, uniform coating evenly distributed over the leather surface. The self- healing performance of the PSB-coated leather was subsequently evaluated, achieving 90% healing efficiency within 20 minutes, with residual marks visible only under microscopic observation. Additionally, the coated leather exhibited improved water-wetting resistance, indicating hydrophobic behavior. The silicareinforced polyurethane matrix further enhanced durability, abrasion resistance, water resistance, and mechanical resilience, ensuring long-term protection under varied environmental conditions.

4. Conclusion

This study successfully developed an aroma-based, synergistic self-healing polyurethane system incorporating a cinnamaldehyde-APTES Schiff base. The material synergistic healing mechanism driven by hydrogen bonding

interactions and UV-triggered dynamic imine bond crosslinking enabled rapid and efficient self-repair, restoring 76% of structural integrity within20 minutes. The Schiff base incorporated polyurethane matrix provided enhanced thermal stability, adhesion, and mechanical durability, while its aromatic Schiff base functionality introduced a subtle yet pleasant fragrance, improving the sensory appeal of coated surfaces. Further PSB coated leather exhibited 90% healing efficiency within 20 mins, effectively recovering tear resistance, flexibility, water resistance and surface integrity. The synergistic combination of self-healing and aroma functionality highlights its potential as an innovative, high-performance coating for soft surfaces, offering both extended durability and enhanced user experience. This approach paves the way for next-generation multifunctional materials that merge self-repair, sensory enhancement, and sustainable design for various advanced applications.

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