

Alginate-Acrylic Hydrogels with Autonomous Self-Healing: A Review of Design Principles and Mechanisms.

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Abstract

A key component in creating high-performance, sustainable materials is the combination of biopolymers with synthetic networks. Alginate-acrylic hybrid hydrogels with self-healing properties are thoroughly examined in this review. These semi-synthetic systems, in contrast to traditional hydrogels, greatly increase their functional lifespan in demanding applications by using dynamic non-covalent interactions to repair structural integrity after mechanical damage. We draw attention to their dual-use in (i) environmental remediation, where they serve as regenerable adsorbents for the removal of organic and heavy metal pollutants, and (ii) sustainable agriculture, where they serve as controlled-release fertilizer carriers and stimuli-responsive soil conditioners. Through repeated cycles of dehydration and rehydration, these materials improve soil aeration, increase water retention, and reduce nutrient leaching, all of which are critically inspected in this review. Finally, we suggest a roadmap for future research, focusing on the standardization of healing protocols and the development of energy-efficient, green synthesis routes to facilitate large-scale industrial adoption.

Keywords Self-Healing Hydrogels, Alginate, Acrylic Polymers, Semi-Synthetic Materials, Water Treatment, Sustainable Agriculture, Controlled-Release Fertilizers

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1. Introduction

Hydrogels, three-dimensional hydrophilic polymer networks, have emerged as pivotal materials in advancing sustainable technologies for water management and agriculture [1, 2, 3]. Among them, self-healing hydrogels (SHHs) represent a transformative class of materials capable of autonomously repairing mechanical damage, thereby extending their functional lifespan and reliability in dynamic environments [4, 5, 6]. This intrinsic repair capability is primarily governed by dynamic reversible bonds such as ionic interactions, hydrogen bonds, and dynamic covalent bonds, which enable the restoration of structural integrity after fracture [7, 8, 9]. Recent research has strategically focused on combining natural and synthetic polymers to create hybrid hydrogels that synergize the advantages of both components [10, 11, 12]. Natural polymers, like alginate (see Scheme 1) derived from marine algae brown seaweed, offer exceptional biocompatibility, biodegradability, and eco-friendliness, along with ion-responsive gelation properties [13, 14, 15]. In contrast, synthetic acrylic polymers (e.g., polyacrylic acid, Scheme 2) provide mechanical robustness, high water absorbency, and tunable physicochemical properties [16, 17, 18]. The integration of alginate with acrylic polymers results in semi-synthetic networks that exhibit not only enhanced mechanical performance but also inherent self-healing functionality [19, 20, 21]. While numerous reviews cover self-healing polymers or alginate-based materials broadly, a dedicated, critical synthesis focusing specifically on the design principles, performance trade-offs, and application-driven optimization of alginate-acrylic hybrid hydrogels is lacking. This review aims to fill that gap by moving beyond general descriptions to a detailed analysis of how molecular interactions translate to macroscale functionality in environmentally relevant settings. Furthermore, the application potential of these hybrid systems in environmental remediation (e.g., water purification via pollutant adsorption) and sustainable agriculture (e.g., as superabsorbent soil conditioners and controlled-release fertilizer carriers) warrants a focused and updated evaluation [22, 23, 24]. Therefore, this review aims to critically examine the current state of knowledge regarding alginate-acrylic hybrid hydrogels with autonomous self-healing properties. Specifically, it will:

- i. Analyse the design and synthesis strategies for creating these hybrid networks.
- ii. Elucidate the molecular and supramolecular mechanisms underpinning their autonomous self-healing behaviour.
- iii. Evaluate their key functional properties, including swelling, mechanical strength, and stimuli-responsiveness.
- iv. Assess their application potential in water treatment and smart agriculture, with an emphasis on controlled-release systems.
- v. Identify current challenges and future research directions to optimize their performance and scalability.

By consolidating recent advances (primarily from the past five years) and providing a mechanistic understanding, this review seeks to serve as a foundational resource for researchers developing next-generation, durable, and environmentally adaptive hydrogel materials [25, 26, 27].

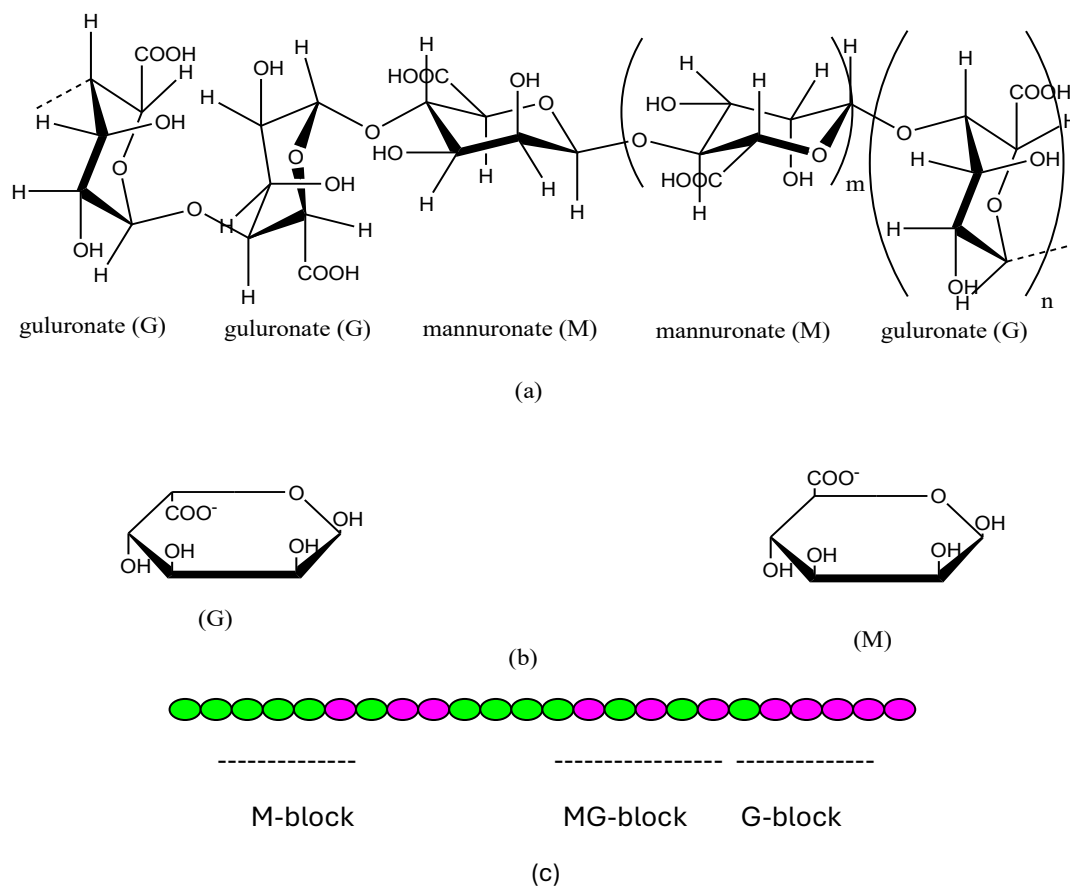
2. Alginate-Acrylic Hybrid Hydrogels: Synthesis, Mechanisms, and Functional Properties

2.1. Synthesis and Network Design:

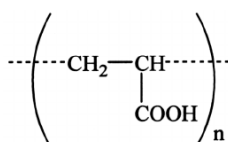
The fabrication of alginate-acrylic hydrogels with self-healing properties typically exploits a dual-network strategy [28, 29, 30]. This strategy synergistically combines the ionotropic gelation of alginate (see molecular structure in schemes 1 and 2) and the free-radical polymerization of acrylic monomers [31, 32, 33]. The main synthesis pathways, along with their comparative advantages and limitations, are summarized in Table 1.

The selection among synthesis methods represents a direct trade-off between simplicity and precise control [51]. The one-pot approach is favored for its efficiency and its ability to create intimately mixed networks that facilitate dynamic bonding [52, 53]. In contrast, the sequential method offers superior tunability, enabling researchers to first establish a robust PAA scaffold before incorporating reversible ionic crosslinks, which is advantageous for achieving targeted mechanical properties [54, 55]. While grafting provides a distinct pathway for molecular-level integration, it may demand greater synthetic expertise [56]. Ultimately, the optimal pathway is fundamentally determined by the target application's (e.g., in water purification for heavy metal adsorption or in agriculture for slow-release fertilizers) requirements for mechanical strength, swelling kinetics, and healing speed [57, 58, 59]. Overall, the choice of synthesis method

directly and critically influences the hydrogel's architecture, swelling behavior, and self-healing efficiency [60, 61].



Scheme 1. (a) Compositional structure of alginic acid, (b) structures of the α -L-gulonate (G) and β -D-mannuronate (M) residues of which it is composed, and (c) schematic representation of the residue sequence



Scheme 2. Molecular structure of poly(acrylic acid)

2.2. Mechanisms of Autonomous Self-Healing

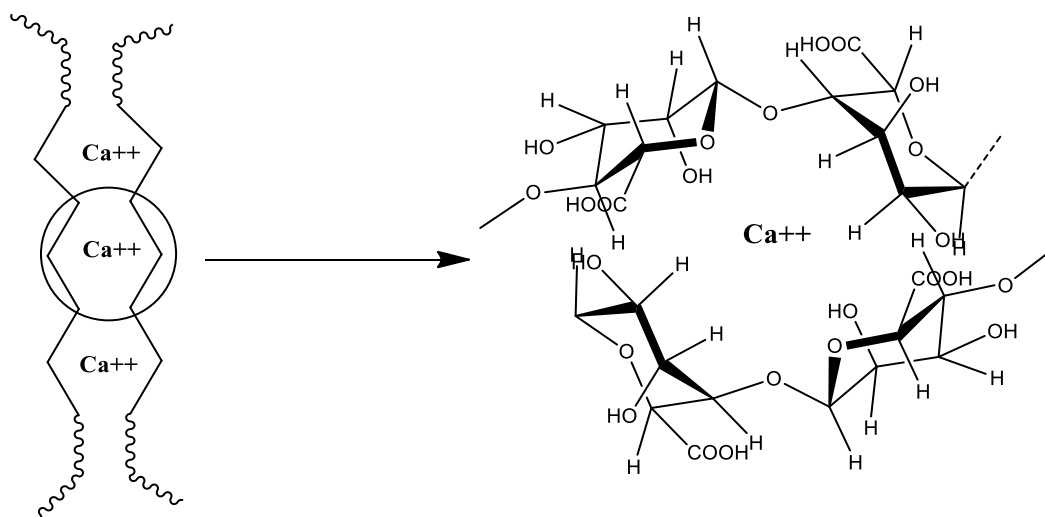
The autonomous self-healing capability in alginate-acrylic hydrogels at room temperature is attributed to the synergistic action of multiple dynamic, non-covalent interactions that can spontaneously reform after damage. This capability primarily relies on three interlinked mechanisms operating in concert: ionic coordination, hydrogen bonding, and polymer chain interdiffusion and entanglement [62, 63, 64].

Table 1. Critical comparison of synthesis methods for alginate-acrylic hydrogels.

Synthesis Method	Key Description	Advantages	Disadvantages
One-Pot, Simultaneous Network Formation	Sodium alginate, acrylic acid monomer, an initiator (ammonium persulfate, APS), and a covalent crosslinker (N,N'-methylenebisacrylamide, MBA) are mixed. Divalent cations (such as calcium ions, Ca^{2+}) are then added to trigger ionic gelation concurrently with free-radical polymerization.	Simplicity and speed [34]. Promotes homogeneous interpenetration of networks from the outset [35]. High potential for creating dense, cooperative interactions [36].	Difficult to independently control the kinetics and density of each network [37]. Risk of phase separation if reaction conditions are not optimized [38, 39].
Sequential Network Formation	1. A covalently crosslinked PAA network is synthesized first. 2. The pre-formed network is soaked in an alginate solution. 3. Ionic crosslinking (e.g., with Ca^{2+}) is performed.	Allows precise, independent control over each network's properties (e.g., crosslink density of PAA) [40]. Ensures a well-defined, interpenetrating structure [41, 42].	More time-consuming and involves multiple steps [43]. Achieving deep and uniform penetration of alginate into the pre-formed PAA network can be challenging [44, 45].
Graft Copolymerization	Acrylic monomers are polymerized directly from active sites on the alginate backbone, creating covalent grafts. The graft copolymer can then be ionically crosslinked.	Creates strong covalent linkages between components, potentially enhancing mechanical integrity [46]. Unique architecture with alginate as a backbone [47].	Synthesis can be complex [48]. May limit the mobility of alginate chains, potentially impairing self-healing driven by chain diffusion [49, 50].

2.2.1. Ionic Coordination

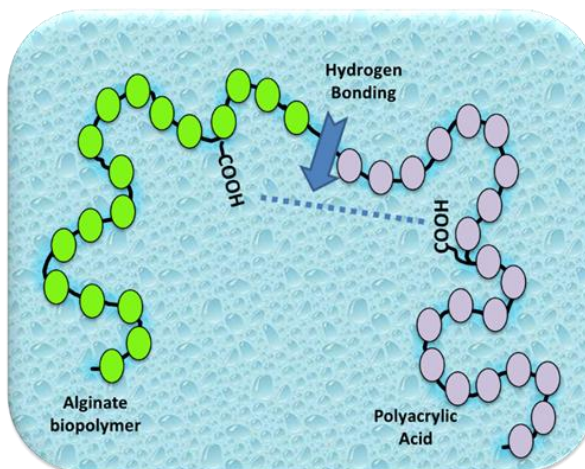
The primary reversible bonds are the ionic crosslinks between alginate's carboxylate groups and divalent cations (e.g., Ca^{2+}) [65]. Upon fracture, these coordinate bonds can break. However, in the hydrated state, the mobility of polymer chains and free ions allows for the diffusion of alginate chains and cations across the cut interface [66, 67]. Given time and contact, the egg-box complexes can re-form, re-establishing the ionic network (in scheme 3) [68, 69, 70]. The kinetics of this ion exchange and re-coordination are critical for healing speed [71].

Scheme 3. Ionic Crosslinking and Re-coordination of alginate via Ca^{2+} Complexes

2.2.2. Hydrogen Bonding

Both alginate and PAA are rich in carboxyl ($-\text{COOH}$) and hydroxyl ($-\text{OH}$) groups [72]. These functional groups form an extensive, cooperative network of hydrogen bonds (see scheme 4) throughout the hydrogel matrix

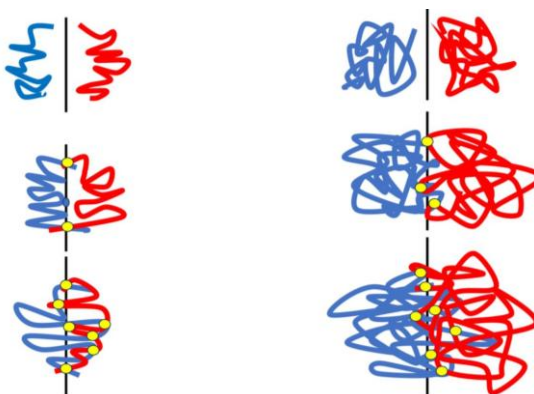
[73, 74]. Hydrogen bonds are dynamic; they can break under stress and readily re-form when the damaged surfaces are brought back into contact [75, 76]. This mechanism is particularly effective in contributing to the initial stages of adhesion and healing [77, 78]. The role of hydrogen bond density and cooperativity has been extensively studied [79].



Scheme 4. Schematic of the reversible hydrogen-bonding network between alginate and PAA chains, facilitating self-healing

2.2.3. Chain Interdiffusion and Entanglement

The viscoelastic nature of the swollen polymer network allows for the interdiffusion of polymer chains across the fracture surface upon contact (see the diffusion process in scheme 5) [80, 81]. This physical entanglement, coupled with the reformation of ionic and hydrogen bonds, leads to the gradual recovery of mechanical strength [82, 83, 84]. Chain mobility, governed by water content and crosslink density, is a key parameter [85]. The healing efficiency, often quantified as the percentage recovery of tensile strength, elongation, or toughness, in these systems, can exceed 90% without any external stimulus, which is a key advantage for practical applications [86, 87, 88]. The healing kinetics and final strength depend on factors such as the density of reversible bonds, water content (plasticizing effect), polymer chain length, and mobility [89, 90, 91].



Scheme 5. Schematic diagram of the diffusion process at the polymer/polymer interface (a process of entanglement establishment)

2.3. Key Functional Properties

The hybrid nature of these hydrogels results in a set of key functional properties that are both tunable and deeply interconnected. These properties, swelling and water retention, mechanical robustness, and stimuli-responsiveness collectively determine the material's performance and suitability for specific applications. Each of these critical attributes is explored in detail below.

2.3.1. Swelling and Water Retention

These hydrogels exhibit significant pH-dependent swelling behavior due to the ionization of carboxylic groups on PAA and alg [92, 93, 94]. At low pH, the -COOH groups are protonated, leading to reduced electrostatic repulsion and lower swelling. As pH increases, the groups deprotonate to -COO⁻, increasing repulsion and water uptake dramatically [95, 96]. This property is crucial for applications like controlled release, where release rates can be modulated by environmental pH [97, 98, 99].

2.3.2. Mechanical Robustness

The dual-network structure often results in a favorable balance of strength and elasticity [100, 101]. The alginate network provides elasticity and toughness through reversible ionic crosslinks, while the covalently crosslinked PAA network contributes to mechanical rigidity and load-bearing capacity [102]. Properties like compressive modulus, tensile strength, and elongation at break can be tailored by varying the polymer ratios and crosslinking densities [103].

2.3.3. Stimuli-Responsiveness

Beyond pH, the hydrogels are sensitive to ionic strength [104]. The presence of monovalent ions (e.g., Na⁺, K⁺) can exchange with Ca²⁺ in the egg-box structures, potentially weakening the network and triggering swelling or degradation, which can be used to control the release of encapsulated agents [105]. Recent formulations also explore temperature or redox responsiveness by incorporating additional functional monomers [106].

2.4. Applications in Environmental and Agricultural Sectors

Alginate-acrylic hydrogels are promising sustainable materials due to their synergistic combination of autonomous self-healing, high absorbency, and stimuli-responsiveness. This multifunctionality ensures durability, efficiency, and intelligent operation in dynamic applications, particularly in environmental remediation and smart agriculture. Their growing potential is reflected in increasing research and patent activity [107].

2.4.1. Water Purification

The anionic nature of the hydrogel network (from alginate and PAA) provides active sites for adsorbing cationic pollutants such as heavy metal ions (Pb²⁺, Cd²⁺, Cu²⁺) and organic dyes through electrostatic interactions and complexation [108]. The self-healing property ensures that the adsorbent material can withstand physical wear and tear in dynamic water treatment systems, potentially extending its operational life and reducing maintenance needs [109, 110]. For instance, recent work demonstrated a self-healing alginate-PAA hydrogel bead that could be reused for multiple adsorption-desorption cycles for methylene blue removal with minimal capacity loss [111]. Field-scale studies are beginning to validate these laboratory findings [112].

2.4.2. Sustainable Agriculture

2.4.2.1 Water Reservoir

When incorporated into soil, these hydrogels absorb and retain large volumes of water during irrigation or rainfall, then release it slowly during dry periods [113]. This reduces water stress on plants and decreases irrigation frequency, which is vital in arid regions [114]. The self-healing feature is critical here, as it allows the hydrogel to recover from compression and dehydration-rehydration cycles in the soil matrix, maintaining its water-holding structure over time [115, 116, 117].

2.4.2.2 Controlled-Release Fertilizer (CRF) Carriers

Nutrients (e.g., N, P, K) or micronutrients (e.g., Fe, Cu, Mn) can be loaded into the hydrogel matrix [118]. Their release is controlled by diffusion through the swollen network and can be triggered by changes in soil moisture, pH shifts caused by root exudates, or microbial activity [119]. This targeted delivery improves nutrient use efficiency, minimizes leaching losses, and reduces environmental pollution from fertilizer runoff

[120]. Patents have been filed for such intelligent delivery systems, highlighting their commercial potential [121]. Recent work integrates sensors for real-time nutrient monitoring [122].

2.4.2.3 Soil Structure Improvement

Hydrogels significantly enhance soil quality through multiple interconnected mechanisms. Their ability to undergo repeated swelling and de-swelling cycles improves soil aeration and porosity by creating and stabilizing macropores within the soil matrix [123]. This improved structure facilitates better gas exchange, enhances water infiltration, and reduces soil compaction, thereby promoting healthier and more extensive root growth [124]. Furthermore, the improved moisture retention and porous habitat foster increased microbial activity, enriching the soil microbiome and accelerating nutrient cycling processes essential for plant health [125]. In addition to modifying soil structure, hydrogels play a crucial role in soil aggregation and erosion control. The hydrogel particles act as binding agents, adhering to soil particles and promoting the formation of stable soil aggregates [126]. This aggregation enhances soil cohesion, increases surface roughness, and improves resistance to both water and wind erosion. By stabilizing the topsoil, hydrogels help prevent nutrient leaching and loss of fertile soil, contributing to sustainable land management. The longevity and stability of these benefits are often enhanced in self-healing hydrogels, which can maintain their structural integrity and functionality through repeated environmental stresses [127].

3. Conclusions and Future Perspectives

3.1. Conclusions

This review has critically examined the development, mechanisms, and applications of semi-synthetic hydrogels based on alginate and acrylic polymers with autonomous self-healing capabilities. These hybrid systems successfully combine the biocompatibility, biodegradability, and ion-responsive gelation of natural alginate with the mechanical robustness, high water absorbency, and tunability of synthetic acrylic polymers. The autonomous self-healing property, primarily driven by dynamic reversible bonds including ionic coordination, hydrogen bonding, and, in advanced formulations, dynamic covalent bonds, enables the restoration of structural integrity after damage, significantly extending the functional lifespan of the material. Key design principles involve tailoring the alginate-to-acrylic polymer ratio, selecting appropriate crosslinking strategies (dual ionic-covalent networks), and controlling the polymerization conditions to optimize swelling behavior, mechanical strength, and self-healing efficiency. The resulting hydrogels exhibit stimuli-responsive behavior (especially to pH and ionic strength), making them "smart" materials for advanced applications. In environmental remediation, these hydrogels show promise for water purification through the adsorption of pollutants and heavy metals. In sustainable agriculture, they function as superabsorbent soil conditioners and controlled-release carriers for water and nutrients, potentially reducing irrigation frequency and fertilizer leaching. Their self-healing nature ensures durability under cyclic hydration and mechanical stress in soil.

3.2. Future Perspectives

Despite significant progress, several challenges and opportunities for future research remain:

- i. **Healing under Complex Conditions:** Most studies demonstrate healing under ideal, laboratory conditions. Future work should quantify healing efficiency under realistic environmental stresses (e.g., varying soil pH, microbial activity, freeze-thaw cycles).
- ii. **Mechanistic Depth:** Advanced in situ characterization techniques, such as in-situ FTIR, Raman spectroscopy, or small-angle X-ray scattering (SAXS), should be employed to observe bond reformation dynamics in real-time during the healing process. Fundamental studies on alginate interactions with various cations are needed to optimize healing efficiency.
- iii. **Multi-Stimuli Responsiveness:** Designing hydrogels that respond to multiple triggers (e.g., pH, temperature, enzymes, or redox potential) could enable more precise control over healing and release profiles for targeted applications. Research should explore alginate derivatives with enhanced functionality, guided by previous work on natural polymer modifications.
- iv. **Scalability and Eco-Footprint:** Developing greener synthesis routes using water-based systems, renewable initiators, and energy-efficient processes is crucial for large-scale, cost-effective production.

The end-of-life fate of these semi-synthetic materials, including their complete biodegradation pathways, requires thorough investigation.

- v. Expanded Application Horizons: Beyond agriculture and water treatment, these hydrogels hold potential in biomedical engineering (e.g., self-healing wound dressings, drug-eluting implants) and soft robotics (durable, hydratable actuators). Exploration into loading and controlled release of micronutrients, pesticides, or beneficial microbes for precision agriculture represents a promising frontier. Alginate-based delivery systems show particular promise for controlled release applications.
- vi. Standardization of Evaluation: The field would benefit from standardized protocols for reporting self-healing efficiency (e.g., recovery of tensile strength, toughness) and release kinetics to allow direct comparison between different material systems.

By addressing these challenges, the next generation of alginate-acrylic self-healing hydrogels can transition from promising laboratory prototypes to robust, sustainable, and intelligent materials capable of addressing critical global challenges in environmental sustainability and food security.

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