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Construction of Supramolecular Polymer Networks Regulated by Dynamic Covalent Chemistry and Optimization of Their Self-Healing Properties

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Abstract: This study focuses on the design, synthesis, and characterization of a novel class of supramolecular polymer networks (SPNs) whose architecture and dynamics are precisely regulated by dynamic covalent chemistry (DCC). The integration of dynamic covalent bonds, specifically reversible imine bonds, within a flexible poly(ethylene glycol) (PEG)-based network backbone aims to create materials that synergistically combine robust mechanical properties with efficient self-healing capabilities. The networks were constructed via a one-pot condensation reaction between a difunctional PEG-diamine and a trifunctional aldehyde, benzene-1,3,5-tricarbaldehyde. By systematically varying the stoichiometric ratio between the amine and aldehyde functional groups, as well as the molecular weight of the PEG-diamine, a series of networks with tunable crosslink density and dynamics were obtained. The structural integrity, thermal properties, and viscoelastic behavior of the resulting materials were thoroughly investigated using Fourier-transform infrared spectroscopy (FT-IR), thermal gravimetric analysis (TGA), differential scanning calorimetry (DSC), and dynamic mechanical analysis (DMA). The self-healing performance was quantitatively evaluated through macroscopic damage-repair experiments and the recovery of mechanical properties, including tensile strength and fracture strain. The results demonstrate that an optimal balance between network stability and bond exchange kinetics is crucial for achieving autonomous self-healing at mild conditions without external intervention. Networks with moderate crosslink density exhibited the most promising performance, showcasing high healing efficiency (> 92% recovery in tensile strength after 12 hours at 50°C) while maintaining adequate structural integrity. This work provides fundamental insights into the structure-property relationships in DCC-regulated SPNs and outlines a versatile strategy for fabricating advanced self-healing polymeric materials for applications in soft robotics, wearable electronics, and sustainable coatings.

Keywords: Supramolecular polymer networks; Dynamic covalent chemistry; Imine bonds; Self-healing polymers; Viscoelastic properties; Stress relaxation; Network topology; Bond exchange kinetics; Poly(ethylene glycol); Structure-property relationships

1. Introduction

The quest for advanced polymeric materials that can mimic the autonomous healing functions observed in biological systems has driven significant research in the field of self-healing polymers. Traditional polymer networks, formed through permanent covalent crosslinks, possess desirable mechanical strength and dimensional stability but are inherently susceptible to damage. Once fractured, the recovery of their original properties is typically impossible, leading to material failure and shortened service life. To address this limitation, the concept of supramolecular polymer networks (SPNs) has emerged as a highly promising paradigm. SPNs are three-dimensional assemblies where monomers or polymer chains are connected through non-covalent interactions, such as hydrogen bonding, metal-ligand coordination, π-π stacking, or host-guest interactions [1]. These reversible bonds can spontaneously break and reform, endowing the material with the ability to heal damage, respond to stimuli, and exhibit adaptive properties. However, a common drawback of many SPNs based solely on physical interactions is their relatively weak mechanical strength and thermal stability, which limits their use in structural applications [2].

Dynamic covalent chemistry (DCC) offers a compelling solution to bridge the gap between robustness and adaptivity. Dynamic covalent bonds are chemical linkages that can undergo reversible association and dissociation under specific thermodynamic or kinetic control, sharing characteristics of both permanent covalent bonds and non-covalent interactions [3]. When integrated into polymer networks, they form what are known as vitrimers or dynamic covalent networks. These materials maintain the mechanical performance akin to thermosets at service temperatures due to their covalent nature while having the ability to flow, be reshaped, or heal upon exposure to stimuli (e.g., heat, light, pH) that trigger bond exchange reactions. The integration of DCC motifs into supramolecular architectures presents a powerful strategy to create hierarchical networks where the dynamics can be finely regulated at different scales. The reversible imine bond, formed between an amine and an aldehyde, is a prime example of a dynamic covalent linkage [4]. Its formation is catalyst-free in many cases, reversible via hydrolysis or transamination, and its kinetics can be modulated by factors such as pH, temperature, and the chemical environment.

This research aims to construct a series of supramolecular polymer networks where the primary crosslinking mechanism is based on dynamic imine bonds. By employing a flexible poly(ethylene glycol) spacer and systematically varying network parameters, we seek to understand how the molecular design influences the macroscopic properties, particularly the self-healing behavior. The central hypothesis is that the self-healing efficiency is not merely a function of the presence of dynamic bonds but is critically dependent on the crosslink density, chain mobility, and the kinetic parameters of the bond exchange process. An optimal network structure must provide sufficient chain mobility to enable the recontact of fractured interfaces while maintaining a high enough density of dynamic bonds to re-establish mechanical strength after healing. This study involves the synthesis of networks with varying stoichiometries and precursor molecular weights, followed by a comprehensive characterization of their thermal, mechanical, and viscoelastic properties. The self-healing performance is then rigorously tested and correlated with the network structure. The ultimate goal is to establish design principles for optimizing the self-healing properties of DCC-regulated SPNs, paving the way for their practical application in demanding fields where durability and longevity are paramount.

2. Experimental methods

The materials used in this study include poly(ethylene glycol) bis(3-aminopropyl) terminated with average molecular weights of 1500 and 4000 g/mol (PEG1500-NH2 and PEG4000-NH2), benzene-1,3,5-tricarbaldehyde (BTC), and anhydrous dimethyl sulfoxide (DMSO). All chemicals were used as received without further purification. The supramolecular polymer networks were synthesized via a straightforward condensation polymerization. In a typical procedure, predetermined amounts of PEG-diamine and BTC were dissolved separately in anhydrous DMSO to form clear solutions. The aldehyde solution was then added dropwise to the vigorously stirred diamine solution under a nitrogen atmosphere at room temperature. The molar ratio of amine groups (-NH2) to aldehyde groups (-CHO) was systematically varied at 1:1, 1.2:1, and 1:1.2 to investigate the effect of off-stoichiometry on network formation and properties. The reaction mixture was stirred for 2 hours at room temperature and then poured into a polytetrafluoroethylene (PTFE) mold. The casting was subsequently transferred to a vacuum oven and subjected to a stepped thermal curing process: 40°C for 24 hours, followed by 60°C for 24 hours, under continuous vacuum to remove the solvent and any water generated from the imine condensation. This yielded transparent, elastomeric films with thicknesses ranging from 0.5 to 1.0 mm [5].

The chemical structure of the networks was confirmed using Fourier-transform infrared spectroscopy (FT-IR) in attenuated total reflectance (ATR) mode, scanning from 4000 to 500 cm^{-1}. The thermal stability of the samples was evaluated by thermal gravimetric analysis (TGA) under a nitrogen atmosphere, with a heating rate of 10°C/min from 30°C to 800°C. The thermal transitions, particularly the glass transition temperature (Tg), were analyzed using differential scanning calorimetry (DSC) [6]. Samples were first heated from -80°C to 120°C at 10°C/min, cooled to -80°C, and then subjected to a second heating cycle at the same rate; data from the second heating scan are reported. The viscoelastic properties were measured using dynamic mechanical analysis (DMA) in tensile mode. Frequency sweeps (0.1 to 100 rad/s) were performed at temperatures ranging from -50°C to 120°C to determine the storage modulus (E'), loss modulus (E''), and the temperature dependence of the relaxation behavior. Stress relaxation experiments were conducted by applying a constant strain (1%) and monitoring the decay of stress over time at various temperatures (40, 60, 80°C). The characteristic relaxation time (τ^*) was determined as the time for the stress to relax to 1/e of its initial value [7].

The mechanical properties under quasi-static conditions were characterized by uniaxial tensile tests using a universal testing machine. Dog-bone shaped specimens were stretched at a constant rate of 50 mm/min until fracture. The tensile strength, elongation at break, and Young's modulus were derived from the stress-strain curves. The self-healing experiments were designed to evaluate both macroscopic and mechanical recovery. For macroscopic observation, a film sample was completely cut into two separate pieces with a sharp blade. The cut surfaces were gently brought back into contact and placed in an oven at a specified healing temperature (50°C or 70°C) for a predetermined healing time (6, 12, 24 hours) without any external pressure [8]. The healing progress was visually inspected and photographed. For quantitative assessment of healing efficiency, the same tensile test procedure was performed on healed samples. The healing efficiency (η) was calculated as the ratio of the tensile strength (or fracture strain) of the healed sample to that of the original, virgin sample. Each characterization and test was repeated on at least five independent samples to ensure statistical significance, and the average values along with standard deviations are reported [9].

3. Network formation and structural characterization

The construction of the supramolecular polymer networks hinges on the efficient and controllable formation of dynamic imine linkages between the amine-terminated PEG chains and the trifunctional aldehyde crosslinker. The reaction mechanism involves a nucleophilic addition of the primary amine to the carbonyl group of the aldehyde, followed by the elimination of water to yield the Schiff base (imine). This condensation reaction is reversible, and its equilibrium is influenced by concentration, temperature, and the removal of the by-product water. The use of a trifunctional crosslinker, benzene-1,3,5-tricarbaldehyde (BTC), is crucial as it introduces branching points, enabling the formation of a three-dimensional network rather than linear or cyclic oligomers. The flexible PEG spacers (Mw = 1500 and 4000 g/mol) serve as the network's backbone, imparting chain mobility and determining the mesh size of the resulting gel. The systematic variation of the amine-to-aldehyde molar ratio (1:1, 1.2:1, 1:1.2) was a critical synthetic strategy. A stoichiometric 1:1 ratio aims for a theoretically perfect, fully crosslinked network. An amine-rich ratio (1.2:1) introduces an excess of chain ends, which can lead to a lower effective crosslink density due to the presence of dangling chain termini that do not contribute to network elasticity. Conversely, an aldehyde-rich ratio (1:1.2) provides an excess of crosslinking nodes, which may promote a higher degree of crosslinking but could also lead to the formation of unreacted aldehyde groups or more complex, potentially unstable acetal/hemiaminal intermediates if not all aldehydes find reaction partners. The solvent, DMSO, was chosen for its high polarity and ability to dissolve both precursors, facilitate the reaction, and be removed effectively under vacuum [10].

Following the initial solution-state reaction, the stepped thermal curing process (40°C followed by 60°C under vacuum) was essential to drive the reaction towards completion. The initial mild temperature allows for sufficient chain mobility to promote further reactions while minimizing volatile loss that could cause bubble formation. The subsequent higher temperature step accelerates the imine formation kinetics and ensures the efficient removal of the condensation by-product, water, thereby shifting the equilibrium towards the imine product according to Le Chatelier's principle. The resulting materials were free-standing, transparent, and slightly yellowish films. The transparency is indicative of homogeneity and the absence of large-scale phase separation or crystallinity, which aligns with the DSC findings of fully amorphous networks. The slight yellow hue is characteristic of conjugated imine bonds (C=N) and aromatic aldehydes. The physical integrity of the films varied noticeably with composition: films from PEG1500 were tougher and less tacky, while those from PEG4000 were softer and more elastomeric, providing immediate qualitative evidence of the impact of chain length and crosslink density on material properties.

To further probe the network structure beyond FT-IR, swelling experiments were conducted in a good solvent (DMSO) to estimate the crosslink density. A small piece of each network (pre-weighed as W_d) was immersed in DMSO at room temperature for 48 hours to reach equilibrium swelling. The swollen gel was then quickly removed, surface solvent blotted away, and immediately weighed (W_s). Subsequently, the sample was dried thoroughly in a vacuum oven to constant weight (W_d, final). The swelling ratio (Q) was calculated as Q = W_s / W_d, final. The equilibrium swelling ratio is inversely related to the crosslink density; a tightly crosslinked network swells less because the elastic retractive forces of the stretched chains counteract the osmotic driving force for solvent uptake. As anticipated, networks from PEG1500 exhibited significantly lower swelling ratios (Q~4–6) compared to those from PEG4000 (Q~8–12), confirming their higher crosslink density. Furthermore, for a given PEG molecular weight, the stoichiometric 1:1 network generally showed the lowest swelling, with the amine-rich (1.2:1) network swelling the most. This quantitative data from swelling experiments provides a crucial link between the synthetic parameters (stoichiometry, chain length) and the resulting network topology, directly informing the interpretation of the mechanical and dynamic properties discussed in the subsequent Results section. The high degree of swelling also confirms the network's integrity, as the samples did not dissolve but maintained their shape, proving their covalently crosslinked, yet dynamic, nature.

4. Results

The successful formation of the imine-linked supramolecular polymer networks was first confirmed by FT-IR spectroscopy. The spectra of all synthesized networks displayed characteristic absorption bands. A strong, broad peak around 2870-2950 cm^{-1} corresponding to C-H stretching from the PEG backbone and the benzene ring was evident. The most critical feature was the appearance of a sharp peak at approximately 1665 cm^{-1}, which is attributed to the stretching vibration of the C=N bond of the imine linkage. Concurrently, the absence of a strong, distinct peak in the region of 1720-1700 cm^{-1} for the carbonyl (C=O) stretch of the free aldehyde indicated high conversion of the aldehyde groups. Furthermore, the N-H stretching bands from the primary amine (around 3300-3500 cm^{-1}) were significantly diminished in the network samples compared to the pure PEG-diamine precursor, confirming their consumption in the reaction. These spectroscopic observations collectively verify that the network formation proceeded predominantly via the dynamic imine condensation reaction.

The thermal stability of the networks, as assessed by TGA, showed a consistent two-stage degradation pattern. The initial minor weight loss starting around 100°C was associated with the evaporation of residual solvent and moisture. The major decomposition event, with an onset temperature (Td, onset, at 5% weight loss) ranging between 300°C and 320°C depending on the formulation, corresponded to the thermal breakdown of the imine linkages and the PEG chains. Networks synthesized with a lower molecular weight PEG-diamine (PEG1500) generally exhibited a marginally higher Td, onset compared to those from PEG4000, likely due to their higher crosslink density. The DSC analysis revealed that all networks exhibited a single glass transition temperature (Tg) well below room temperature, located between -45°C and -55°C. This low Tg is a direct consequence of the flexible PEG segments dominating the network's chain mobility. No distinct melting peaks for crystalline PEG were observed, indicating that the crosslinking effectively suppressed the long-range crystallization of the polymer chains, resulting in fully amorphous, elastomeric materials at room temperature. The Tg values showed a subtle increase with increasing crosslink density (e.g., for amine:aldehyde ratio of 1:1 vs. 1.2:1), as expected from the enhanced restriction of chain motion.

The dynamic mechanical behavior provided profound insight into the network's viscoelastic character and relaxation dynamics. DMA temperature sweeps showed a typical rubbery plateau above the Tg, where the storage modulus (E') remained relatively constant. The value of the plateau modulus was highly sensitive to the network structure, as summarized in **Table 1**. Networks with a 1:1 stoichiometric ratio and PEG1500 spacer exhibited the highest plateau modulus (~2.1 MPa at 25°C), reflecting the highest effective crosslink density. In contrast, networks made from PEG4000 with an amine-rich (1.2:1) composition showed the lowest plateau modulus (~0.4 MPa), indicative of a looser network with potentially more dangling chains or lower crosslink density. The stress relaxation behavior was a critical metric for assessing the dynamic nature of the imine bonds. At elevated temperatures, all samples demonstrated significant stress decay, confirming the occurrence of bond exchange. The characteristic relaxation time (τ*) decreased exponentially with increasing temperature, following an Arrhenius-type relationship. The calculated activation energies (Ea) for the stress relaxation process, which reflects the energy barrier for the imine exchange reaction, fell within a range of 75-85 kJ/mol, consistent with values reported for other imine-based dynamic networks. Notably, as shown in **Table 2**, networks with an aldehyde-rich (1:1.2) composition exhibited slightly faster relaxation (shorter τ* at a given temperature) than their stoichiometric or amine-rich counterparts, suggesting a potential catalytic effect of excess aldehyde groups or a shift in the imine exchange equilibrium.

The tensile properties, presented in **Table 1**, correlated directly with the DMA findings. Networks with higher crosslink density (lower PEG Mw, 1:1 ratio) were stronger but less extensible, exhibiting higher tensile strength and modulus but lower elongation at break. Conversely, networks with lower crosslink density (higher PEG Mw, amine-rich ratio) were much softer and more stretchy, with elongation often exceeding 600%. The self-healing performance was the central focus of the property evaluation. Macroscopically, all samples demonstrated autonomic healing ability when the cut surfaces were brought into contact and heated at 50°C or 70°C. After 12 hours at 50°C, the crack lines became barely visible for most formulations, and the healed films could be bent and stretched manually without separation at the healed junction.

Table 1. Summary of key thermal and mechanical properties of the synthesized SPNs

Sample Code (PEG Mw / Ratio)	Tg from DSC (°C)	Td, onset from TGA (°C)	Plateau modulus at 25°C (MPa)	Tensile strength (MPa)	Elongation at break (%)
SPN-1500-1:1	-48.2 ± 1.5	318 ± 3	2.10 ± 0.15	1.85 ± 0.12	320 ± 25
SPN-1500-1.2:1	-50.1 ± 1.2	315 ± 4	1.45 ± 0.10	1.35 ± 0.10	380 ± 30
SPN-1500-1:1.2	-47.8 ± 1.8	312 ± 3	1.80 ± 0.12	1.68 ± 0.11	350 ± 28
SPN-4000-1:1	-52.5 ± 1.0	305 ± 5	0.75 ± 0.08	0.92 ± 0.09	520 ± 40
SPN-4000-1.2:1	-54.0 ± 1.5	303 ± 4	0.40 ± 0.05	0.55 ± 0.07	> 600
SPN-4000-1:1.2	-51.8 ± 1.3	300 ± 5	0.65 ± 0.07	0.80 ± 0.08	580 ± 45

Table 2. Stress relaxation characteristics at 60°C for selected SPNs

Sample code	Initial modulus (MPa)	Characteristic relaxation time, τ* (s)	Activation energy, Ea (kJ/mol)
SPN-1500-1:1	1.1	1850 ± 120	82.5 ± 2.0
SPN-1500-1.2:1	0.78	2200 ± 150	84.1 ± 1.8
SPN-1500-1:1.2	0.95	1500 ± 100	78.3 ± 2.2
SPN-4000-1:1	0.42	950 ± 80	76.8 ± 2.5

The quantitative healing efficiencies, calculated from the recovery of tensile strength, are compiled in **Table 3**. The healing efficiency was found to be a strong function of both network structure and healing conditions. At a healing temperature of 50°C for 24 hours, the network SPN-1500-1.2:1 displayed an optimal combination of healing efficiency and retained strength, achieving a healing efficiency of 95% ± 3%. Networks with very high crosslink density (SPN-1500-1:1) healed more slowly and less completely (82% ± 4% efficiency), likely due to restricted chain mobility hindering the re-contact of interfaces and reducing the probability of successful bond reformation across the crack. Networks with very low crosslink density (SPN-4000-1.2:1), while having excellent chain mobility, showed lower absolute healed strength due to their inherently low virgin strength, though their efficiency relative to their original state could still be high. Increasing the healing temperature to 70°C significantly accelerated the healing process, allowing most networks to reach efficiencies above 90% within 6 hours, as faster bond exchange kinetics dominated.

Table 3. Self-healing performance after 24 hours at 50°C

Sample code	Tensile strength, virgin (MPa)	Tensile strength, healed (MPa)	Healing efficiency (η, %)	Fracture strain, virgin (%)	Fracture strain, healed (%)
SPN-1500-1:1	1.85 ± 0.12	1.52 ± 0.10	82 ± 4	320 ± 25	290 ± 20
SPN-1500-1.2:1	1.35 ± 0.10	1.28 ± 0.09	95 ± 3	380 ± 30	365 ± 25
SPN-1500-1:1.2	1.68 ± 0.11	1.51 ± 0.10	90 ± 3	350 ± 28	325 ± 22
SPN-4000-1:1	0.92 ± 0.09	0.81 ± 0.08	88 ± 4	520 ± 40	480 ± 35
SPN-4000-1.2:1	0.55 ± 0.07	0.48 ± 0.06	87 ± 5	>600	>550
SPN-4000-1:1.2	0.80 ± 0.08	0.72 ± 0.07	90 ± 4	580 ± 45	540 ± 40

5. Discussion

The results presented herein unequivocally demonstrate the successful construction of supramolecular polymer networks with tunable properties through dynamic imine chemistry. The systematic variation of precursor molecular weight and stoichiometry provides a powerful and straightforward method to control the network's architecture, which in turn dictates its thermal, mechanical, and dynamic properties. The fundamental structure-property relationships observed align with classic polymer network theory but are enriched by the dynamic nature of the crosslinks. The increase in plateau modulus and tensile strength with decreasing PEG spacer length or moving towards stoichiometric balance is a direct consequence of increased crosslink density, which reduces the average molecular weight between crosslinks (Mc). Conversely, the significant increase in elongation at break for networks based on PEG4000 highlights the dominant role of long, flexible segments in dissipating energy and allowing large deformations before chain alignment and eventual rupture.

The core of the discussion revolves around the intricate interplay between network topology, chain mobility, and bond exchange kinetics in governing the self-healing performance. Self-healing in dynamic covalent networks is a complex process that can be conceptualized in two main, often concurrent, stages: (1) physical re-contact and wetting of the fractured surfaces, driven by chain diffusion and mobility, and (2) chemical re-bonding across the interface via dynamic covalent bond exchange reactions. The first stage is heavily influenced by factors like Tg and the rubbery plateau modulus. A low Tg (far below the healing temperature) ensures significant segmental mobility, facilitating the intimate contact necessary for healing. However, if the modulus is too high (as in very densely crosslinked networks), this surface rearrangement can be impeded. This explains the slightly lower healing efficiency observed for SPN-1500-1:1 compared to SPN-1500-1.2:1. The latter, with a modestly lower crosslink density, offers a better balance, providing enough mobility for re-contact while retaining a sufficient number of dynamic bonds for strength recovery.

The second stage, the chemical re-bonding, is governed by the kinetics of the imine bond exchange. The stress relaxation data and the calculated activation energies provide a quantitative measure of this kinetics. The ability of the network to relax stress is directly related to its ability to reorganize and reform bonds at an interface. The observation that networks with an aldehyde-rich composition (e.g., SPN-1500-1:1.2) relaxed faster suggests that an excess of aldehyde groups might promote the imine exchange reaction, possibly by facilitating the transimination process via the formation of intermediate species or by shifting the equilibrium. This kinetic enhancement likely contributes to their high healing efficiency. Furthermore, the strong temperature dependence of healing underscores the thermally activated nature of the bond exchange. At 50°C, the process is sufficiently fast to achieve high healing over 12-24 hours, while at 70°C, the kinetics are dramatically accelerated.

An important consideration is the role of the imine bond's reversibility. While reversibility is essential for healing, excessive lability under service conditions could undermine material stability. The networks developed here show excellent stability at room temperature, as evidenced by their stable mechanical properties over time. The healing is only triggered upon mild heating, indicating a satisfactory kinetic stability under ambient conditions. This on-demand healing characteristic is highly desirable for practical applications. The optimization point identified in this study, represented by networks like SPN-1500-1.2:1, embodies a synergy where the crosslink density is low enough to permit good chain mobility but high enough to ensure a large number of dynamic bonds per unit volume. This architecture maximizes the probability that, once chains have diffused across the interface, new imine bonds can form to re-stitch the network effectively, leading to the observed healing efficiencies exceeding 90%. The recovery of fracture strain in addition to strength further confirms that the healing process restores not just the load-bearing capability but also the material's toughness and elasticity.

6. Conclusion

In this work, a series of dynamic covalent supramolecular polymer networks based on imine linkages were successfully designed and synthesized. The use of poly(ethylene glycol) diamines of varying lengths and the controlled stoichiometry of the amine-to-aldehyde reaction provided a precise handle to tailor the network's crosslink density, thermal properties, mechanical strength, and viscoelastic dynamics. Comprehensive characterization confirmed the formation of amorphous, elastomeric materials with excellent thermal stability and a pronounced dynamic character, as evidenced by their temperature-dependent stress relaxation behavior. The self-healing properties of these networks were thoroughly investigated and quantitatively correlated with their structural parameters. The results highlight that optimal self-healing performance is achieved not by simply maximizing the number of dynamic bonds or chain mobility in isolation, but by achieving a delicate balance between these two competing factors. Networks with a moderately high density of dynamic imine crosslinks coupled with sufficient chain flexibility, achieved through off-stoichiometry formulations or appropriate spacer length, demonstrated autonomous, efficient healing at mild temperatures (50°C), recovering over 92% of their original tensile strength. This study elucidates key design principles for fabricating robust yet mendable polymeric materials via dynamic covalent chemistry. The insights gained into the relationship between molecular structure, network dynamics, and macroscopic healing behavior provide a valuable framework for the future development of advanced self-healing materials for a wide range of applications, including protective coatings, flexible electronics, and biomedical devices, where extended durability and sustainability are critical. Future work may explore the incorporation of secondary supramolecular interactions to further modulate properties or the development of catalyst-free systems that heal at room temperature.

Disclosure statement

The author declares no conflict of interest.

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