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Expanding the Frontiers of Flame-Retardant Hydrogels: Recent Advances in Applications, Design Strategies, and Multifunctional Performance

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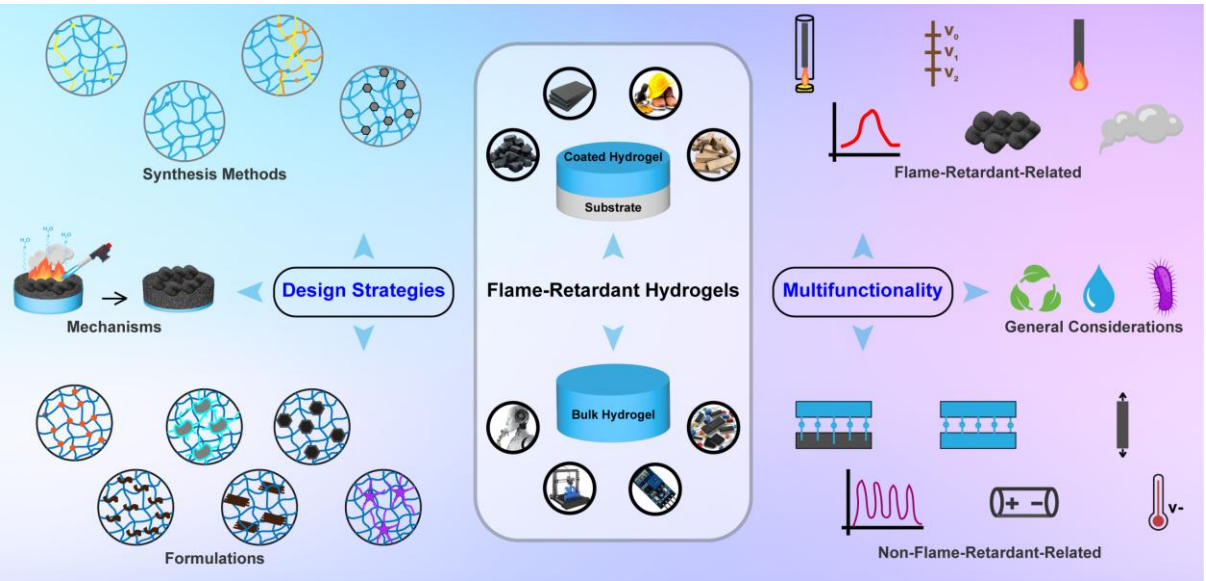
Abstract

Flame-retardant hydrogels are multifunctional materials that offer fire resistance, mechanical flexibility, self-adhesion, water retention, and self-healing properties. This review discusses recent developments in these materials, categorizing them into two types: coatings and bulk materials. Coating-type hydrogels provide fire protection for the surfaces of various materials, including coal, foam, wood, textiles, and safety gear. In contrast, bulk hydrogels possess flame-retardant properties suitable for applications in robotics, 3D printing, sensors, and energy storage. Moreover, this review highlights recent advancements in various design strategies for flame-retardant hydrogels, including their synthesis methods, mechanisms, formulations, and performance indicators. The synthesis techniques include physical, covalent, and dynamic crosslinking, allowing diverse network architectures. Flame-retardant mechanisms involve heat absorption, gas-phase dilution, char formation, intumescence, and radical quenching. The formulations typically incorporate additives such as polymers, phosphorus compounds, inorganic fillers, and nanomaterials. Key performance indicators assess both flame-retardant and non-flame-retardant functionalities, essential for advanced applications of flame-retardant hydrogels. Specifically, this review explores the multifunctional capabilities of flame-retardant hydrogels and compares these findings to previous studies, emphasizing their potential to broaden their applications in various fields. The review concludes by offering valuable insights into future directions for the development of flame-retardant hydrogels in next-generation fire-safe technologies.

Keywords: Flame-retardant hydrogels, applications, design strategies, multifunctional performance

Graphical Abstract



This review highlights recent advances in flame-retardant hydrogels, focusing on innovative design strategies including synthesis methods, mechanisms, and formulations for their use as coatings or bulk materials, emphasizing their multifunctional capabilities by incorporating both flame-retardant and non-flame-retardant functionalities.

1. Introduction

Fires pose a serious danger to public safety, infrastructure, and the environment, resulting in considerable loss of life and economic damage globally. To address these hazards, strict fire-safety regulations have been implemented in various industries, leading to the development of innovative flame-retardant technologies [1, 2]. Flame-retardant hydrogels have gained significant attention as materials designed for fire prevention owing to their unique ability to combine flame resistance with multifunctional properties [3].

Traditionally, flame retardants have been added to polymers and used as coatings or fillers to prevent ignition and control the spread of flames [4–7]. These substances include phosphates [8], halogen compounds [9], metal hydroxides [10], nitrogen-based agents [11], layered silicates, and nanomaterials (NMs) [12], each of which offers unique mechanisms for flame inhibition. Recently, the incorporation of flame-retardant additives into hydrogels (three-dimensional polymer networks that can hold large amounts of water) has led to safer, more innovative, and adaptable fire-protection solutions [13]. Hydrogels have emerged as a class of materials with great potential owing to their multifunctional properties, which include high strength, flexibility, self-adhesion, self-healing ability, and high-water retention [14–18]. These properties make them suitable candidates not only for use in conventional sensors and electronics, but also for innovative flame-retardant systems. Their function has evolved into two main forms: (1) as flame-retardant coatings that safeguard the surfaces of flammable materials like wood [19], coal [20], rigid polyurethane foams (RPUFs) [21], skin [22], and textiles [23]; and (2) as bulk materials where flame resistance is integrated throughout the hydrogel matrix for applications in energy storage devices, sensors, and robotic components [24, 25].

Unlike many other materials, multifunctional hydrogels are characterized by a particular set of features that render them versatile and capable of performing multiple functions [26]. Such features include soft mechanical properties, self-healing capability, anti-

freezing behavior, long-term water retention, and adhesion. Softness and flexibility are important aspects in the design of materials used in wearable sensors [27–29]. The self-healing function enables the repair of damaged sites and recovery of lost functionality. The anti-freezing capability of hydrogels allows them to operate at low temperatures without the risk of freezing, which is useful in a wide range of applications, including flexible sensors and stretchable electronics [30]. Similarly, resistance to dehydration enables these hydrogels to function over longer durations while maintaining their flexibility and electrical conductivity in smart electronic devices. Adhesion is essential for wearable hydrogels to firmly attach to the skin surface [14, 24, 25]. Hydrogels must adhere strongly to substrate materials to prevent peeling or detachment when applied as flame-retardant coatings [19]. Multifunctional hydrogels are highly valued for their easily tunable functional properties, leading to numerous applications [31, 32]. Furthermore, the incorporation of flame-retardant features into these hydrogels significantly enhances their market appeal and acceptance.

In recent years, significant advancements have been made in the development of hydrogels with enhanced flame-retardant properties. These include innovative synthesis techniques employing physical [33], chemical, or dynamic crosslinking [34], as well as the strategic incorporation of functional additives such as phosphorus compounds, carbon-based NMs, bio-based flame retardants, and hybrid fillers [4, 35–39]. Previous review articles on flame-retardant hydrogels have primarily concentrated on individual aspects, such as their flame-retardant mechanisms, chemical compositions, or applications [40–45]. However, to address the evolving demands of advanced technologies, such as wearable electronics, flexible energy devices, biomedical systems, and coatings, understanding the multifunctional performance of flame-retardant hydrogels is essential.

Multifunctionality is crucial in these hydrogels because it not only enhances their overall performance and reliability but also significantly expands their range of applications.

Integrating flame retardancy with features such as mechanical strength, electrical conductivity, self-healing properties, and environmental adaptability makes these hydrogels versatile for applications in wearable electronics, biomedical devices [46–48], smart textiles, and energy-storage systems. This combination enhances the efficiency, minimizes the number of required components, and supports the development of next-generation, high-performance, and sustainable solutions. This broader perspective highlights the importance of writing this review, especially considering recent advancements in material-design strategies for next-generation flame-retardant materials.

This review provides a timely and significant contribution to this field by means of a comprehensive overview of the recent developments in flame-retardant hydrogels, categorizing them primarily into two types: coatings and bulk materials. The design principles are analyzed, and their multifunctional performance is assessed. Unlike previous reviews that focused mainly on mechanisms or specific applications, this article emphasizes the integration of flame retardancy with additional properties such as mechanical robustness, environmental sustainability, and adaptability to emerging technologies. By critically evaluating synthesis strategies, mechanisms, formulations, and performance benchmarks, this review identifies current trends and knowledge gaps in the field and sets the stage for future innovations in sustainable application-specific hydrogel systems, positioning them as promising candidates for next-generation fire-safe and multifunctional materials.

2. Main Categories of Flame-Retardant Hydrogels

Flame-retardant hydrogels are used in various applications because they enhance fire protection, thermal insulation, and safety without compromising the material performance. These hydrogels are particularly valuable in areas such as coal, RPUFs, domestic materials, textiles, skin protection, personal-safety equipment, robotics, 3D printed components, energy storage,

sensors, and fire-detection technologies. Figure 1 summarizes the structures and primary applications of flame-retardant hydrogels, which can be categorized into two main types based on their applications.

2.1. Flame-Retardant Hydrogels as Fire-Resistant Coatings

Flame-retardant hydrogels in this category, as shown in Figure 1b, act as protective layers on substrates, providing a surface barrier against heat and oxygen to retard combustion. Notably, applications in this category include coatings on coal [20, 49–57], RPUFs [21, 58–61], fire-fighting textiles and fabrics [23, 62–69], gloves [69, 70], and domestic materials, including wood [19, 65, 71–76] and grass [65]. The flame-retardant mechanism typically depends on the specific additives used and high moisture content in the hydrogel layer, which helps absorb heat and delay ignition [49, 52, 53]. This is often followed by the formation of a char layer or intumescent barrier upon heat exposure, which provides additional insulation to the underlying substrate [50, 51, 58, 60]. The key design factors for flame-retardant hydrogels used as fire-resistant coatings include enhancing adhesion [63, 64], achieving uniform coverage [70], and ensuring a rapid response to fire exposure [69].

2.2. Flame-Retardant Hydrogels in Bulk Form

In this category, flame-retardant properties are incorporated into the bulk of the hydrogel material (Figure 1b). Applications include skin-burn protection [22], robotics [77], 3D-printed components [78, 79], and devices used in energy storage, such as electrolytes for capacitors [80–87] and batteries [88, 89], as well as electronics, sensors, and fire detection systems. Flame-retardant additives are evenly distributed throughout the hydrogel material, making flame resistance an intrinsic property of the material. These hydrogels function as standalone components, such as gel polymer electrolytes (GPEs) for supercapacitors (SCs) [80–82], in which flame retardancy helps inhibit the internal spread of fire through mechanisms such as endothermic decomposition and char development. Key design considerations include balancing the

mechanical strength and durability with flame resistance to ensure sustained performance under thermal stress [83–87].

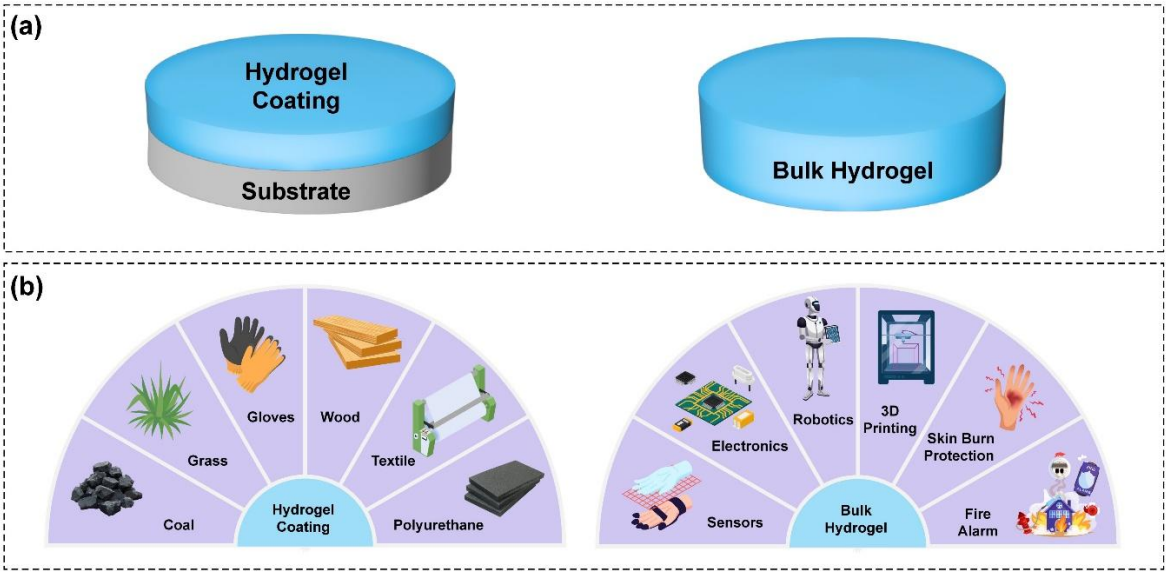


Figure 1. Schematic representation of flame-retardant hydrogel structures and their applications. (a) Hydrogels used as surface coatings and bulk materials. (b) Application areas of hydrogel coatings and bulk hydrogels, highlighting their multifunctionality.

2.3.Discussion

Flame-retardant hydrogels can be broadly divided into two categories: coatings and bulk materials. Coating-type hydrogels serve as protective layers on flammable surfaces such as textiles [67, 69] and coal [20], whereas bulk hydrogels integrate flame-retardant additives into the hydrogel matrix, making them suitable for applications in robotics [77] and energy devices [88, 89]. Coating hydrogels function by suppressing combustion on the surfaces to which they are applied [65, 69, 71]. By contrast, bulk hydrogels possess inherent flame-retardant properties, indicating that they are less prone to ignition and can help suppress internal combustion [85, 86].

3. Recent Advances in the Applications of Flame-Retardant Hydrogels as Fire-Resistant Coatings

Flame-retardant hydrogels act as protective layers on surfaces, significantly reducing heat and oxygen exposure to prevent combustion. These hydrogels are commonly utilized as coatings on various substrate materials, such as coal [20, 49–57], RPUFs [20, 49–57], fire-retardant textiles and fabrics [23, 62–69], gloves [69, 70], and domestic items such as wood [19, 65, 71–76] and grass [65], as shown in Figure 2c. In coal storage, hydrogel coatings help prevent spontaneous combustion, thereby mitigating the economic losses and environmental risks associated with coal fires [20, 54, 90]. For RPUF, flame-retardant hydrogels create a protective barrier that prevents heat transfer and the release of flammable gases, thereby enhancing fire safety in insulation and furniture [21, 58, 61]. Flame retardancy is essential for domestic materials like wood, bamboo veneers, grass, and window panels. It protects structures, furniture, and infrastructure, reduces fire risks, and provides more time for safe evacuation [63, 66, 82]. In personal protective equipment (PPE), such as gloves, textiles, fabrics, and paper, hydrogel coating enhances fire resistance and protects against burns and high temperatures [22, 63, 70]. Hydrogels are used as coatings on textiles to improve the fire resistance of protective clothing, industrial fabrics, and firefighting gear [42, 52, 84].

3.1.Approaches of Flame-Retardant Hydrogel Coatings on Substrates

The development of flame-retardant hydrogels involves the dissolution and combination of flame-retardant additives and polymers to formulate a hydrogel solution, as shown in Figure 2a. According to previous studies, these solutions can be applied using various techniques, including impregnation via dip coating, spraying, blading, infiltration, or brushing, direct pouring (controlled casting), layer-by-layer (LBL) assembly, attachment of a pre-polymerized hydrogel layer, and in situ polymerization on a substrate. These key processes are highlighted in Figure 2b.

In the dip coating method, a substrate is submerged in a hydrogel solution, pulled out at a regulated pace, and subsequently dried or cured to form a consistent coating [21, 51, 52, 59, 61, 62, 67]. Spray coating involves atomizing a hydrogel solution and applying it onto a substrate through a pressurized nozzle [20, 49, 50, 55, 65, 76]. Blading involves depositing a hydrogel precursor uniformly across the surface using a blade. The gap between the blade and the surface controls the coating thickness [73]. Infiltration is the process by which the hydrogel solution penetrates into the pores, voids, and capillary structures of the substrate before final gelation [54, 57]. Brushing is a manual technique whereby a brush or roller is used to spread the hydrogel solution onto a surface [55, 75]. Direct pouring (controlled casting) is the coating technique in which hydrogel is poured directly onto the substrate surface, but its shape is confined by molds or spacers [71]. LBL assembly entails the sequential deposition of several ultra-thin layers of hydrogel along with flame-retardant materials onto a substrate [91, 92]. The attachment of a pre-polymerized hydrogel layer involves separately preparing the hydrogel, curing it into a solid layer, and then affixing it to the substrate [23, 60]. In-situ polymerization is a method where hydrogel monomers and crosslinkers are polymerized directly onto the surface of a substrate, either through physical crosslinking, which does not require curing, or through covalent crosslinking, which involves a curing process, resulting in a strongly bonded hydrogel layer [19, 59, 63, 68, 72]. In these coating processes, the hydrogel coating forms upon completion of the gelation. During formation, crosslinking reactions improve the strength of the hydrogel coating. Additionally, the polymer concentration can be adjusted to regulate viscosity and gelation time.

For instance, when focusing on the application of hydrogels as flame-retardant coatings for coal, it is essential to select appropriate base polymers, additives, and water content to tailor the specific properties of the hydrogel solutions that protect coal. For example, hydroxypropyl methylcellulose (HPMC), polyethylene glycol (PEG), and chitosan (CS) create thermosensitive hydrogels with high water retention and fire resistance [49]. Lignin-based

hydrogels, made from lignin, acrylic acid, and N-isopropyl acrylamide, enhance thermal stability [90], while eco-friendly options utilize polyvinyl alcohol (PVA) and sodium alginate [51, 93]. Iron-based hydrogels, incorporating sodium carboxymethyl cellulose and ferric citrate, exhibit strong adhesion to the coal surface and provide oxygen barrier properties [53].

The presence of flame-retardant additives in the base hydrogel solution is crucial. For instance, hybrid shell bio-fillers like di(triethanolamine) titanate diisopropyl enhance fire resistance of coal by non-reactive gas phase formation for dilution of combustion [51], and expanded microspheres (EMs) in HPMC-CS-EM hydrogels support early water release and oxidation inhibition to retard combustion in coal [55]. Piceatannol-loaded nanocomposite hydrogels effectively reduce spontaneous combustion in coal by targeting reactive free radical groups [56].

After formulating the flame-retardant hydrogel solution, it can be applied to coal using three methods: direct spraying [54], injection into goafs [51], or dipping [90]. After applying the hydrogel solution to the coal, a hydrogel coating forms as the gelation process completes. During hydrogel formation, crosslinking reactions improve the strength of the hydrogel coating. For instance, chemical crosslinking with glutaraldehyde or borax [52] and physical crosslinking through hydrogen bonding enhances the stability of hydrogel coatings [90]. Multi-network crosslinked gels using guar gum, sodium tetraborate, and PEG boost fire inhibition performance with stable hydrogel coating performance [52].

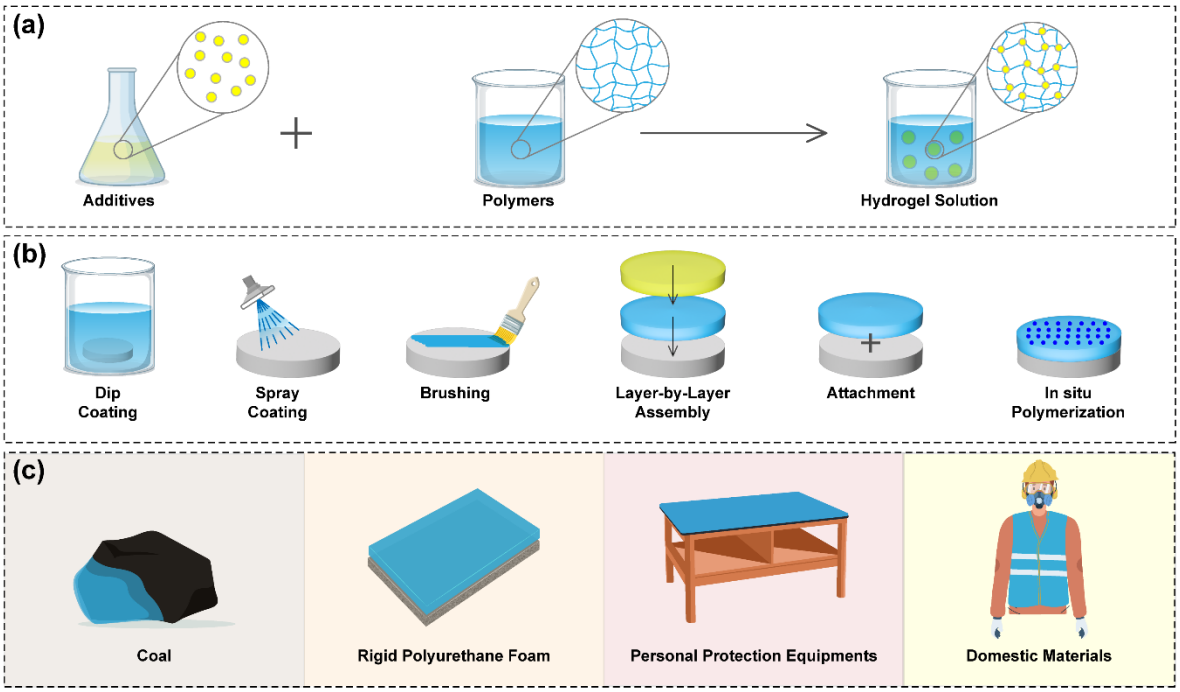


Figure 2. Overview of: (a) the preparation of flame-retardant hydrogel solutions, (b) recent strategies for applying these hydrogels onto substrates, and (c) representative applications as flame-resistant coatings.

Table 1 presents recent advances in substrate materials, application goals, and coating techniques, along with examples relevant to the potential areas of use.

Table 1. Recent advances in substrate materials, application goals, and techniques for flame-retardant hydrogel coatings, along with examples related to the potential areas of use.

Substrate Material	Objective	Application methods for flame-retardant hydrogel	Example
Coal	To prevent the spontaneous combustion of coal, a flame-retardant protective barrier should be created.	Injection [51] or spraying [54] onto high-risk coal seams and mined areas using pumps and nozzles [90] allows the hydrogel solution to penetrate coal pores and cracks.	Mixing a base polymer with water and functional additives like hydroxypropyl methylcellulose (MC), polyethylene glycol, chitosan (CS), sodium alginate (SA), metal ions (Fe, Al), antioxidants (e.g., Piceatannol), and fire-retardant additives [20, 49–57].
RPUF	To create an effective fireproof barrier on RPUF, a protective layer is needed to delay ignition, reduce heat release, and suppress smoke, while	Casting and drying [60], brushing or spraying [59, 61], and application of pre-polymerized hydrogel solution [21].	Dual-nucleophilic ester-induced T-P5-PAA hydrogel [60], polyacrylamide-MXene hydrogel nanocomposite [59], double-network hydrogel (DH) coating composed of polyacrylic-polydopamine

	preserving the mechanical integrity of the material.		[21], and self-healing and recyclable hydrogel coating made of polyvinyl alcohol (PVA)-borax [61].
Domestic materials (wood, bamboo, and grass)	To enhance the fire resistance of unspecified wood [19, 73, 76], pine wood [72], poplar wood [71], bamboo veneers [75], and grass [65], providing high water retention, thermal insulation, and self-healing capabilities.	Spraying [65, 72], blade technique [19], painting (brushing) [73], spraying and absorption [76], and soaking (immersion or dipping) [71, 74].	Self-healing cellulose nanocrystal (CNC)-borax hydrogels [72], MC/silica (SiO_2) hybrid hydrogels made with DOPO-ITA-modified cellulose and SiO_2 nanoparticles (NPs) [65], PVA/phytic acid (PA) hydrogel coatings [19, 94], PVA/PA/MXene-based coatings [73], SiO_2-based hydrogel coatings [76], and gelatin (Gel)[[95]/poly(acrylamide-co-acrylic acid)/calcium chloride ($CaCl_2$) hydrogels [71, 74].
Firefighting and personal	To enhance the fire resistance of textiles and fabrics	Doctor blading technique [66, 70], dipping [62, 66, 69, 70], LBL assembly approach [63], spray	Multi-walled nanotube (MWNT)-Gel hydrogel paste [70], multifunctional

protection equipment	(cellulosic fabric [69], cotton fabric [66], polylactic acid fabric [64], polyester fabric [63]), nitrile and elastomeric gloves [70], PPE and wearing [62, 96], and cellulosic paper [70], helping to safeguard against burns and exposure to high temperatures.	coating [63, 64], and in-situ polymerization [62].	conductive cellulose fabric after immersion in ammonium salt of ethylene glycol diphosphoric acid [69], poly(N-isopropylacrylamide) [PNIPAAm]/SA/PVA hydrogel [66], polypyrrole (PPy)/CS hydrogel [64], calcium alginate (CA)/PVA/expandable graphite (EG) hydrogel [62], and acrylamide-SiO_2 NP hydrogel [96].

4. Recent Advances in the Applications of Bulk-Form Flame-Retardant Hydrogels

In bulk-form flame-retardant hydrogels, flame-retardant additives are evenly distributed throughout the hydrogel matrix, making flame resistance an intrinsic property of the material. Unlike flame-retardant hydrogel coatings that attach to substrates, these hydrogels function as standalone components, such as skin-burn protection [22], robotics [77], 3D-printed components [78, 79], and energy-storage devices, including electrolytes for capacitors [80–87] and batteries [88, 97], as well as electronics, sensors, and fire detection systems [71, 74, 95, 97–101], as summarized in Figure 3d. Flame-retardant hydrogels have shown significant potential for use in fire alarms and strain sensors. [71, 74, 98–101]. They offer significant advantages for improving the reliability and safety of fire-detection systems in high-risk industrial settings. Flame-retardant hydrogels show great promise as electrolytes for SCs owing to their exceptional energy-storage capacity [80, 81, 83, 84, 86, 87], self-adhesion [80], self-healing [80, 82], low-temperature resistance [80–84], and water-retention properties [80–82, 85]. Fire resistance in energy storage devices is essential to suppress thermal runaway, prevent electrolyte ignition, inhibit cell-to-cell propagation, and ensure safe operation under abuse conditions by simultaneously controlling heat, oxygen, and reactive species. Similarly, hydrogels are increasingly used in robotics because of their flame-retardant properties, flexibility, and ability to protect sensitive components. When combined with montmorillonite (MMT) [77] or multi-walled carbon nanotubes (MWNTs) [70], they form a stretchable skin that can withstand high temperatures, enabling robotic components to sense and operate safely under fire-prone conditions. Furthermore, these hydrogels are now rapidly fabricated via vat photopolymerization, forming well-defined layered structures that resist heat and slow combustion, making them ideal for eco-friendly 3D-printed components in electronic, aerospace, and construction applications [102].

4.1.Approaches of Flame-Retardant Hydrogels in Bulk Form

To create effective flame-retardant hydrogels in bulk, it is important to incorporate flame-retardant additives into the hydrogel networks. The synthesis methods are similar to those of general hydrogel synthesis, with the key difference being the integration of flame-retardant additives. Figure 3b summarizes the general procedure for fabricating hydrogel networks. Hydrogels with integrated flame-retardant properties are developed using this procedure.

Several methods for hydrogel synthesis have been reported in the literature, as summarized in Figure 3a. One approach involves physical crosslinking, where water-soluble, unmodified, or modified polymers are combined with crosslinking agents to form hydrogel networks. In this method, polymer chains are crosslinked through non-covalent interactions such as ionic, hydrophobic, or hydrogen bonding [33, 103–105]. Another method involves covalent crosslinking, in which polymers react with chemical crosslinkers to form hydrogel networks via covalent bonds. This process can occur by forming covalent bonds between unmodified or modified polymer chains, or by polymerizing synthetic monomers in the presence of chemical crosslinkers and initiators [106–108]. A third approach uses modified polymers and crosslinkers through single or multiple dynamic bond reactions, including Schiff-base, hydrazone, boronic ester, and disulfide bonds, which produce dynamic covalent crosslinks within hydrogel networks. This method is particularly attractive because it allows the introduction of self-healing and self-adhesive properties into hydrogels [109–112]. Such attributes can also be achieved in physically crosslinked hydrogels when multiple physical crosslinks, including hydrogen bonds and metal-polymer coordination, act as dynamic connections [109, 113–115]. Moreover, combining physical, covalent, and dynamic covalent crosslinks can yield these attributes [116–118]. Based on their network composition, whether physical, chemical, or a combination of both, hydrogels can be categorized into single-, double-, and triple-network structures [119–121], which are shown in Figure 3c. These networks provide structural stability, thereby improving mechanical properties and other key functionalities. Different fabrication

methods or their combinations yield various performance characteristics, including stretchability, energy dissipation, fracture toughness, and multifunctional features such as freezing resistance, dehydration resistance, self-adhesion, and self-healing [122–124]. NMs can also be introduced into hydrogel networks to create composite hydrogels and further enhance their functional performance [124–126].

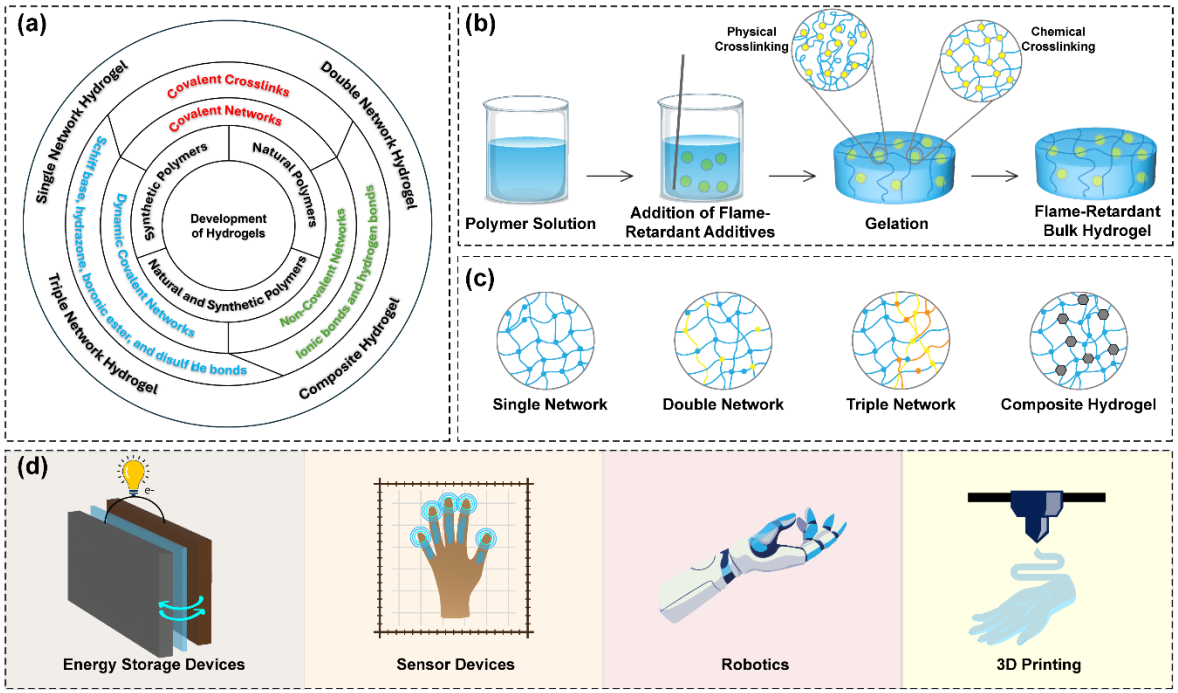


Figure 3. Advances in bulk flame-retardant hydrogels: (a) general strategies for hydrogel development, (b) typical synthesis steps incorporating flame-retardant additives, (c) various network structures for enhanced properties, and (d) application areas.

5. Mechanisms of Flame Retardancy in Hydrogels

The flame-retardant properties of hydrogels, including flame-resistant hydrogel coatings and bulk flame-retardant hydrogels, arise from various mechanisms that delay ignition or extinguish fires once ignited. The underlying mechanisms are largely similar for both the coating and bulk forms. Understanding these mechanisms allows the rational design of

materials by integrating appropriate flame-retardant additives and optimizing their compositions through different hydrogel synthesis approaches.

These mechanisms are illustrated in Figure 4, which depicts the sequential stages of the process. In stage 1, water is released from the hydrogel, providing a cooling effect. In stage 2, the polymer chains ignite following dehydration. In stage 3, several flame-retardancy mechanisms are involved, including charring, the release of non-flammable gases, and intumescence. Finally, in stage 4, the fire is extinguished. Also, the key flame-retardant mechanisms and their associated additives are summarized in Table 2, which outlines their functions and effects on fire suppression.

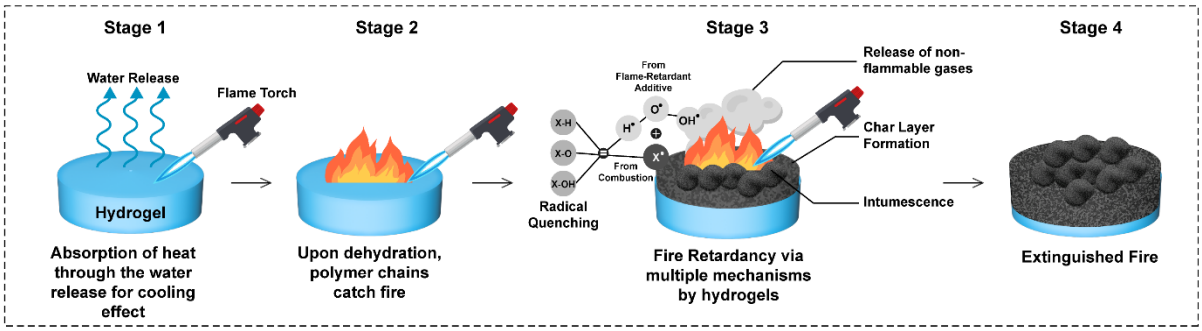


Figure 4. Schematic illustration of the stages and mechanisms involved in the flame-retardant actions of hydrogels, applicable to both hydrogel coatings and bulk materials.

5.1. Absorption of Heat Through the Water Content for Cooling Effect

Hydrogels are polymer networks that retain significant amounts of water. When formulated correctly, these hydrogels can absorb heat upon exposure to heat or flame, causing the water to evaporate as the heat or flame reaches the polymer network, providing a cooling effect. This process lowers the temperature of the surrounding area and dilutes the flammable gases. As a result, conditions favorable for ignition are postponed, minimizing the spread of flames. Introducing small hygroscopic organic molecules, such as glycerol [82, 127], and hygroscopic fillers, such as spindle-shaped aluminum hydroxide [71], into a hydrogel network can enhance

the interaction between the molecules or fillers and water. This results in a smoother water release pattern at high temperatures, improving the cooling effect.

5.2.Non-Flammable Gas (Gas Phase) Release for Dilution of Combustion

Hydrogels can contain various functional groups such as amine (-NH_2), carboxyl (-COOH), ammonium (NH_4^+), and hydroxyl (-OH). Flame-retardant polymers or flame-retardant additives can provide these functional groups. For example, alginate [66] and chitosan [127, 128] serve as flame-retardant polymers and are rich in -NH_2 and -COOH functional groups, respectively, along with -OH groups. When exposed to flames, these polymers release their functional groups, forming non-flammable gases such as H_2O, N_2, CO_2, and NH_3. These gases constitute the gas phase. This process dilutes any flammable volatile compounds and displaces oxygen in the combustion zone during ignition. As a result, flame propagation is inhibited, ultimately extinguishing the fire. Similarly, ammonium salts provide abundant NH_4^+ ions, which produce NH_3 gas when exposed to flame, thus enhancing the flame retardancy of hydrogels [84].

5.3.Protective Char Layer (Condensed Phase) Formation for Thermal Barrier and Limiting Access to Oxygen

Hydrogels can incorporate either polymer networks or additives that create a protective carbon-rich layer when exposed to flame. This layer forms through the release of functional groups such as -OH, -NH_2, and -COOH, accompanying degradation and carbonization processes [101]. This protective layer disrupts the flame's interaction with the hydrogel surface and its internal structure; it is called the char layer. The char layer possesses thermal barrier properties, reduces heat transfer, and limits access to oxygen; hence, further combustion is prevented, thereby promoting flame starvation and smothering [129].

For example, naturally occurring char-forming polymers can be integrated into hydrogel networks. Some of these polymers include chitosan [83, 84, 127, 128, 130], lignin [90, 131],

starch [132], alginate [66, 87, 101], agarose [85], and carrageenan [86]. Additionally, carbon-based materials such as graphene oxide (GO) and carbon nanotubes (CNTs) [70, 100] can also serve as char-forming additives. Small biomolecules like tannic acid (TA) [54, 133] can perform similar functions in promoting char formation, thereby enhancing the flame-retardant properties of hydrogels. The use of phosphate-based flame-retardant additives can catalyze the formation of thick char layers in the formulations [19]. Moreover, the inclusion of inorganic fillers like silica [120], MXene [73], kaolin [134], basalt [135], and mineral residues [58, 84] can enhance the char layers by making them denser and harder. This effectively reduces oxygen access to the combusting material. The char layer constitutes the condensed phase. Similar to carbon-rich compounds, inorganic salts such as lithium chloride (LiCl) [82], sodium chloride (NaCl) [101] contribute to the formation of lithium carbonate and sodium carbonate as a mineral layer, which provides a barrier effect similar to that of a char layer.

5.4.Intumescence for Thermal Resistance Through Entrapped Air Volume

Intumescent additives can be incorporated into hydrogels to impart flame-retardant properties. For instance, expandable graphite is an intumescent additive in hydrogels, as it can hold sulfuric acid between its carbon layers. When the hydrogel is exposed to fire, the expandable graphite produces gases such as CO_2 and SO_2 through a redox reaction. This process increases the pressure between the carbon layers, causing the graphite to expand in volume. The expanded graphite traps a significant volume of entrapped air (gas phase), which provides resistance to heat transfer when exposed to heat and flames [62].

5.5.Radical Quenching for Interruption of the Combustion Process

Hydrogels can incorporate radical quenching additives to exhibit flame-retardant properties. When combustion is initiated upon exposure to flame, high-energy free radicals such as Ḣ, Ȯ, and ȮH are generated in the gas phase, which sustain the combustion chain reaction. By converting these radicals into low-energy free radicals, the combustion process can be slowed

down, resulting in an extinguishing effect. Furthermore, when combined with polymers, these additives enhance dehydration, converting polymers into a thick, carbon-rich char. This char formation retards the combustion process and helps to prevent the transfer of external heat and oxygen, ultimately controlling the spread of fire and extinguishing the flames.

Commonly used additives include LiCl [82], NaCl [101] phosphorus-containing compounds such as organophosphates (e.g., resorcinol bis(diphenyl phosphate) [22], triethyl phosphate [136], sodium polyacrylate (PA-Na), and phytic acid (PA) [19, 137, 138] and inorganic phosphates (such as sodium dihydrogen phosphate [98] and ammonium polyphosphate (APP) [135, 139]). These additives consume the combustion-promoting free radicals, thereby quenching them and extinguishing the flame. For instance, LiCl produces $\dot{\text{Cl}}$ free radicals that react with high-energy free radicals, generating low-energy $\text{H}\dot{\text{Cl}}$ free radicals that quench the combustion. Phosphorus-containing flame-retardant compounds similarly provide $\text{P}\dot{\text{O}}_x$ free radicals for quenching.

Table 2. Mechanisms of flame retardancy in hydrogels.

Mechanisms	Explanation	Key Additives/Materials	Effect
Cooling Effect	The absorption of heat by water leads to evaporation, which in turn lowers the surrounding temperature.	Water, hygroscopic organic molecules such as glycerol (GLY) [82, 127], and fillers like spindle-shaped aluminum hydroxide [71].	- Water absorbs heat and evaporates, which lowers the temperature and delays ignition. - GLY helps in retaining and gradually releasing water, thereby prolonging the cooling effect. - Aluminium hydroxide releases water vapor when heated, which further cools the material and forms a protective barrier.

Gas-phase Dilution	Release of non-flammable gases such as CO_2, NH_3, and water dilutes flammable gases and displaces oxygen.	Flame-retardant polymers, such as alginate [66] and CS [127, 128], along with ammonium salts [84].	Flame-retardant polymers like alginate and CS, along with ammonium salts, contain functional groups that generate gaseous phases upon exposure to flames, helping to dilute combustion.
Char Formation	A carbon-rich layer forms during degradation, serving as a thermal barrier and restricting oxygen access.	CS [83, 84, 127, 128, 130], lignin [90, 131], starch [132], alginate [66, 87, 101], and carbon-based materials (e.g., GO, CNTs [70, 100]), phosphorus-containing additives such as main-chain poly(phosphoester)s , LiCl [82], and sodium chloride (NaCl) [101]serve as potential char formers.	- When heated, the char-forming components of the hydrogel degrade to create a protective carbon layer that reduces heat transfer and limits oxygen access, preventing further combustion and inhibiting flame spread.

			- LiCl and NaCl contribute to the formation of lithium carbonate and sodium carbonate as a mineral layer, which provides a barrier effect similar to that of a char layer.
Intumescence	Flame-retardant additives expand when exposed to heat, trapping air in the gas phase to provide thermal resistance.	EG and sulfuric acid [62] can potentially act as intumescent agents.	Intumescence enhances flame retardancy by causing additives like EG to expand when heated. This creates CO_2 and sulphur dioxide gas layers that form an insulating barrier, reducing heat transfer.
Radical Quenching	This process involves neutralizing free radicals that promote combustion, which helps	LiCl [140], phosphorus-based compounds such as organophosphates (e.g.,	Additives like LiCl or phosphorus compounds can quench free radicals generated

	to slow down the chain reaction of combustion.	resorcinol bis(diphenyl phosphate, RDP) [22], triethyl phosphate, sodium phytate (PA-Na), and PA [19, 137, 138]) and inorganic phosphates (such as sodium dihydrogen phosphate [98] and ammonium polyphosphate (APP) [135, 139])	during combustion, turning them into low-energy ones. This slows the combustion process and reduces flame spread.

6. Recent Developments in Flame-Retardant Hydrogel Formulations

This section highlights the incorporation of different flame-retardant additives into hydrogel networks to enhance their flame-resistant properties, as observed in flame-resistant hydrogel coatings and bulk-form flame-retardant hydrogels. An effective flame-retardant hydrogel should utilize various flame-retardancy mechanisms to enhance its overall performance. When multiple flame-retardant mechanisms work together, they significantly boost their ability to resist flames. In this context, different approaches for integrating flame-retardant additives into hydrogels have been examined, considering the various mechanisms of flame retardancy.

6.1. Incorporating Phosphorus-Containing Small Molecules or Compounds as Flame-Retardant Additives

In this approach, phosphorus-containing small molecules or compounds, such as resorcinol bis(diphenyl phosphate (RDP) [22], triethyl phosphate [136], PA-Na, and PA [19, 137, 138], are integrated as flame-retardant additives into hydrogel networks. For instance, Zhao et al. developed a PVA/PA hydrogel through noncovalent interactions (hydrogen bonds), as shown in Figure 5a [19]. PA was dissolved in a PVA solution and then applied to a wood surface using a blade. The coated wood was subsequently frozen at -20 °C to form the hydrogel layer. In this hydrogel, PA provides flame-retardant functionality by acting as a carbonization catalyst, encouraging the initial drying and charring of the wood. A more compact and thicker char layer is formed, which impedes heat and oxygen, thereby reducing further combustion.

Li et al. reported a flame-retardant polyelectrolyte hydrogel based on polyacrylamide (PAM) and the polysaccharide pullulan (PUL) that used PA as a flame-retardant additive for zinc-ion capacitors (ZICs) [81]. The PA promoted char formation and inhibited further polymer decomposition. PA was integrated into the hydrogel via hydrogen bonding during free-radical polymerization to form the PAM/PUL/PA hydrogel (Figure 5b). Xue et al. developed a hydrogel for skin protection using xanthan gum (XG) and RDP-coated starches. In this system,

the organophosphate compound RDP was incorporated through hydrogen bonding, imparting flame-retardant properties. The RDP promoted char formation and reduced heat release. The RDP-modified starch and XG were mixed to form a hydrogel network via hydrogen bonding (Figure 5c) [22].

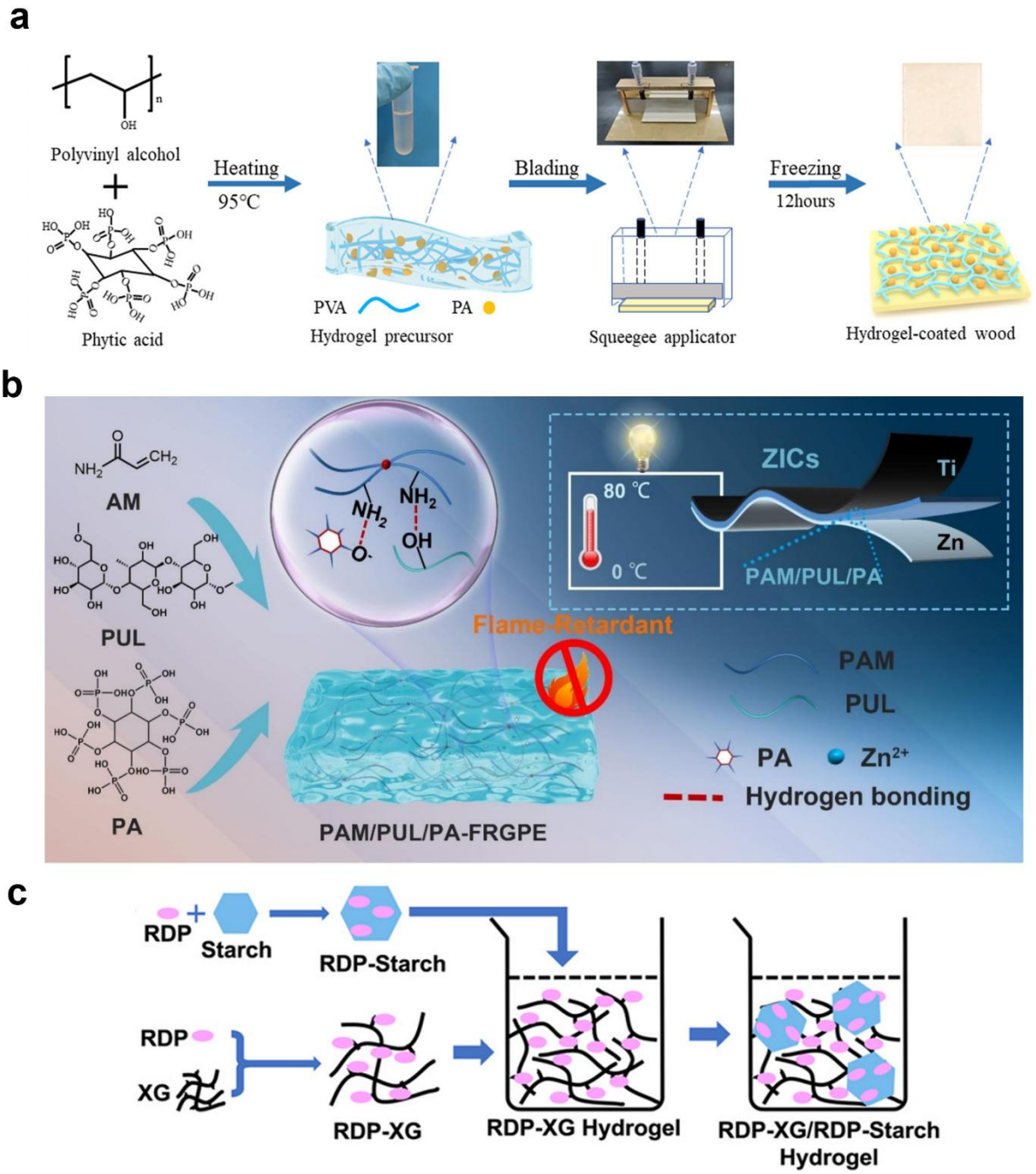


Figure 5. Representative flame-retardant hydrogels incorporating phosphorus-containing small molecules: (a) Synthesis of a flame-retardant hydrogel coating for wood utilizes non-covalent interactions (hydrogen bonds), composed of PVA and PA, adapted with permission

from Ref. [19] Copyright 2021, Elsevier B.V. (b) Development of a flame-retardant polyelectrolyte hydrogel (PAM/PUL/PA-FRGPE) based on the polysaccharide PUL, which incorporates PA through hydrogen bonding as a flame-retardant additive for zinc ion capacitors, adapted with permission from Ref. [81] Copyright 2024, Elsevier B.V. (c) Fabrication of a flame-retardant hydrogel for skin protection using XG and RDP-coated starch, crosslinked via hydrogen bonding, adapted with permission from Ref. [22] Copyright 2021, American Chemical Society.

6.2. Incorporating Phosphorus-Containing Long-Chain Compounds as Flame-Retardant Additives

This method involves incorporating phosphorus-containing long-chain compounds into hydrogel networks to enhance the flame-retardant performance. The long-chain additive APP has been shown to be particularly effective. For example, Chen et al. developed a hydrogel that incorporated modified APP (MAPP). The intumescent properties of MAPP contributed to the formation of a protective char layer upon exposure to flames. Initially, APP was coated with SiO_2 and further modified to introduce carbon-carbon double bonds, resulting in MAPP. The MAPP was then added to an alginate/poly(acrylamide-co-stearyl methacrylate) [alginate/P(AAm-co-SMA)] hydrogel via free-radical polymerization in a one-pot synthesis, allowing covalent crosslinking with the hydrogel (Figure 6a) [139].

Zheng et al. incorporated water-soluble APP into polyacrylamide (PAAm) hydrogel networks reinforced with basalt fibers (Figure 6b) [135]. The hydrogels were synthesized by directly adding APP to the polymerization mixture. The inclusion of woven basalt fibers significantly enhanced both the mechanical strength and thermal stability. This SiO_2-based reinforcement material, which was capable of withstanding high temperatures, synergistically contributed to the fire resistance of the hydrogel. APP further improved the flame retardancy by forming a protective char layer that inhibited direct flame contact.

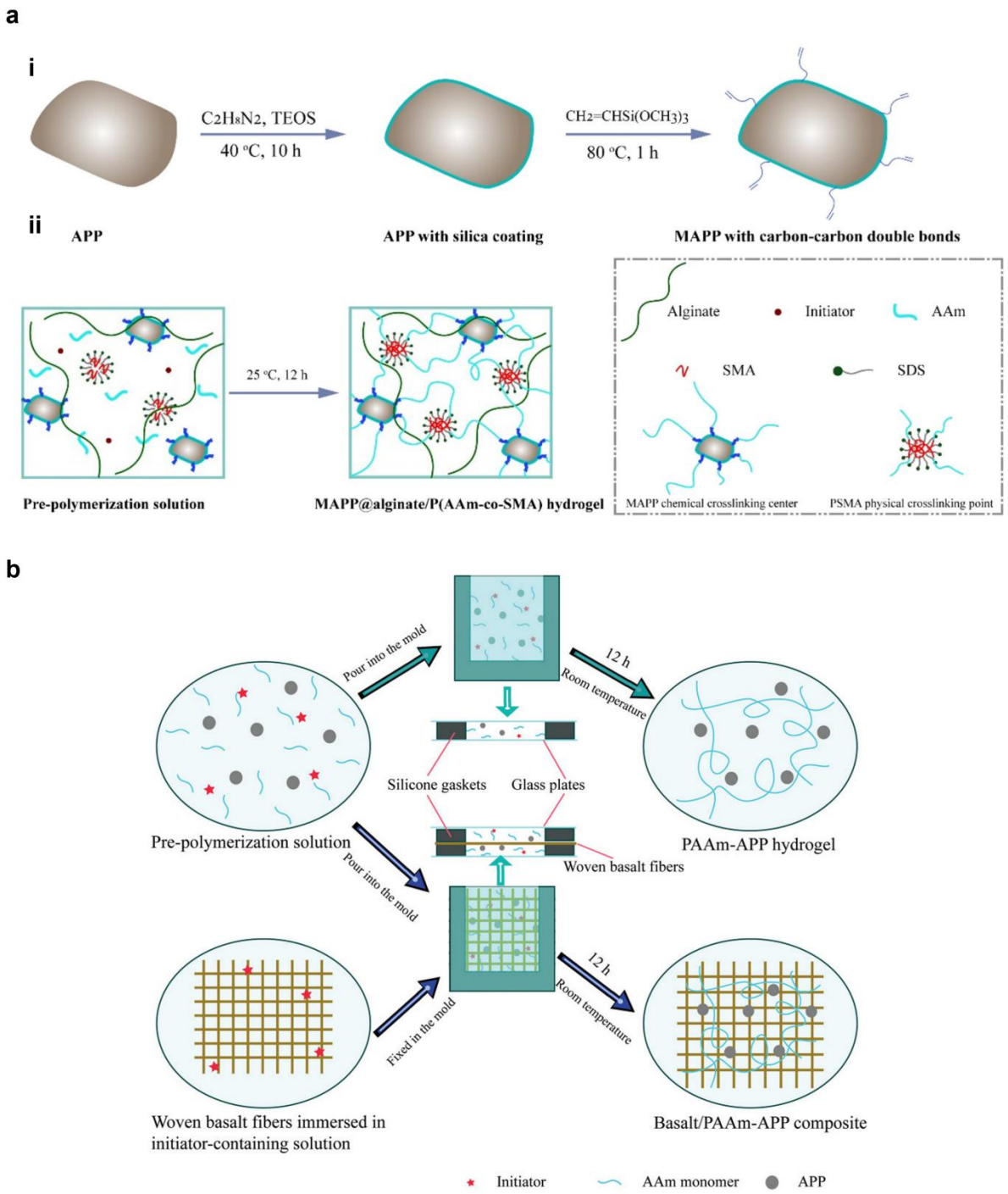


Figure 6. Synthesis strategies for flame-retardant hydrogels incorporating APP and basalt fibers: (a) Synthesis of MAPP@alginate/P(AAm-co-SMA) flame-retardant hydrogel using APP modified by carbon–carbon double bonds, adapted with permission from Ref. [139]. Copyright 2021, Wiley Periodicals LLC. (b) Synthesis of basalt/PAAm-APP flame-retardant hydrogel using water-soluble APP, adapted with permission from Ref. [135]. Copyright 2020, Wiley-VCH GmbH.

6.3.Incorporating Polymers as Flame-Retardant Additives

In this approach, polymers are incorporated as flame-retardant additives into hydrogel networks, using either covalently crosslinked polymer networks or noncovalently crosslinked structures. Examples of such polymers include CS [83, 84, 127, 128, 130], lignin [90, 131], starch [132], alginate [66, 87, 101], agarose (Ag) [85], and carrageenan [86]. Zuo et al. recently developed a 3D-printed hydrogel using all-natural polymers, including itaconic acid (IA), sodium alginate (SA), and gelatin (GE) [102]. These polymers were modified to include reactive polymerization sites and were utilized as 3D printing inks to produce covalently crosslinked hydrogels via photopolymerization in the presence of a poly(ethylene glycol) diacrylate (PEGDA) crosslinker, as shown in Figure 7a. The resulting hydrogels, itaconic acid (IAE)/gelatin-based (GEV)/sodium alginate (SAV) monomers, exhibited improved flame-retardant properties owing to enhanced char formation and the release of nonflammable gases, which acted as barriers against heat and oxygen (Figure 7b).

In another study, Huang et al. developed a hydrogel coating using non-covalently crosslinked double networks of PAAm and polydopamine (PAAm–PDA), employing hydrogen bonding, hydrophobic interactions, and mechanical interlocking to improve the flame retardancy of RPUF, as shown in Figure 7c-i. In a standard method, RPUF was first cleaned, treated with PDA derived from dopamine in NaOH (pH 11), and subsequently coated with a PAAm hydrogel precursor solution, which was then cured using either UV light (20 W, 365 nm) or heat (60 °C for 30–60 min), as illustrated in Figure 7c-ii. This treatment enhanced the formation of a thick, carbon-rich char layer, which slowed combustion and improved flame resistance [21].

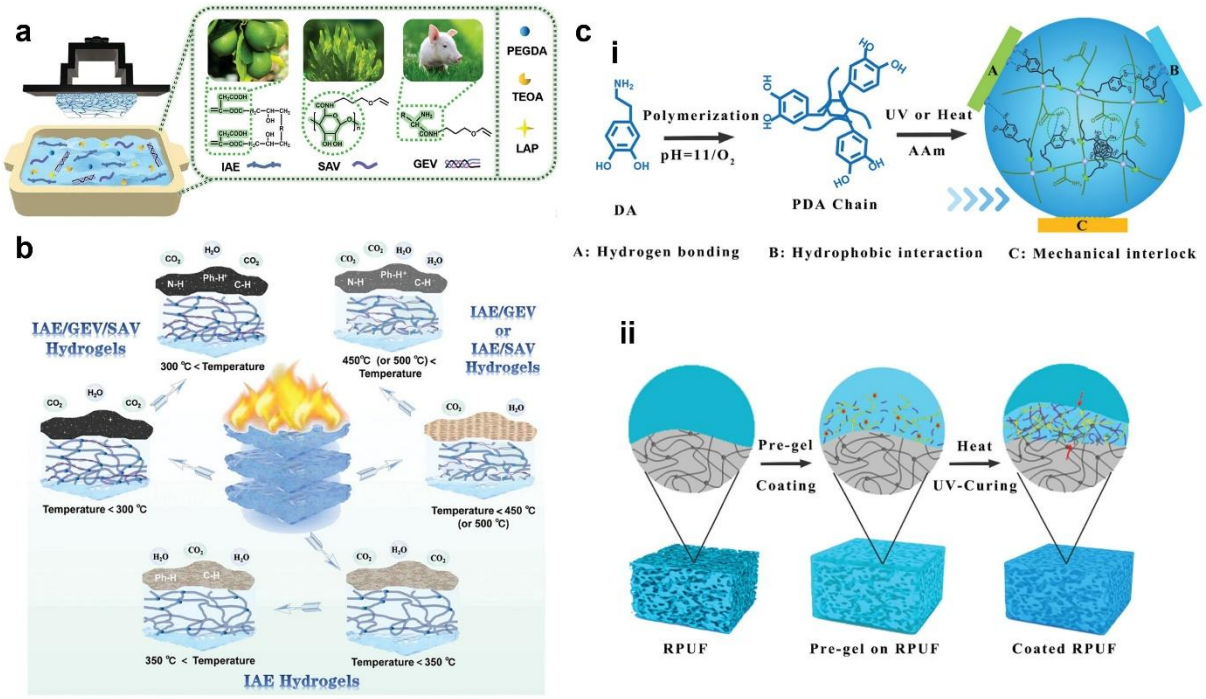


Figure 7. Design strategies for flame-retardant hydrogels based on natural polymers and polydopamine: (a) Components of 3D printing inks based on modified natural polymers: IA, SA, and GE. (b) Proposed combustion pathway of the IAE/GEV/SAV hydrogel, illustrating the release of gaseous byproducts and providing a comparison with control hydrogels, adapted with permission from Ref. [102]. Copyright 2023, Wiley-VCH GmbH. (c)- i) Synthesis route and schematic illustration of a PAAm–PDA hydrogel coating and ii) schematic representation of the coating process on RPUF using PAAm–PDA hydrogel, adapted with permission from Ref. [21] Copyright 2021, The Royal Society of Chemistry.

6.4.Incorporating Inorganic Fillers as Flame-Retardant Additives

In this approach, inorganic fillers such as SiO_2 [120], MXene [73], kaolin [134], bentonite [141], basalt [135], borax [72], and lithium acetate [85–87]are incorporated, along with composite fillers such as Mxene@Halloysite nanotubes (HNTs) [142] and mineralized black phosphorus@SiO_2 nanofibers [58].

In a representative study, a hydrogel coating composed of PVA, PA, and MXene was developed to enhance the flame resistance of wood. This hydrogel was synthesized using a

one-pot heating approach followed by freeze–thaw cycles, resulting in a uniform and stable coating on the wood surface, as illustrated in Figure 8a [73]. It improved the flame retardancy through water-evaporation-induced cooling, the formation of a physical barrier, phosphorus-based char generation (from PA), and enhanced thermal stability. Notably, the MXene contributed to flame resistance by forming a robust char layer and reducing the heat release.

In a separate investigation, hydrogels with flame-retardant properties were fabricated using cellulose nanocrystals (CNCs) derived from wood and cross-linked via boronate–ester bonds. Borax ($Na_2B_4O_7 \cdot 10H_2O$) powder was gradually added to the CNC solution under stirring, as shown in Figure 8b [72]. Dynamic boronate ester bonds were formed between the CNCs and borax ions. The CNCs facilitated carbonization and protective char formation, whereas the borax released water vapor to suppress combustion and generate an insulating glassy layer. This combination significantly enhanced the flame retardancy of the hydrogel.

In another study involving composite fillers, HNTs were embedded within multilayer MXene nanosheets to create a "sandwich filling" configuration. The MXene@HNT composite was applied to the surface of RPUF via surface coating. A double-network hydrogel (DH) composed of SA and CS was used as an outer protective layer to form a flame-retardant system, as shown in Figure 8c [122]. The MXene@HNT structure produced a dense network that blocked heat and smoke penetration. During combustion, the MXene was converted to titanium dioxide (TiO_2), while the HNT decomposed into aluminum oxide (Al_2O_3) and silicon dioxide (SiO_2), which enhanced the formation of a protective char layer. This layer slowed the heat transfer and reduced the flammability. Additionally, the hydrogel released water and flame-inhibiting gases (N_2, NH_3) at high temperatures, which helped dilute the oxygen in the combustion zone.

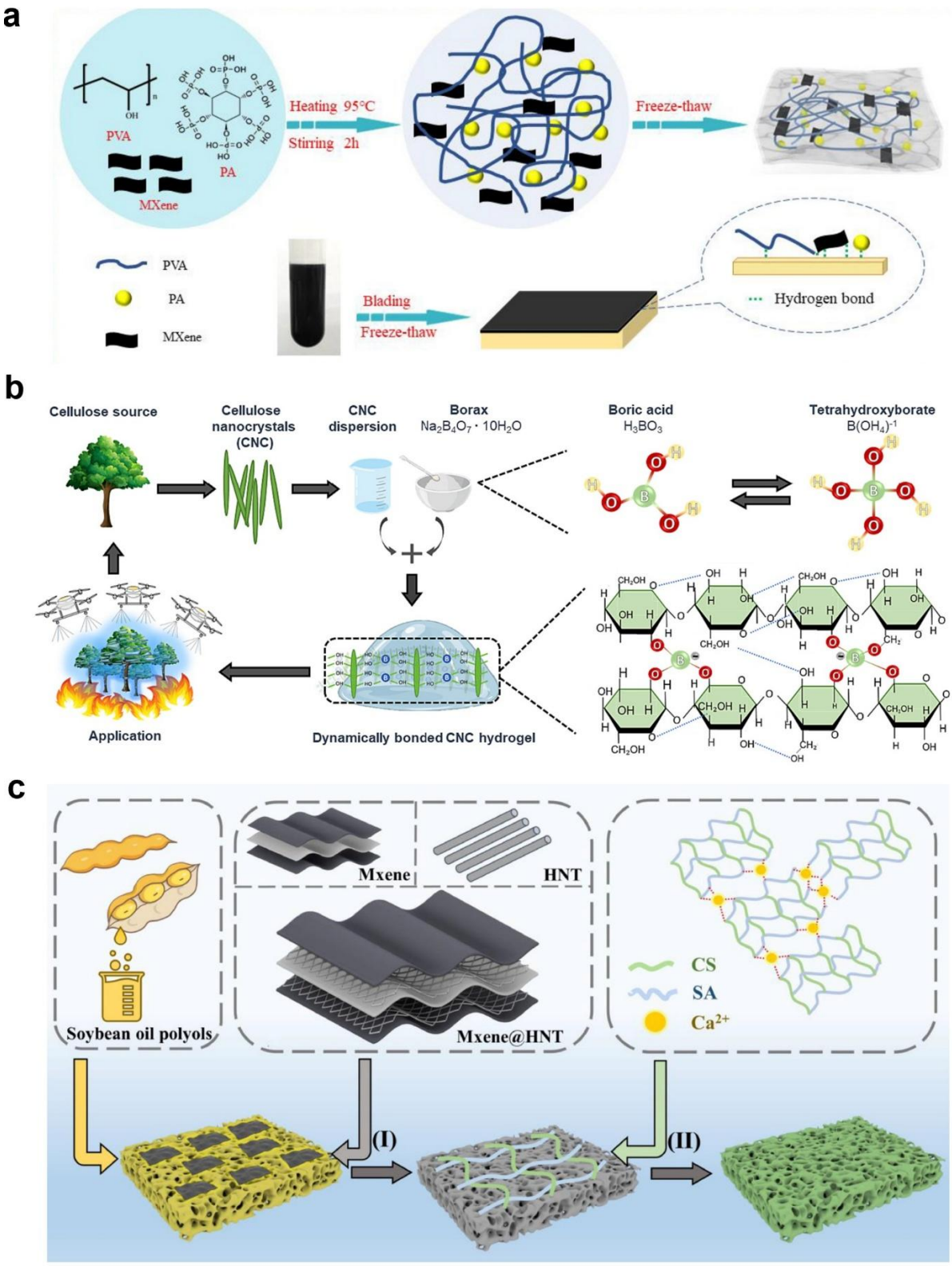


Figure 8. Preparation and flame-retardant mechanisms of inorganic filler-based hydrogels: (a) Illustration of the preparation process for the PVA/PA/MXene hydrogel and subsequent coating of this hydrogel on wood, adapted with permission from Ref. [73]. Copyright 2023, Springer

Nature. (b) Schematic of flame-retardant hydrogel synthesis and its use in preventing combustion of cellulosic materials. It includes the molecular structure of borax, ions formed in water, and the dynamic crosslinking mechanism between the CNCs and borate ions forming the hydrogel network, adapted with permission from Ref. [72]. Copyright 2024, Elsevier Ltd. (c) Schematic of the preparation of eco-friendly fire-retardant RPUF/M@H/DH: I) MXene@HNT forms the flame-retardant inner layer and II) a DH hydrogel forms the flame-retardant outer layer, adapted with permission from Ref. [142]. Copyright 2024, Elsevier Ltd.

6.5.Incorporating Carbon-Based Fillers as Flame-Retardant Additives

In this method, carbon-based nanofillers such as CNTs [70, 100, 143], graphite [62], and GO [100, 141, 144–146] are integrated into a hydrogel matrix to enhance the flame retardancy. For instance, a flame-retardant hydrogel known as a sodium alginate/graphene/carbon nanotube (SGC) hydrogel was developed using a 3D printing technique with a composite ink composed of SA, GO, and single-walled CNTs (SWCNTs), as shown in Figure 9a [100]. SA was dissolved in water at 90 °C, followed by the addition of GO, SWCNTs, and $CaCl_2$ as cross-linking agents. The mixture was extruded using a direct inkjet 3D printer and subsequently cross-linked with Ca^{2+} to enhance its structural stability. When exposed to a flame, the hydrogel released water via evaporation, and its outer layer carbonized because of the presence of GO and CNTs, forming a protective barrier.

In another study, a flame-retardant hydrogel was fabricated by combining Ag with GO to form a GPE (Figure 9b) [146]. A 4 wt% Ag solution was prepared in 2 M lithium acetate (LiOAc) at 90 °C, followed by the addition of GO (1–3 wt%). After stirring for 30 min and cooling in a polypropylene dish, a flexible Ag/GO-GPE membrane was obtained. GO improved the thermal stability and residual mass at 800 °C. During combustion, Li^+ ions were converted into lithium carbonate, which formed a protective layer that reduced oxygen infiltration and slowed flame propagation.

In a separate investigation, a flame-retardant hydrogel was developed by incorporating GO into a PAM hydrogel, cross-linked with nickel ions (Ni^{2+}), as shown in Figure 9c [145]. Functional groups in the GO formed hydrogen bonds with the PAM framework, and carboxyl groups in both components interacted with Ni^{2+}, which improved the mechanical strength and electrical conductivity. Upon exposure to fire, water evaporated endothermically, and the GO decomposed into reduced GO (rGO), which generated a protective char layer that blocked oxygen and heat transfer.

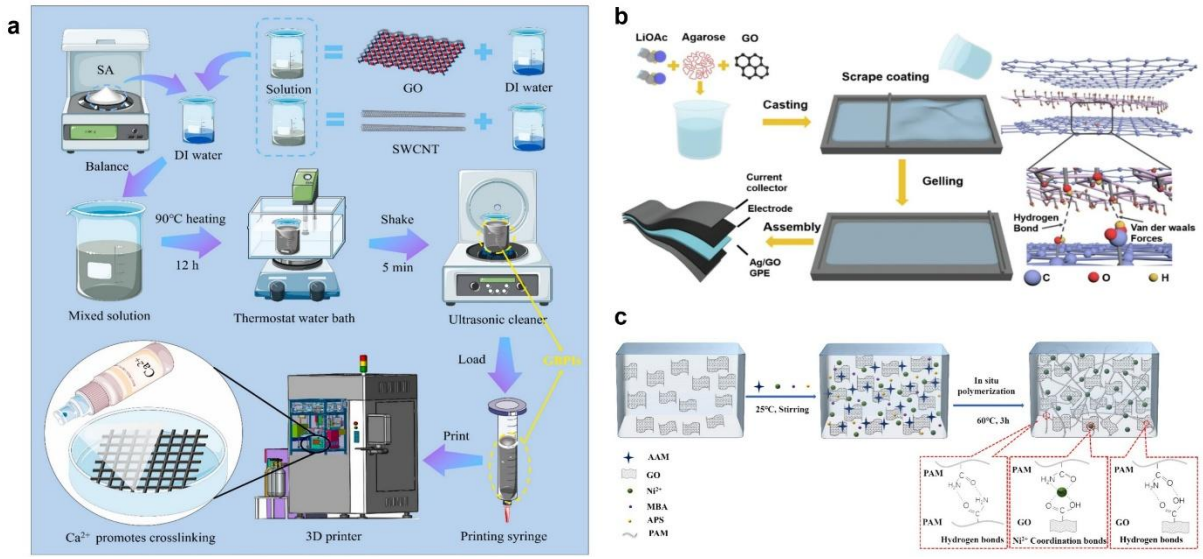


Figure 9. Fabrication and structural features of carbon-based flame-retardant hydrogels: (a) Schematic of the manufacturing process for SGC hydrogel using 3D printing, adapted with permission from Ref. [100]. Copyright 2023, Elsevier Ltd. (b) Preparation method of Ag/GO GPE, including casting, coating, and gelling steps, adapted with permission from Ref. [146]. Copyright 2023, Elsevier B.V. (c) Schematic of the design and in-situ polymerization of PAM/GO/Ni hydrogel, highlighting the hydrogen bonding and coordination interactions, adapted with permission from Ref. [145]. Copyright 2024, American Chemical Society.

6.6.Incorporating Hybrid Flame-Retardant Additive

In this method, a flame-retardant additive is first synthesized using at least two other additives and then incorporated into the hydrogel to improve its flame-retardant properties. Mahaninia

et al. developed a flame-retardant hydrogel from CS by using a vanillin-based phosphorus crosslinker (PA) to form a dynamic covalent network [128] (Figure 10a). The PA crosslinker, which was synthesized through the esterification of vanillin and phosphorus oxychloride, possessed aldehyde groups that reacted with the amine sites of CS via Schiff base reactions, resulting in imine (C=N) bonds within the hydrogel. In addition to CS, phosphorus-containing crosslinkers generated phosphoric acid derivatives upon heating. These derivatives enhanced the char formation and inhibited degradation, thereby improving the flame-retardant efficacy. The phosphorus also acted in the gas phase by deactivating oxygen radicals, thereby slowing the combustion process.

In another study, Xu et al. synthesized a dual-nucleophilic ester compound that combined TA and PA, referred to as T-P5 [60]. TA, cyanoguanidine, urea, dimethyl sulfoxide, and cyclohexane were mixed and heated under reflux at 105 °C in a nitrogen atmosphere. A solution of PA was gradually added, and the reaction was continued for 2 h. The resulting T-P5 product was precipitated, washed with ethanol, and dried under vacuum at 70 °C for 48 h. T-P5 was incorporated into a polyacrylic acid (PAA) hydrogel network (Figure 10b) via free-radical polymerization, where it interacted with the PAA chains through hydrogen bonding. This interaction imparted flame-retardant properties to the T-P5-PAA hydrogel. The T-P5 contained phosphorus groups that broke down at high temperatures, leading to the formation of phosphoric acid derivatives. These derivatives promoted char formation and inhibited free radical activity, thereby suppressing combustion. They also reduced flame spread, significantly improving the fire resistance.

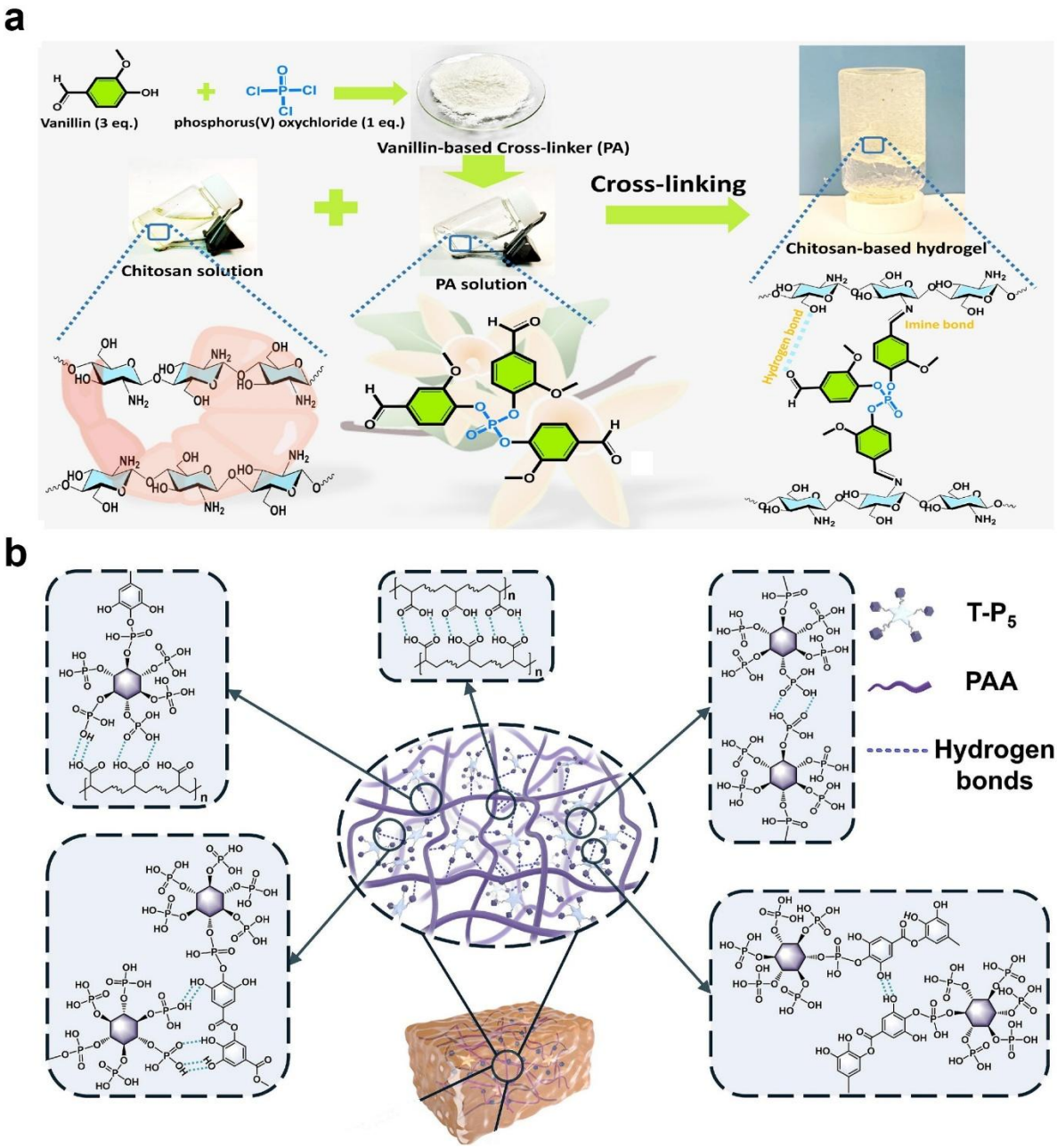


Figure 10. Flame-retardant hydrogels synthesized using dynamic covalent and hydrogen bonding crosslinkers: (a) Synthesis of a CS-based flame-retardant hydrogel (CSPA2) using a vanillin-derived phosphorus crosslinker (PA) through dynamic covalent bonding, adapted with permission from Ref. [128]. Copyright 2023, Elsevier B.V. (b) Fabrication of the T-P5-PAA flame-retardant hydrogel using the T-P5 crosslinker via hydrogen bonding with PAA, adapted with permission from Ref. [60]. Copyright 2025, Elsevier Inc.

6.7.Discussion

The strategies for creating flame-retardant hydrogels include the incorporation of various additives that increase the flame retardancy through synergistic mechanisms. These mechanisms include heat absorption, catalytic char formation, and the creation of physical barriers to enhance fire resistance while preserving the functional characteristics of the hydrogel.

Phosphorus-based additives, such as small molecules (e.g., PA) or larger compounds (e.g., APP), can be integrated into the hydrogel network via hydrogen bonding or covalent crosslinking. This integration promotes the formation of early-stage char and reduces combustion. In addition, flame-retardant polymers (e.g., CS, lignin, and alginate), inorganic fillers (e.g., MXene, silica, and borax), and carbon-based fillers (e.g., GO and CNTs) have been included in hydrogel systems to improve their thermal stability and mechanical strength.

Hybrid additives consisting of multiple functional components can further enhance the performance by combining distinct flame-retardant mechanisms within a single crosslinked framework. Collectively, these diverse strategies facilitate the development of hydrogels that can function effectively as surface coatings or bulk materials for various applications. These applications range from protective surfaces to energy-storage systems, in which achieving a balance between fire resistance, mechanical robustness, and multifunctionality is essential.

Table 3 provides an overview of the different types of additives, including phosphorus-containing substances, flame-retardant polymers, inorganic fillers, carbon-based fillers, and hybrid additives, detailing their integration methods, associated mechanisms, representative examples, and specific applications in both coatings and bulk materials.

Table 3. Overview of flame-retardant additives, including integration methods, their mechanisms, and representative examples for developing flame-retardant hydrogels.

Method	Additive Type & Examples	Integration Method & Mechanism	Representative Examples & Applications
Phosphorus-containing small molecules/compounds	PA [19, 137, 138], triethyl phosphate [136], and RDP [22].	Integrated through non-covalent interactions (such as hydrogen bonding) in the hydrogel matrix. They serve as catalysts for the carbonization process and enhance the formation of char at an early stage to postpone ignition.	PVA/PA hydrogel layer applied to wood [19]; PAM/PUL/PA-FRGPE for zinc ion energy storage devices [81]; hydrogel for skin protection made from RDP-coated starch [22].
Phosphorus-containing long-chain compounds	APP [135] and MAPP [139].	Incorporated via covalent crosslinking, the additives promote intumescence and create	MAPP@alginate/P(AAm-co-SMA) hydrogel [139]; PAAm-APP

		a char layer that shields the substrate during burning.	hydrogel strengthened with basalt fibers [135].
Flame-retardant polymers	CS [83, 84, 127, 128, 130], lignin [90, 131], starch [132], alginate [66, 87, 101], agarose [85], and carrageenan [86].	Incorporated through either covalent or non-covalent crosslinking, these polymers improve flame retardancy by encouraging char formation and emitting non-flammable gases during combustion.	3D-printed hydrogels (IAE/GEV/SAV) utilizing chemically-modified natural polymers [102]; PAAm–PDA hydrogel layer for rigid polyurethane foam (RPUF) [21].
Inorganic fillers	SiO_2 [120], MXene [120], kaolin [134], bentonite [141], basalt [135], borax [72], lithium acetate [85–87], composite fillers (e.g., MXene@HNTs [142] and mineralized black phosphorus@silica NF [58])	Dispersed throughout the hydrogel matrix to ensure thermal stability and create a physical barrier. They enhance cooling (through water evaporation) and aid in the formation of strong char.	Coating wood with a hydrogel made of PVA/PA/MXene [73]; hydrogels derived from CNC and borax [72]; MXene@HNT composites utilized on RPUF in the form of eco-friendly fire-

			retardant RPUF/M@H/double network hydrogel [142].
Carbon-based fillers	CNTs [70, 100, 143], graphite [62], and GO [100, 141, 144–146]	Incorporated into the hydrogel framework to improve char development during carbonization and minimize heat transfer, thereby creating a protective shield when exposed to flames.	SGC hydrogel (SA, GO, SWCNT) produced through 3D printing [100]; agarose/GO composite gel polymer electrolytes [146]; PAM/GO/Ni hydrogel [145] designed for enhanced mechanical and flame-resistant characteristics.
Hybrid flame-retardant additives	Hybrid additives synthesized from multiple components (e.g., vanillin-based phosphorus crosslinker [128], T-P5 from TA and PA [60])	Create dynamic networks through covalent or hydrogen bonding (for instance, using Schiff-base reactions) that integrate various flame-retardant strategies like char formation, deactivation of	Hydrogels made from chitosan (CSPA2) utilizing phosphorus crosslinkers derived from vanillin [128]; T-P5-PAA hydrogels that incorporate dual-nucleophilic ester compounds [60] to increase

		free radicals, and inhibition in the gas phase.	char formation and reduce combustion.

7. Flame-Retardant-Related Performance of Flame-Retardant Hydrogels

The performance indicators of flame-retardant hydrogels can be categorized into two groups: flame-retardant-related and non-flame-retardant-related, as shown in Figure 11. The evaluation of flame-retardant hydrogels requires a comprehensive assessment of both categories.

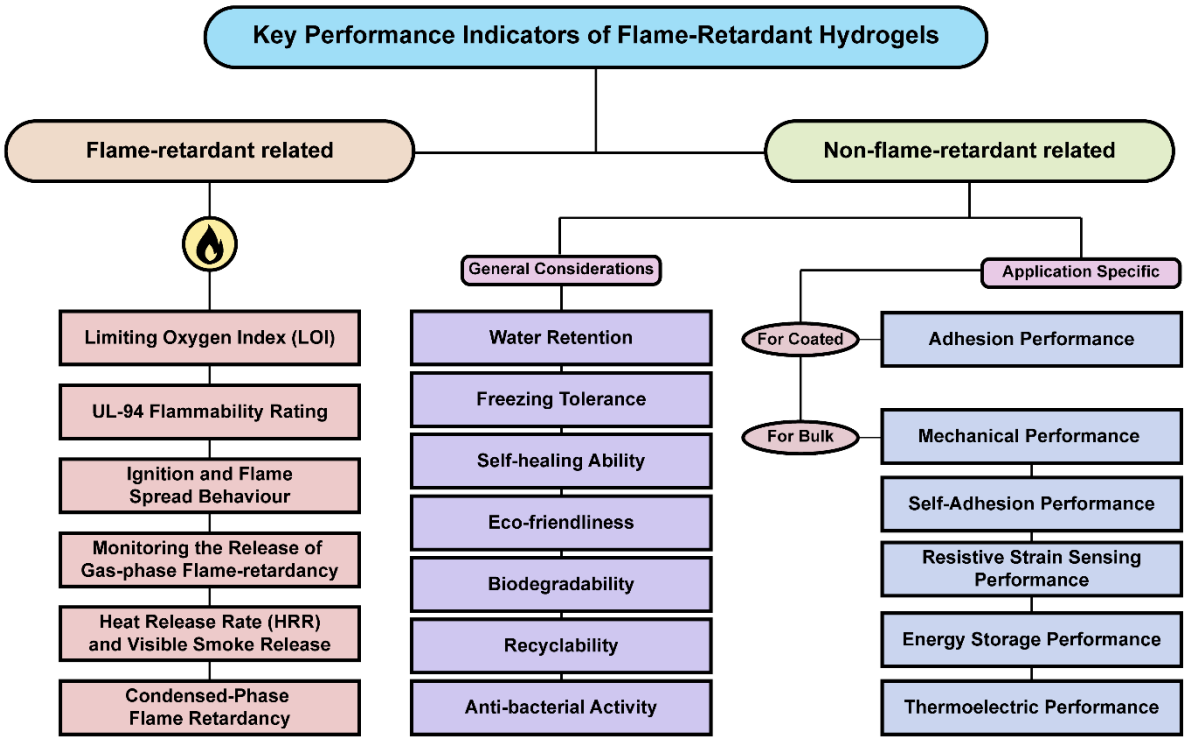


Figure 11. Key performance indicators for flame-retardant hydrogels include both flame-retardant- and non-flame-retardant-related indicators, which apply to hydrogel coatings as well as bulk forms.

Flame-retardant-related performance indicators directly measure the ability of a hydrogel to resist ignition, slow flame spread and reduce fire hazards. These indicators are applicable to both flame-retardant hydrogel coatings and bulk flame-retardant hydrogels. Collectively, these indicators enable a comprehensive evaluation of the flame-retardant performance of hydrogels. The key indicators include the limiting oxygen index (LOI), ignition and flame-spread behavior, Underwriters Laboratories Standard 94 (UL-94) flammability rating, heat-release rate, visible smoke release, and thermal-insulation capacity. In addition,

condensed-phase flame retardancy can be assessed based on the maximum degradation temperature, char formation, and residual mass percentage. Gas-phase flame retardancy is also monitored.

The LOI is a measure of the minimum oxygen concentration required to sustain combustion in a hydrogel; higher values indicate better flame retardancy. ASTM D2863 provides a standard method for testing the LOI in polymer-based materials [147, 148]. Hydrogels typically exhibit high LOI values, often exceeding 70%, because of their high water content and the formation of an insulating char layer [60, 75].

The UL-94 flammability rating classifies hydrogels based on their flame resistance, following the protocols outlined in ASTM D3801 for vertical position and ASTM D635 for horizontal position [127, 149–151]. These methods provide useful information on ignition time, flame-spread rate and behavior, dripping behavior, and fire-retardant class (V-0/V-1/V-2 ratings). Longer ignition times and slower flame-spread rates indicate better flame retardancy [58, 74, 134].

The Heat Release Rate (HRR) test evaluates the flame retardancy of a hydrogel by measuring the heat released during combustion and monitoring smoke production, in accordance with ASTM E1354, using a cone calorimeter [131, 134, 151]. The key parameters include the heat release rate (HRR), which denotes the instantaneous rate of heat release during combustion; the peak heat-release rate (pHRR), which indicates the maximum combustion intensity; and the total heat release (THR), which represents the total energy emitted. Flame-retardant hydrogels typically exhibit low HRR, pHRR, and THR values, delayed ignition, and high char yields, indicating reduced flammability [41, 62, 75, 78].

Visible smoke release is evaluated by attenuating a light beam with a photodetector to calculate the smoke production rate (SPR) and total smoke release (TSR). A lower optical

density indicates reduced smoke generation, indicating better visibility and lower toxicity risks for the environment [60, 75, 152].

A stable condensed phase of a flame-retardant refers to the solid residue left on the surface of the material after burning. This mainly consists of char, inorganic compounds, or protective coatings formed during thermal degradation, which contribute to flame-retardant behavior. Monitoring the condensed-phase flame retardancy in hydrogels involves evaluating three key factors: the maximum degradation temperature (T_{max}), char formation, and residue percentage [65, 66, 74, 98]. The T_{max} can be studied using Thermogravimetric Analysis (TGA) with Derivative Thermogravimetry (DTG) curve. A higher value of T_{max} is favorable for the flame retardancy of a hydrogel [23, 90]. The quality of char layer formation is assessed using a Scanning Electron Microscope (SEM) to monitor compact char or porous/cracked char. The compact char indicates the stable condensed phase [62, 65]. The residue percentage in TGA indicates char yield if TGA is performed in an N_2 gas atmosphere, and ash (inorganic compounds) if performed in air. Both types of residues contribute to flame retardancy; however, char yield specifically relates to the condensed phase [60].

Monitoring gas-phase flame retardancy involves analyzing the volatile combustion products that contribute to flame suppression [153]. These non-combustible gases interfere with the combustion chemistry by releasing inert gases, such as water vapor, CO_2, and NH_3, or active radicals, such as phosphorus- or halogen-containing species. These substances effectively quench flame-propagating radicals, such as hydrogen ($H^{\bullet}$) and hydroxyl ($OH^{\bullet}$) radicals. Thermogravimetric analysis (TGA) combined with Fourier-transform infrared spectroscopy (FTIR), known as TG-FTIR, is commonly used to monitor and identify the gases released during decomposition [62, 84, 98, 102].

7.1.Evaluation of Flame-Retardant Performance of Hydrogel Coating – An Example

The flame-retardant performance of hydrogel-coated bamboo (HB) shows significant improvement over untreated bamboo (OB), as illustrated in Figure S1. The cone calorimetry results (Figure S1a and S1b) indicate a 20.3% reduction in pHRR and a 53.1% decrease in THR for HB. This enhancement is attributed to the formation of a protective char layer, which is catalyzed by phosphoric acid from APP. Although HB initially produces more smoke (Figure S1c and S1d), this is due to the release of incombustible gases such as NH_3 and H_2O, which help to dilute flammable vapors. The TGA shown in Figure S1e reveals that HB decomposes in four stages and retains 46.4% char, compared to 25.9% for OB. The TG-IR spectra (Figure S1f) confirm reduced flammable gas release and the presence of NH_3 in HB. The LOI increases from 22.5% in OB to 55.9% in HB (Figure S1g), indicating improved flame resistance. Thermal insulation tests (Figure S1h) demonstrate that HB heats up more slowly than OB, confirming its effectiveness in resisting heat transfer. [75]. The improvements arise from the roles of each component: the hydrogel absorbs heat through evaporation, ammonium polyphosphate produces phosphoric acid and non-flammable gases that dilute oxygen, and the polymer network forms a char barrier that blocks heat and gas diffusion. Optimal performance relies on balancing heat absorption (water content), gas-phase inhibition (phosphorus-nitrogen compounds), and condensed-phase protection (char formation). Component selection would align with the primary hazard: thermal insulation benefits from high water retention, fire suppression needs gas-releasing phosphorus, and structural fire protection is enhanced by char-forming polymer-silicon networks. Thus, the best formulation depends on the specific application, prioritizing tailored mechanisms over single additives.

7.2.Evaluation of Flame-Retardant Performance of Bulk Form Hydrogel – An Example

The flame-retardant properties of bacterial cellulose (BC)/SiO_2 composites, as depicted in Figure S2, show a notable enhancement compared to pure BC nanofiber membrane (BCNF). Thermogravimetric analysis (Figure S2a) indicates that both materials maintain approximately

40% residue at 500 °C, but BC/SiO_2 exhibits superior char stability. In alcohol lamp experiments (Figure S2b, S2c), BCNF burns fiercely, whereas BC/SiO_2 extinguishes itself once the ignition source is removed. Cone calorimetry (Figure S2d, S2e) reveals a delayed ignition (from 25 s to 181 s) and a decrease in total heat release (from 29.68 MJ/m² to 21.99 MJ/m²) for BC/SiO_2. These findings confirm that SiO_2 significantly improves the flame resistance of the composite [154]. The performance improvement of the hydrogel-derived laminate is mainly due to the in-situ formation of SiO_2 at the interfacial gelation layer. This silica network, created from water glass and tannic acid, serves as a protective barrier that slows heat transfer, suppresses volatilization, and limits combustion. For hydrogel design, it is crucial to select components that form an inorganic protective phase without degrading the polymer network, specifically a polyphenol–silicate system that generates fire-resistant SiO_2. The optimal composition varies by application: higher silica is preferred for fire-safety materials, while applications requiring flexibility may need a balanced inorganic content to prevent brittleness.

7.3.Discussion

Based on flame-retardant-related performance indicators, flame-retardant hydrogels show strong performance across these tests due to their combined condensed-phase and gas-phase mechanisms. In LOI, the protective char layer (condensed phase) acts as a physical barrier, slowing the release of flammable gases and producing non-combustible gases (non-combustible gas phase) when exposed to fire [13]. The non-combustible gases such as H_2O, CO_2, and NH_3 dilute the oxygen concentration, therefore preventing O_2 from sustaining combustion [23]. This can be attributed to the crosslinking between flame-retardant additives and the hydrogel network [13].

In UL-94 vertical burning tests, the hydrogel-coated substrate achieves a V-0 rating after flame application, indicating fire suppression and delay, primarily due to the high-water content in the hydrogels. The absorbed heat causes water to evaporate, cooling the surface and

diluting the released gases [10, 19]. Similarly, cone calorimetry further confirms fire retardancy, as evidenced by reduced pHRR and THR values, indicating extended time to peak burning and reduced overall fire risk. Among the parameters, water evaporation plays a major role in the start, and the carbonized char layer formed by phosphorus or nitrogen-containing molecules plays a major role in the later stages. In hydrogels composed of inorganic additives, the physical barrier effect of a dense char can further improve flame retardancy by stabilizing the residue and suppressing secondary HRR peaks [73, 125, 142].

The main mechanism for enhanced flame retardancy is char formation, which acts as a persistent thermal barrier by restricting heat transfer and oxygen diffusion, reducing the release of flammable volatiles, and slowing thermal decomposition. An early and dense char layer improves flame resistance, and higher residual char yields generally correlate with superior thermal stability and prolonged protection during combustion testing [66, 155]. Among the gas-phase contributors, H_2O and NH_3 play distinct but complementary supportive roles during the initial stage of flame exposure. Water evaporation absorbs latent heat, providing immediate surface cooling and diluting combustible gases near the substrate interface, thereby delaying ignition [13]. NH_3, released from nitrogen-containing functional groups (e.g., $–NH_2$, NH_4^+) upon thermal decomposition, acts as a non-combustible gas-phase diluent by lowering the local oxygen concentration in the combustion zone [76]. Both effects are transient; once water has evaporated and NH_3 release is complete, sustained flame retardancy depends on the condensed-phase char layer [13, 42, 128]. Overall, char formation constitutes the dominant and long-lasting mechanism, whereas H_2O and NH_3 release serve as early-stage gas-phase contributors that delay ignition and retard initial flame spread.

7.4.Challenges in UL-94 and Cone Calorimetry Tests for Flame-Retardant Hydrogels

The UL-94 and calorimetry tests were developed for solid polymers that maintain their shape. On the other hand, hydrogels are polymer-based materials with over 50% water content,

making them soft and unable to retain rigid shapes. Testing their fire-retardancy directly could lead to misleading results due to their water-rich nature [41]. The UL-94 test requires a self-supporting hydrogel bar specimen, but hydrogels can deform under gravity and heat, which can affect fire-retardant performance. It is recommended that the issue be resolved by supporting the hydrogel bar specimen with a metal mesh frame or other metallic structures [73, 100].

Hydrogels also present a challenge due to their high-water content, which may result in a virtual V-0 rating from significant water evaporation, even if the polymer network itself is flammable [156]. The same challenge can also be encountered in cone calorimetry due to the water-rich nature of hydrogels [13]. True evaluation of a hydrogel's flame retardancy starts only after water has evaporated. To enhance accuracy, it is advisable to conduct the UL-94 and cone calorimetry tests on the hydrogel in both hydrated and dried states separately. This approach would help differentiate the effect of water on fire delay and provide an accurate response of the polymer content to fire [41, 157].

8. Non-Flame-Retardant-Related Performance and Multifunctional Aspects of Flame-Retardant Hydrogels

Aside from flame-retardancy, hydrogels must exhibit a wide range of other non-flame-retardant properties to ensure the mechanical, structural, and other functional abilities that are crucial for real-world applications. These performance indicators, summarized in Figure 11, include general considerations for determining the longevity of hydrogels and other application-specific functionalities for both coated and bulk hydrogels. These influential factors play a crucial role in enhancing the functional durability of flame-retardant hydrogels, ensuring their resilience and effectiveness under various conditions. The effectiveness of flame-retardant hydrogels depends not only on their intrinsic flame resistance but also on their ability to maintain structural integrity, adhesion, and various other functional properties before exposure

to fire [158]. Specifically, these additional properties increase confidence in the practical application of these hydrogels.

On the other hand, in this review, multifunctional aspects refer to the combination of two or more non-flame-retardant-related functions or properties, with the main flame-retardant-related performance being essential. Multifunctional aspects are essential in flame-retardant hydrogels for two main reasons: (1) they enhance the performance and versatility, and (2) they broaden the application scope [159]. First, multifunctionality enhances performance and versatility: properties such as adhesion, mechanical stability, water retention, self-healing, and sensing capability ensure that the hydrogel remains attached, intact, and functional under thermal exposure [160]. Without these supporting functions, even a highly flame-resistant hydrogel may crack, detach, dry out, or fail prematurely, allowing heat and oxygen to reach the substrate. Thus, auxiliary properties reinforce the effectiveness of the flame-retardant mechanism itself by preserving barrier continuity over time. Second, multifunctionality broadens the scope of applications [160]. Different applications require different operational conditions—for example, coatings need strong adhesion and durability, wearable safety devices require flexibility and biocompatibility, and smart fire-warning systems demand electrical responsiveness [161]. By integrating multiple non-flame-retardant functions with flame resistance, hydrogels can be tailored to specific environments, increasing user confidence and enabling their use across various applications.

8.1. Non-Flame-Retardant-Related Performance of Flame-Retardant Hydrogels

8.1.1. Adhesion Properties

Substrate adhesion is crucial for flame-retardant hydrogel coatings, as it ensures the coating remains intact during fire exposure and provides a uniform protective barrier [21, 73, 96]. Strong adhesion prevents delamination, enhances durability under thermal and mechanical stresses, and effectively blocks heat and oxygen. If the adhesion fails, the flame-retardant performance

of the coating is significantly reduced [19, 60, 75]. For example, Xu et al. assessed the adhesive characteristics of a thermosensitive ionic hydrogel, Gel/PAM/GLY/TA/NaCl (gelatin/polyacrylamide/glycerol/tannic acid/sodium chloride, PGTS), which exhibited flame-retardant properties [162]. The findings of this research are presented in SI Figure S8 (Section S2). The gel electrolyte exhibits stable electrochemical behavior under bending, thereby ensuring strong interfacial contact with the electrodes and preventing gaps that could lead to heat transfer or ignition. Hydrogel formulations should employ dynamic yet robust bonding chemistries to ensure adhesion and structural integrity. Optimal compositions vary by application: fire-protective formulations require stronger adhesion to maintain barrier integrity, whereas flexible wearable electronics benefit from softer networks for improved conformability.

In the context of devices, self-adhesion enables a hydrogel to adhere to a surface without additional adhesives, thereby improving its applicability in wearables, sensors, and capacitors. Coupled with durability and flexibility, this property ensures that hydrogels can endure stress, maintain functionality, and reliably sustain electronic performance [81, 137, 145]. For instance, Zhang et al. evaluated the self-adhesive performance of bio-based hydrogel electrolytes (PSBGL) composed of peach gum polysaccharide (PGP) and sisal nanofibers (NFs) for use in flexible SCs [82]. The details of their research findings are provided in SI Figure S10 (Section S3.2). The hydrogel achieves strong self-adhesion and flame retardancy by combining dynamic interfacial bonding with char-forming components. Hydrogel design should integrate multifunctional groups to provide surface bonding and thermal barriers, rather than addressing these properties separately.

8.1.2. Longevity and Sustainability Aspects

When assessing the longevity of a hydrogel, it is crucial to consider factors such as the water retention [23, 99], freezing tolerance [16, 84, 123, 124, 163], eco-friendliness [1, 148], biodegradability [64,

[164], recyclability [61, 83, 127, 165, 166], and antibacterial properties [36, 116, 118, 126, 148, 167]. These features enhance the durability and sustainability of a hydrogel, making it more reliable and commercially viable for various applications [13].

Water retention, which provides cooling during a fire [23, 99]; freezing tolerance, which makes it suitable for operation in a cold environment [163, 165]; and self-healing capabilities, which enable the repair of minor damage to restore function [73, 80]. For example, Yang et al. evaluated the water-retention capacity of gelatin/poly(acrylamide-*co*-acrylic acid)/lithium bromide/sodium phytate/glycerol (GLY-GAPL) hydrogels [74]. The details of their findings are presented in SI Figure S3 (Section S1.1), which shows a significant enhancement with the addition of GLY and PA-Na. Zuo et al. reported the freezing tolerance of IA/cellulose-based hydrogels [163], as shown in SI Figure S4 (Section S1.1). The performances of IAB and IAB/HPCS hydrogels, as illustrated in SI Figure S4, highlighted their excellent stability under extremely cold conditions. Zhu et al. investigated the self-healing behavior of biomass hydrogel electrolytes [80], as shown in Figure S5 (Section S1.1). These studies demonstrate that hydrogel performance is governed by a dense, multi-interaction network that minimizes dehydration and freezing, while enabling self-healing. This indicates improved performance results from a synergistic design that provides anti-freezing, moisture-retaining elements and dynamic non-covalent interactions. The ideal composition varies with the intended application, including freezing tolerance, moisture retention, and self-healing, without sacrificing the flame-retardant performance.

In addition, eco-friendliness, biodegradability, and recyclability contribute to environmental safety and sustainability [61, 83, 102, 127, 134]. Eco-friendly materials reduce emissions and are less dependent on nonrenewable resources. Biodegradable materials break down naturally without generating toxins, whereas recyclable materials help conserve resources and reduce landfill waste [102, 127, 130]. For instance, Xu et al. investigated the

recyclability of dual-crosslinked CS-LA-$B(OH)_4Na$ GPEs [83]. The details of their findings are presented in SI Figure S6 (Section S1.2). The dual-cross-linked chitosan gel electrolyte retains its electrochemical performance even after damage, thanks to reversible imine bonds and borate ester interactions. This dynamic network allows for structural reconstruction without disrupting ion transport. Hydrogel design should incorporate reversible network-cross-linking chemistries for reprocessing while maintaining conductivity and mechanical integrity. However, the optimal composition varies by application: energy storage devices benefit from recyclable dynamic networks for enhanced sustainability, whereas strong, irreversible cross-linking networks are prioritized for greater dimensional stability over recyclability.

8.1.3. Antibacterial and Biocompatible Properties

Antibacterial properties help prevent microbial degradation, which increases the lifespan of a flame-retardant hydrogel [23, 66, 128, 148, 167, 168]. Biocompatibility is crucial for flame-retardant hydrogels in medical applications, because it ensures that they will not cause toxic or inflammatory reactions in biological tissues [79]. While flame resistance is important, the material must also be nontoxic and safe for the body, especially in applications such as wound dressings or implants, where both fire protection and biological safety are essential [22, 128]. Mahaninia et al. developed CS-vanillin hydrogels with flame-retardant properties, significant antibacterial activity, and biocompatibility, making them suitable for applications involving skin contact [128]. Their research findings are detailed in SI Figure S7 (Section S1.3), which together demonstrate antimicrobial activity and cytocompatibility. The performance improvement stems from a balanced structure: bio-derived chitosan provides safety and antibacterial action, while a vanillin-based crosslinker enhances antimicrobial functionality and bonding. Optimized crosslink density governs adhesion, hydration, and cellular compatibility. These findings suggest hydrogel design should focus on synergistic component selection, prioritizing biopolymer compatibility, functional crosslinkers, and controlled network density.

The optimal formulation varies by application; for example, skin-contact coatings require balanced antimicrobial and biocompatible properties, while structural fire barriers may prioritize thermal resistance.

8.1.4. Mechanical performance

The functionality of bulk flame-retardant hydrogels is closely related to their mechanical properties, such as their strength, toughness, flexibility, and stretchability [169]. In applications such as sensors and energy-storage devices, factors such as the electrical conductivity and thermoelectric performance are crucial [162, 170]. Self-adhesion can further enhance the functionality for specific applications. These properties ensure durability and versatility, enabling the hydrogel to withstand physical stress, support electronic performance, and effectively bond to target surfaces.

The key mechanical properties of bulk flame-retardant hydrogels include their strength, toughness, flexibility, and stretchability. Strength refers to the resistance to breaking, whereas toughness indicates the ability to absorb energy and resist cracking. Flexibility allows for bending without damage, making the material suitable for movement or shape changes. Stretchability is the extent to which a hydrogel can elongate without losing integrity, a property crucial for wearable or flexible devices. These properties ensure their durability and functionality under mechanical stress [71, 98, 171]. For example, mechanical performance is critical for the use of flame-retardant hydrogels in fire-safety systems. Zaidi et al. evaluated the mechanical properties of CS-reinforced flame-retardant hydrogels for supercapacitor applications [84]. The findings of this study are shown in SI Figure S9 (Section S3.1). The results indicate that the composite hydrogel offers improved mechanical strength and excellent flame retardancy. This enhancement occurs because the network structure reinforces the matrix without interfering with the flame-retardant mechanism; the polymer framework provides mechanical integrity, while the chemical composition promotes protective char formation

during burning. Thus, hydrogel formulations should integrate structural reinforcement with char-forming or thermally stable components instead of relying on a single additive.

8.1.5. Applications-Specific Functionalities

Resistive hydrogel strain sensors are crucial for wearable electronics owing to their straightforward design, flexibility, and high sensitivity. These sensors operate by altering their electrical resistance when stretched or compressed [137, 140]. Unlike capacitive or piezoelectric sensors, resistive hydrogel sensors are easier to fabricate, respond effectively to static and dynamic strains, and function reliably in wet, cold, and biocompatible environments. Their applications include motion tracking, health monitoring, soft robotics, and human–machine interaction [25, 172]. Most flame-retardant, strain-sensing hydrogels rely on resistive strain-sensing mechanisms [137, 140, 145, 172]. For example, Li et al. evaluated the strain-sensing capabilities of a multifunctional CNT-based acrylamide-acrylic acid copolymer hydrogel, designated GEL-CNT-ILS. This novel copolymer gel was synthesized using CNTs, acrylamide, and acrylic acid [140]. Their detailed research findings are presented in SI Figure S11 (Section S3.3).

In energy-storage devices, such as hydrogel batteries and SCs, several key properties are essential for optimal performance [161]. These include a high specific capacitance or energy density, which allows compact energy storage, and high electrical conductivity to facilitate efficient electron transport [80, 83]. In addition, fast charge–discharge rates are important for rapid energy transfer. To ensure smooth ion transport, hydrogel materials must exhibit excellent cycling stability and high ionic conductivity. Furthermore, a wide electrochemical stability window is necessary to ensure safe operation over a broad voltage range [173, 174].

Recent studies have highlighted the potential use of flame-retardant hydrogels as electrolytes for safe SCs [80, 82–88, 101, 136, 146, 175], particularly ZICs [81]. The details of these research findings are illustrated in SI Figure S12 (Section S3.4). Furthermore, hydrogel-based

battery components with flame-retardant properties hold promise for energy-storage applications, particularly as separators [176], electrodes [177], and electrolytes [136, 176, 178] in lithium-ion batteries (LIBs).

For example, Wang et al. investigated the feasibility of using a flame-retardant CG-PAM@PAN-SiO_2 quasi-solid-state hydrogel electrolyte (QSE) in Li||Li symmetric batteries [88]. The results of their study are presented in SI Figure S13 (Section S3.4). Moreover, flame-retardant hydrogels are safe alternatives to flammable commercial polyolefin-based separators during thermal runaway or short circuiting. They enhance the electrochemical performance by offering high porosity, wettability, and ionic conductivity [176, 177]. For instance, a flame-retardant hydrogel (melamine–PA supramolecular polymer assembly, MAPA) was used to construct an LIB anode, which enhanced the safety by reducing flammability [177]. Detailed findings are provided in SI Figure S14 (Section S3.4). When MAPA is used as an anode in lithium-ion batteries, it shows stability and specific capacity, which is attributed to the abundant functional groups, such as phosphate and π bonds of MAPA. Additionally, MAPA's flame-retardant properties are assessed using two types of widely used electrolytes: the quasi-solid gel electrolyte of solid supercapacitors (polyvinyl alcohol, PVA) and ethylene carbonate and dimethyl carbonate (1:1 vol, ECDC) for LIBs. Excellent flame-retardant properties are displayed by both the PVA/MAPA produced by electrospinning technology and the ECDC/MAPA produced by ultrasound sonication technology.

The thermoelectric properties of flame-retardant hydrogels allow them to function beyond fire resistance [161]. When heated, these hydrogels convert thermal energy into electrical energy, enabling them to act as self-powered fire or temperature sensors. This capability enables the hydrogel to generate voltage signals in response to rising temperatures, which can trigger alarms, shut down electrical systems, and send emergency alerts [171, 179]. Therefore,

these materials enhance the safety and responsiveness of fire protection systems, making them both protective and intelligent.

Furthermore, thermoelectric hydrogels can harness heat under fire-related conditions, thereby generating electricity for power-critical devices during emergencies. Their combined flame resistance and energy conversion capabilities make them suitable for wearable electronics and compact building materials, where both safety and energy efficiency are essential [170, 180, 181]. Li et al. developed a multifunctional CNT-based hydrogel (CNT and ionic liquid-doped gel, GEL-CNT-ILS) with excellent thermoelectric properties via radiation polymerization. Multi-walled CNTs and ionic liquids were incorporated into an acrylamide-acrylic acid copolymer gel [140]. The details of their research findings are presented in SI Figure S15 (Section S3.5). Radiation polymerization produces adjustable cross-linking, ionic liquids facilitate thermoelectric conversion via the Soret effect, and MWCNTs offer electrical conductivity and photothermal properties. The optimal formulation depends on the intended use: balanced compositions work well for multifunctional sensing and flame-retardant applications, greater absorption doses favor thermoelectric production, and GEL-CNT-ILS is excellent for desalination.

8.1.6. Discussion

The hydrogel performance is governed by the choice of polymer, its synthesis route, the type of fillers used, integration of NPs, and different hybrid compositions. The choice of polymer sets the mechanical baseline and determines intrinsic flame retardancy, sustainability, and biodegradability. In this regard, polysaccharide-based polymer matrices remain the most effective and widely accepted choice due to their abundant functional groups, biodegradability, and renewable origins [81, 82]. Crosslinkers are mainly responsible for water retention and improved char formation during combustion. Dual or hybrid crosslinking networks typically exhibit different performance characteristics, such as mechanical robustness, durability, and

flame resistance, whereas dynamic crosslinking strategies are particularly advantageous for providing self-healing, self-adhesion, and flame resistance [60, 68, 105, 117]. The use of green crosslinkers, such as phytic acid [19, 138] and calcium alginate (CA) [62], further strengthens their sustainability profile while simultaneously improving their multifunctionality, such as flame retardancy, adhesion, thermal insulation, and mechanical properties [19, 62, 138]. The filler selection is also important as it can introduce flexibility and flame retardancy through endothermic cooling and char formation. Notably, the integration of hybrid and NMs, such as bio-derived NPs [147] or Mxene@HNT [142] can provide enhanced mechanical strength, superior flame resistance, and improved overall thermal stability. The polysaccharide-based hydrogel engineered through hybrid crosslinking and reinforced with multifunctional nano- or hybrid fillers represents the most effective and sustainable strategy for achieving a balance of flame-retardant and non-flame-retardant properties.

8.2. Multifunctional Aspects of Flame-Retardant Hydrogels

8.2.1. Enhancing Performance and Versatility through Multifunctionality

Multifunctional aspects are crucial in flame-retardant hydrogels because they enhance the reliability in specific applications and the versatility across various fields. Multifunctional properties refer to additional characteristics beyond flame retardancy. Multifunctionality enhances the performance and versatility of hydrogels through properties such as adhesion, mechanical stability, water retention, self-healing, and sensing. These features ensure that the hydrogel remains intact and functional under thermal exposure [100, 158, 182]. Without them, even flame-resistant hydrogels may crack or detach, compromising their protective barrier. Thus, these properties enhance the effectiveness of the flame-retardant mechanism by maintaining barrier continuity over time.

For instance, in practical settings such as electronics, biomedical devices, and construction, flame resistance alone is insufficient. Additional properties, such as mechanical

strength, flexibility, anti-freezing ability, and conductivity, are also essential [84, 128, 137]. By integrating these features, hydrogels can withstand harsh conditions, serve multiple functions, and satisfy complex requirements. Table 4 summarizes recent studies on flame-retardant hydrogels that exhibit enhanced performance and versatility through multifunctionality. These features not only improve the performance reliability but also broaden their applicability, making them suitable for advanced and sustainable uses [80–87].

Zhu et al. reported a comprehensive green hydrogel electrolyte composed of SNFs, PGP, borax (B), glycerol (G or GLY), and zinc sulfate ($ZnSO_4$) salt in water. The resulting hydrogel electrolyte, SPBG-$ZnSO_4$, exhibited multifunctional properties, including flame retardancy, freezing resistance, and self-adhesion (Figure 12a)**,** which enhanced its suitability as a flame-retardant hydrogel electrolyte for SCs (Figure 12b). After dehydration, the PGP and SNFs enhanced the flame retardancy by forming a char layer and releasing non-flammable gases that assisted in suppressing combustion. Dynamic physical crosslinking through borate ester bonds and hydrogen bonds between the PGP and SNFs, provides a hydrogel with self-healing capability, even at a low temperature of -20 °C in the presence of salt (Figure 12c). Additionally, as shown in Figure 12d, the ability of SNFs and PGP to form hydrogen bonds with non-metallic substrates, along with their coordination with metallic substrates, endowed the hydrogel electrolyte with excellent adhesion, achieving an adhesion strength of approximately 10^2 kPa on various substrates. The presence of GLY significantly improved the dehydration resistance under various temperature and humidity conditions (Figure 12e).

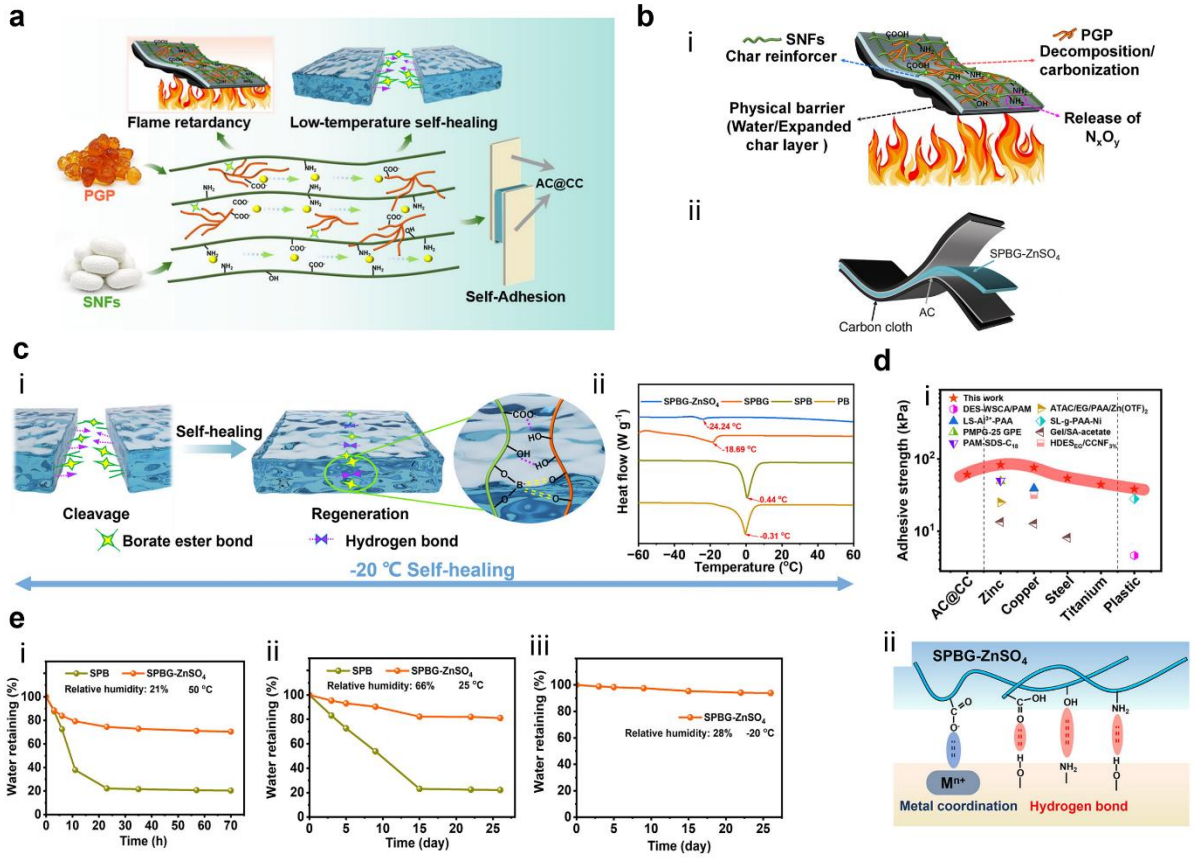


Figure 12. Multifunctional properties of SPBG-$ZnSO_4$ flame-retardant hydrogel electrolyte: (a) Multifunctional characteristics of the SPBG-$ZnSO_4$ hydrogel electrolyte, including its flame retardancy, low-temperature self-healing, and self-adhesion. (b)- i) Flame-retardant mechanism of the hydrogel electrolyte and, ii) its application in a SC device. (c)- i) Mechanism of self-healing and, ii) low-temperature resistance of the hydrogel electrolyte. (d)- i) Adhesion performances on various substrates and, ii) the proposed adhesion mechanism based on hydrogen bonding and metal coordination. (e) Water-retention behavior of the hydrogel electrolyte under various humidity and temperature conditions. Adapted with permission from Ref. [80]. Copyright 2024, American Chemical Society.

Table 4. Recent updates on flame-retardant hydrogels, showing enhanced performance and versatility achieved through multifunctionality.

Year	Flame-retardant hydrogel system	Properties beyond the flame retardancy	Flame-retardant additive	Properties contributed by the flame-retardant additive	Ref.
2025	MC/PAAm + SiO_2@APP (C-Si-P-N coating)	- High adhesion (125.9 kPa) - Abrasion resistance - Thermal insulation - Structural integrity under flame	SiO_2@APP (tetraethyl orthosilicate-treated APP)	- Reinforces mechanical structure - Contributes to thermal barrier via C-Si-P-N synergy	[75]
2025	CNC/lignin/borax	- Improved hydrogen bonding - Water retention - Rheology - Elasticity - Surface adhesion	Lignin and borax	- Lignin enhances char formation - Borax forms boronate esters, improves thermal barrier and gel strength	[183]

2025	Triazolium-containing PVA-borax hydrogel	- Antibacterial activity (>99% against E. coli and S. aureus) - Mechanical strength enhancement - Biocompatibility (low cytotoxicity at low concentrations)	Triazolium ionic salts	- Antibacterial activity - Improved mechanical strength - Higher water retention - Biocompatibility for skin contact - Thermal stability	[167]
2025	PAM/PUL/PA	- High tensile strength (92 kPa) - Stretchability (1535%) - Ionic conductivity (22.54 mS/cm) - Energy density (97.57 Wh/kg) - Long cycle life	PA	Hydrogen bonding improves mechanical strength and ionic conductivity, regulates Zn^{2+} deposition to prevent dendrites, and enhances adhesion and thermal stability	[81]

		- High adhesion - Thermal insulation			
2025	CPG-L_2 (CA/PAAm/GLY/(lithium bromide, LiBr)	- High thermoelectric performance (Seebeck coefficient of 9.31 mV/K) - Tensile strength (1.24 MPa) - Elongation at break (689%) - Fast fire response (0.07 s) and long warning duration (855 s) - Self-healing - Water retention	CA + water	Improves mechanical stability through cross-linking, enhances water retention, contributes to thermoelectric sensitivity, and enables self-healing via reversible bonding	[171]

2024	SPBG-$ZnSO_4$ (SNFs/PGP/borax/GLY with $ZnSO_4$)	- Low-temp self-healing at −20 °C - Tensile strength: 20.7 kPa - Elongation at break: 234.9% - Adhesion to carbon cloth: 60.7 kPa - Ionic conductivity: 7.68 mS/cm - Water retention: 81% after 26 days at 25 °C - Capacitance retention: 98.2% after 40,000 cycles	SNFs GLY $ZnSO_4$	- SNFs: char reinforcement (21.9% residue at 600 °C), nitrogen oxide release for flame suppression - GLY: water retention, mechanical flexibility - $ZnSO_4$: enhanced ionic transport, dehydration resistance, adhesion support	[80]
2024	IAB/HPCS hydrogel (IA-based + hydroxypropyl cellulose + PEGDA crosslinker)	- Tensile strength (95.2 kPa) - Surface temp (~94 °C) after 12 min	IA + HPCS	- Improved water retention - Enhanced mechanical strength due to hydrogen	[163]

		- Better water retention (only 56% water loss) - Freezing tolerance (–13 °C).		bonding and van der Waals interactions - Delays thermal decomposition as a result of water retention - Improves thermal insulation	
2024	Gly-GAPL [Gel/Poly(AAm-co-AA)/LiBr/PA-Na/ GLY]	- Seebeck coefficient (8.66 mV/K) - Tensile strength (up to 296.6 kPa) and elongation (~706.7%) - Adhesion strength (46.23 kPa) - Water retention (25 °C for 7 days and ≤14.2% mass loss)	LiBr and PA-Na	- LiBr: increases thermoelectric sensitivity via ion mobility and improves water retention by binding water molecules - PA-Na: enhances mechanical strength and thermal char formation, increases water retention,	[74]

		- Transparency (>82% at 800 nm thickness) - Fire-warning activation (<2 s)		and forms dense flame-retardant layers	
2023	Gel/CS_{30}/ammonium sulfate/ammonium chloride	- Mechanical toughness (3.81 MJ/m^3) - Fracture energy (26 kJ/m^2) - Anti-freezing (−30 °C) - Ionic conductivity (72 mS/cm at 26 °C, 39 mS/cm at −30 °C) - Capacitance (426 mF/cm^2 or 129 F/g), retention (−30 °C): 84% after 2000 cycles	CS precipitated in-situ via salt soaking	Mechanical reinforcement: improves modulus, toughness, and fracture energy	[84]

2023	CS-based dynamic covalent hydrogel crosslinked with vanillin-derived phosphorus agent (CSPA2 hydrogel)	- Self-healing (Complete healing in ~30 min) - Adhesion strength (23.2 ± 3.0 kPa) - Thermal insulation (0.29 ± 0.03 $W^{-1}mK^{-1}$) - Antimicrobial activity (~99% S. aureus, ~80% E. coli kill rate) - Biocompatibility (Enhanced HaCaT cell growth over 5 days)	Vanillin-derived phosphorus-containing aldehyde crosslinker	- Self-healing via imine bonds - Antimicrobial effect due to phenolic and imine groups - Enhanced thermal insulation (retains unbound water) - Improved adhesion via π–π stacking and hydrogen bonding - Biocompatibility due to natural vanillin origin	[128]
2023	PVA/borax	- Self-healing (complete reformation in 1–9 min depending on formulation)	Borax	- Enables borate ester bond formation for 3D network - Enhances self-healing and elastic properties	[61]

		- Recyclability (crushed hydrogels reformed into a single piece)		- Provides recyclability as a result of reversible bonding	
2023	PVA/PA/MXene coating	- Self-healing without external stimulus - Water content ≥ 90% retention after 15 days - Swelling ratio up to 31.08% - Mechanical strength (stress: 0.124 MPa, strain: 310%)	MXene ($Ti_3C_2T_x$ nanosheets)	- Improves self-healing via hydrogen bonding - Enhances water-retention and swelling - Increases mechanical strength - Physical barrier: improves char layer compactness and thermal stability - Improves adhesion to wood surface	[73]
2022	P(DPOP-Si)	- Adhesion strength: up to 3.5 MPa (glass), 1.6 MPa	Diphenylphosphine oxide (DPOP), as part of the siloxane monomer	- π-π stacking: enables strong, reversible cohesive energy and transparency	[165]

	A phosphorus-doped aromatic siloxane	(steel), 1.4 MPa (wood), 200 kPa (Teflon) - Durability (stable under pH 1–14, −10 to +90 °C, and 85% humidity) - Recyclability (100% recovery and >100 reuse cycles) - Transparency: 98% in 400–800 nm range and low haze (~2.5%) - Coating durability: 97% of weight retained after 50 machine wash cycles		- Hydrolytic stability: water-insensitive interactions - High durability: retains mechanical and structural properties under high humidity, acid/base, and temperature - Recyclability: enables ethanol-responsive disassembly/reassembly mechanism for closed-loop use	

2022	PAA-SA-TA	- Water retention: ~10% higher than PAA-only hydrogel - Swelling ratio (g/g): 151 - Air-leakage blocking: 42.9% better than PAA	SA and TA	- Enhanced self-adhesion to coal, glass, or gravel - Improves water absorption and retention - Forms hydrogen bonds for strong adhesion - Enables dense surface coverage for air-leakage blocking and fire suppression	[54]
2022	SiO_2/PAAm + PA-doped PPy + GLY	- Stretchability: 1896% at RT; 1883% at −20 °C - Adhesion: up to 79.7 kPa - Conductivity: 0.11 S/cm - Gauge factor (GF): ~2.90 across 0–500% strain range	PA	- Improves conductivity through doping of PPy - Enhances adhesion via PA–Fe^{3+} complexation - Maintains hydrogel flexibility and conductivity after burning	[137]

		- Stable operation from −20 °C to +600 °C		- Reduces heat transfer and surface temperature (71.2 °C after 1200 s at 200 °C)	
2021	PVA + PA	- Adhesion strength: 151.68 kPa (↑179.96%) - Tensile strength of hydrogel (0.96 MPa), maintains wood tensile strength (~25 MPa) - Antibacterial effect (no colonies on day 5 for 2 mm coating) - Thermal insulation: delay in back-surface temp rise to 150 °C in 53 s	PA	- Enhances adhesion to wood via hydrogen bonding - Improves hydrogel mechanical strength - Provides antibacterial properties	[19]

2021	PNIPAAm/SA/silver NP hydrogel with $CaCl_2$, laminated on cotton fabric	- Water retention: up to 19.6% at 3000 min with 7% $CaCl_2$ - Heat resistance: withstands 1200 °C for 30 min - Antibacterial rate: >96% against E. coli and S. aureus - Thermal insulation: surface stays at <90 °C on 120 °C heat plate	SA and $CaCl_2$	- Improves hydration and water retention as a result of ion–water bonding - Enhances thermal stability (T_5% up to 235 °C) - Densifies porous structure for better insulation	[23]
2021	Gel–CS–GLY biogel coating	- Self-healing (full recovery after heating at 60 °C for 70 s) - Full recyclability (biogel resoluble in water and reusable)	CS and GLY	- CS: enhances hydrogen bonding and adhesion, improves elongation and char strength, and improves self-healing ability - Tensile strain increases 4-fold with CS	[127]

		- Biodegradability (natural food-safe components) - Adhesion strength: 0.685 MPa (doubled with CS) - High transparency in visible light		- GLY: retains water content (anti-dehydration and frost resistance), enhances hydrogen bonding for self-healing	

8.2.2. Expanding Application Scope through Multifunctionality

Another key advantage of flame-retardant hydrogels is the ability to integrate multiple functional properties, which broadens their range of applications, including the use of hydrogels beyond flame retardancy. Different applications require specific operational conditions—coatings need strong adhesion, wearable safety devices demand flexibility, and smart fire-warning systems require electrical responsiveness [161]. By combining multiple functions with flame resistance, hydrogels can be customized for various environments, enhancing user confidence and broadening their application [159]. This multifunctionality turns flame-retardant hydrogels into adaptable safety systems for diverse uses. For instance, when features such as biocompatibility and antibacterial activity are incorporated, these hydrogels can serve a dual purpose: providing fire safety while also functioning as biomaterials [128, 184, 185]. Although flame resistance is not typically required for biomedical applications, a hydrogel that is both flame-retardant and biologically functional is highly valuable for use in wound dressings or implant coatings. Furthermore, combining functionalities for applications such as energy storage, sensors, and fire alarms can significantly enhance their overall utility. This approach significantly increases the versatility and applicability of flame-retardant hydrogels.

Recently, the PAM/GO/Ni^{2+} hydrogel developed by An et al. [145] demonstrates impressive multifunctionality in strain, temperature, and flame sensing. For strain sensing (Figure 13a), its high electrical conductivity (Figure 13a-i) arises from the synergistic network formed by GO nanosheets and Ni^{2+} coordination with carboxyl groups in the PAM matrix, which enhances electron transport. The GF increases from 2.47 to 6.62 with strain (Figure 13a-ii), as conductive pathways are disrupted during stretching. Figure 13aiii–iv demonstrates the effectiveness of the hydrogel in detecting wrist and elbow movements.

For temperature sensing (Figure 13b), the deoxygenation of GO by Ni^{2+} restores the sp^2 carbon network, increasing the carrier density and causing a significant drop in resistance

as the temperature increases. This results in a high temperature sensitivity, as indicated by a temperature coefficient of resistance (TCR) value of −1.67 °C^{-1} in the 30–40 °C range (Figure 13b-ii), with the system capable of triggering an alarm (Figure 13b-i).

For flame sensing (Figure 13-c), strong hydrogen bonding and water retention enhance its thermal stability (Figure 13c-i). Exposure to a flame causes water evaporation and the reduction of GO to rGO, which sharply decreases its resistance (Figure 13c-iii). Prolonged exposure results in structural damage and increased resistance, as demonstrated by the flame alarm system shown in Figure 13c-ii.

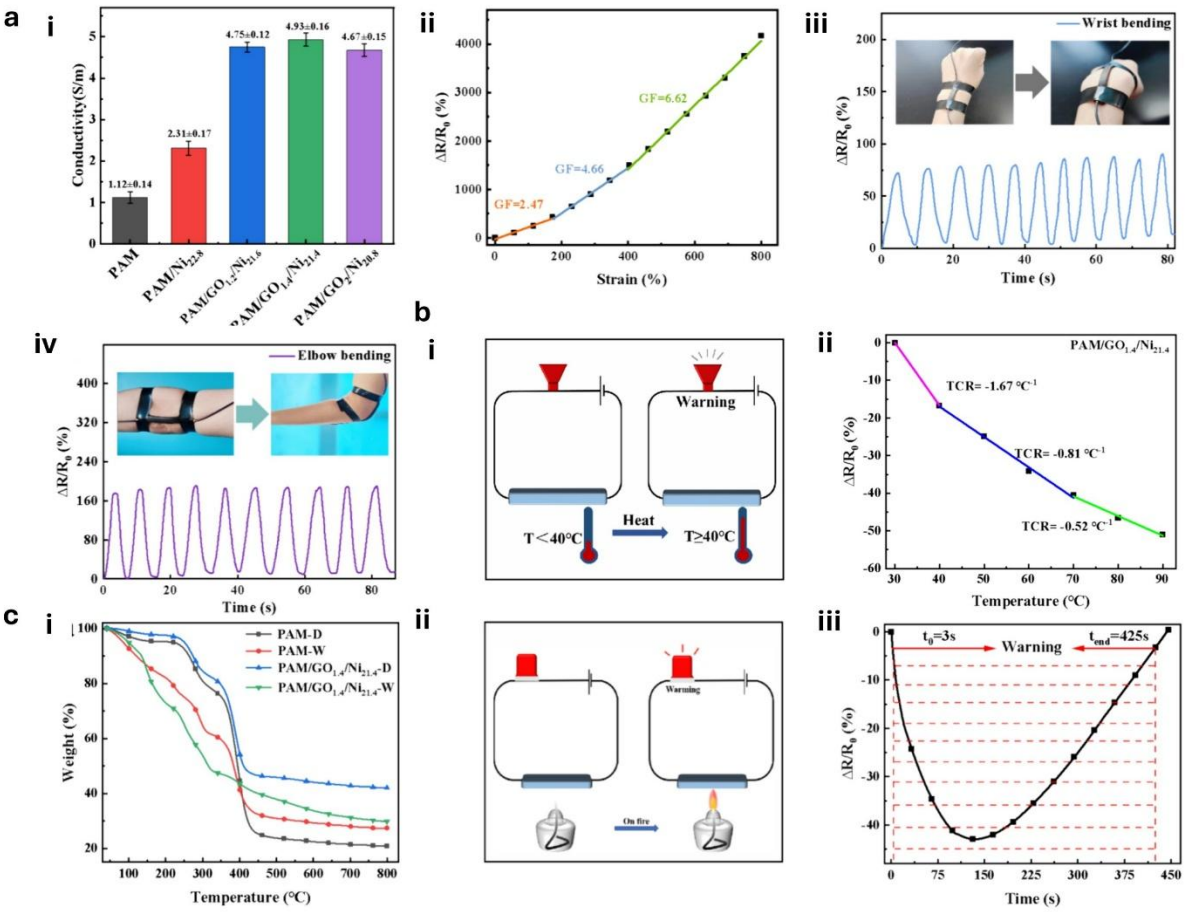


Figure 13. Multifunctionality of PAM/GO/Ni composite hydrogels in strain, temperature, and flame sensing: (a) Electrical characteristics of the PAM/$GO_{1.4}$/$Ni_{21.4}$ hydrogel-based strain sensor: i) electrical conductivity, ii) relative change in the resistance ($\Delta R/R_o$) and corresponding GF across strain levels from 0 to 450%, iii) wrist flexion, and iv) elbow flexion

for detecting human motion. (b) Temperature-sensing performance: i) a schematic of the temperature-alert system and ii) fitting curve illustrating the relationship between $\Delta R/R_o$ and temperature for the PAM/$GO_{1.4}$/$Ni_{21.4}$ hydrogel. (c) Flame-sensing capabilities of the PAM/$GO_{1.4}$/$Ni_{21.4}$ hydrogel: i) the thermogravimetric (TG) curve, ii) a schematic of the flame-alert mechanism, and iii) changes in the relative resistance under flame conditions. Adapted with permission from Ref. [145]. Copyright 2024, American Chemical Society.

8.2.3. Discussion

The hydrogel performance is governed by the combined effects of polymer selection, network preparation approach, and the addition of functional fillers or nanomaterials. Bio-derived polymer matrices such as PGP and silk nanofibers offer mechanical flexibility, adhesion, and environmental compatibility [80]. Dynamic or hybrid crosslinking networks formed by hydrogen bonding or borate ester interactions provide self-healing, anti-freezing, and anti-drying, thereby maintaining barrier integrity under thermal or mechanical stresses. Fillers are commonly introduced for electrical responsiveness and sensing abilities [61]. The incorporation of nanofillers like GO coordinated with Ni^{2+} improves electron transport pathways, resulting in enhanced strain sensitivity [145]. The hybrid hydrogel systems combining dynamic crosslinking with nano or supramolecular fillers improve mechanical strength, adhesion, and electrical functionalities without compromising on flame retardancy and sustainability [72, 109, 111, 112, 114, 128]. However, the optimal composition of multifunctional flame-retardant hydrogel depends on the specific application area: wearable devices benefit from flexible, conductive networks, whereas protective devices benefit from more rigid networks. In contrast, protective coatings demand better adhesion with dehydration resistance and flame resistance. The hydrogels created through hybrid crosslinking and reinforced with multifunctional nano or supramolecular fillers represent the most effective strategies for achieving a balance between flame retardancy and multifunctional properties.

Table 5 presents recent studies on flame-retardant hydrogels, emphasizing the expansion of their application scope through multifunctionality. It includes details on the hydrogel systems, their multifunctional applications, the flame-retardant additives used, and the additional properties or functions imparted by these additives.

Table 5. Recent studies on flame-retardant hydrogels, emphasizing the expansion of their application scope through multifunctionality.

Year	Flame-retardant hydrogel system	Applications or uses beyond flame retardancy	Flame-retardant additive	Properties or functions contributed by the flame-retardant additive	Ref.
2025	GEL-CNT-ILS	- Seawater desalination - Flame sensing - Energy generation - Motion sensing	CNTs + ionic liquid (1-butyl-3-methylimidazolium chloride)	-	[140]
2024	PAM/GO/Ni^{2+}	- Strain sensing - Temperature sensing - Flame sensing	Mainly GO and nickel ions (Ni^{2+}) (indirect role, mainly synergistic with GO)	The conversion of GO to rGO significantly reduces resistance, triggering electrical alarms within ~3 s of flame exposure and strain-sensing functions	[145]
	SA, graphene, and CNTs	- Fire alarm - Electronic skin	SA, graphene, and CNTs	Graphene and CNTs contribute to electrical conductivity	[100]

2024		- Flexible circuit			
2023	LiBr-infiltrated PAM hydrogel	- Humidity sensing - Respiratory monitoring - Non-contact sensing	LiBr	LiBr solution treatment improves water retention and conductivity for sensing	[67]
2022	SA/poly(acrylic acid-co-acrylamide)/NaCl	Overheat alarm	SA	-	[101]
2022	Poly(acrylamide-co-methacrylic acid) hydrogel	- Fire-resistant architectural glass - Smart safety coatings - Flame-retardant transparent layers in electronics or biomedical fields	APP + ammonium dihydrogen phosphate + urea + GLY	-	[186]
2022	MFCP-GLY/P	- Electromagnetic interference	Fe_3O_4 NPs and MXene nanosheets	- MXene: electrical	[172]

	organohydrogel (MXene–Fe_3O_4–CNF–PAAm–GLY/paraffin)	shielding (99.625%) - Strain sensing (GF ≈ 0.56) - Human skin protection from burns		conductivity, EMI attenuation via dielectric/conductive losses, structural integrity - Fe_3O_4: magnetic loss for enhanced EMI absorption, interfacial polarization, stability of emulsion droplets - MXene + Fe_3O_4 shells: Scattering/reflection of incident EM waves, self-extinguishing property	

9. Conclusions and Perspectives

9.1.Conclusions

Flame-retardant hydrogels have emerged as a promising class of multifunctional materials. They are used in a variety of applications ranging from coatings on flammable surfaces to bulk materials employed in energy storage, robotics, and smart sensing devices. Owing to their high water content, tunable polymer networks, and the incorporation of various flame-retardant additives, these hydrogels effectively inhibit ignition and combustion through multiple mechanisms, including heat absorption, the release of nonflammable gases, char formation, intumescence, and radical quenching.

This review categorized flame-retardant hydrogels into two main types: coating-type hydrogels, which provide surface-level fire protection for combustible substrates; and bulk hydrogels, which are embedded within materials (such as devices) to deliver intrinsic fire resistance. A wide range of additives, including phosphorus compounds, inorganic fillers, carbon-based NMs, and hybrid systems, have been explored to enhance both the flame-retardant performance and multifunctionality across diverse applications.

Importantly, these hydrogels demonstrate excellent compatibility with eco-friendly, self-healing, and recyclable systems, making them practical candidates for sustainable fire-safety solutions in advanced technologies.

9.2.Perspectives

9.2.1. Augmenting the Scalability and Manufacture of Flame-Retardant Hydrogels

Recent studies on flame-retardant hydrogels have focused on coatings on substrates such as coal, RPUF, and PPE. Various application techniques have been explored, including dip coating, spray coating, and in-situ polymerization, using both synthetic and bio-based materials. Additionally, the use of bulk-form hydrogels is gaining interest in fields such as energy storage, robotics, and 3D printing [187].

However, challenges remain in scaling-up advanced nanocomposite hydrogels, such as those incorporating black phosphorus [58] or MXene [21, 59, 73], owing to their high costs, complex synthesis procedures, and environmental safety concerns. The transition to sustainable bio-based hydrogels with competitive flame retardancy is an active area of research. The mass production of hydrogels with uniformly distributed additives and consistent performance remains a significant hurdle. Advances in additive manufacturing such as 3D printing, along with continued improvements in polymerization methods, could help address these challenges [188].

9.2.2. Improving the Performance and Sustainability of Flame-Retardant Hydrogels

The performances of flame-retardant hydrogels under real-world fire conditions, which are affected by factors such as humidity, abrasion, and prolonged heat exposure, have not yet been thoroughly evaluated. Future research should focus on developing flame-retardant additives that are inexpensive, easy to implement, and environmentally safe.

It is important to assess the performance of flame-retardant hydrogels in actual fire scenarios by considering variables such as fluctuating humidity, mechanical stress, and sustained thermal exposure. Such assessments are particularly relevant for coatings applied to PPE and structural wood because they would assist in clarifying how these external conditions influence the durability and effectiveness of coatings.

The use of biocompatible, non-toxic, and biodegradable components in hydrogels aligns with international environmental and safety standards, particularly for applications in biomedical and consumer products.

9.2.3. Enhancing the Sustainability of Flame-Retardant Hydrogels with a Focus on Post-Fire Exposure

There is growing interest in developing halogen-free, eco-friendly, and biodegradable hydrogels from renewable sources such as IA, Gel, and cellulose without compromising their

performance, thereby promoting sustainability [102, 163]. During fire exposure, organic polymers (e.g., CS and alginate), organic additives (e.g., TA), and carbon NMs (e.g., CNTs, graphene, and GO) within the flame-retardant hydrogel structures release CO_2. This contributes to environmental concerns such as global warming. Although CO_2 is non-flammable and is generally considered to be a non-hazardous gas, its cumulative effects can exacerbate climate change.

Future research should aim to develop hydrogel formulations that provide flame-retardant functionality while minimizing the use of organic matter, such as polymers and carbon-based materials, including CNTs and GO. The objective should be to create flame-retardant hydrogels using minimal amounts of structural components that contribute to CO_2 emissions. Emphasis should be placed on exploring formulations that incorporate carbon-free additives or ingredients such as water and phosphorus-containing compounds. These alternatives can decompose or evaporate upon exposure to fire, producing eco-friendly by-products such as water and NH_3 instead of CO_2.

9.2.4. Addressing the Trade-Off Between Mechanical and Functional Aspects While Maintaining Flame-Retardancy

In the pursuit of flame retardancy, there is often a trade-off between mechanical properties, such as stretchability and toughness, and functional aspects, such as electrical conductivity, self-healing, and adhesion. In particular, a trade-off commonly arises between mechanical strength and adhesion strength [182, 189]. Therefore, achieving excellent flame retardancy without compromising mechanical and functional properties remains a significant challenge. Enhancing the bonding between hydrogel coatings and various substrates, particularly under thermal or mechanical stress, is crucial. Techniques such as surface functionalization, composite interface engineering, and dynamic bonding strategies may help improve the durability of coatings.

Future research should focus on developing multinetwork architectures that utilize dynamic interactions [190, 191]. Although such approaches may initially increase synthesis complexity and cost, the long-term objective is to achieve scalable and economically viable systems once structural optimization is established. Nanoparticle incorporation has been widely explored to modulate microenvironments and enhance functional performance across diverse polymer and biomaterial platforms [26, 79, 192]. In particular, integrating nanomaterials such as GO, MMT, and CNTs into bio-hydrogel matrices through hybrid composite strategies, combined with hierarchical nano-structuring and emerging fabrication technologies, may generate synergistic improvements in flame retardancy, electrical conductivity, and environmental durability [140, 188, 193, 194].

9.2.5. Evaluating the Effect of Thermal and Environmental Conditions on the Performance of Flame-Retardant Hydrogels

Anti-freeze characteristics are essential for maintaining the operational capabilities of hydrogels in cold environments as they prevent the freezing of water within the hydrogel matrix [137]. Conversely, dehydration resistance is crucial for preserving the structure and water content of hydrogels over extended periods, thereby ensuring their functional durability and flame retardancy. Researchers have successfully integrated these characteristics into flame-retardant hydrogels using various methods [69, 84, 88].

However, there is a lack of studies investigating how fluctuations in the temperature and humidity of an operational environment affect the flame-retardant performance. For example, how does the flame-retardant performance of a well-developed hydrogel change when the temperature rises from room temperature to 40 °C? Similarly, how does the ambient humidity influence its effectiveness? Maintaining moisture stability under dry or elevated temperature conditions is vital for maintaining long-term flame retardancy. Approaches that

incorporate hygroscopic materials or humidity-responsive designs can offer effective solutions to these challenges.

9.2.6. *Incorporating a Resistance to Microbial Attacks in Flame-Retardant Hydrogel*

In service environments, flame-retardant hydrogels are exposed to various microbes, including bacteria and fungi. Without antimicrobial properties, these hydrogels could undergo microbial degradation, which would compromise their structural integrity. Although some studies have been conducted on the incorporation of antimicrobial properties into flame-retardant hydrogels, this work remains limited [128]. Because hydrogels are often used in contaminated or high-risk environments, it is essential to develop and integrate antimicrobial functionalities to extend their operational lifespan [195].

9.2.7. Developing Flame-Retardant Hydrogels for Multiple Applications with Multifunctional Properties

Considering the broad scope of flame-retardant hydrogels, researchers have reported the development of materials with multifunctional properties (Table 4) and diverse applications (Table 5). However, the field appears to have reached a plateau. To expand the potential and versatility of flame-retardant hydrogels, it is essential to develop products that integrate multifunctional properties for multiple applications.

For instance, future hydrogel designs should focus on combining fire resistance with other essential features such as electrical conductivity, thermoelectric properties, antimicrobial activity, and pressure sensitivity to meet the demands of wearable technologies, intelligent construction materials, and autonomous systems. Furthermore, the development of such hydrogels should prioritize the use of eco-friendly materials.

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The authors report there are no competing interests to declare.

Author Contributions:

Syed Farrukh Alam Zaidi, Abdullah Malik, and Aiman Saeed contributed equally to this work.
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Abdullah Malik: conceptualization, data curation, formal analysis, methodology, visualization, and writing–original draft.
Aiman Saeed: conceptualization, data curation, formal analysis, methodology, visualization, and writing–original draft.
Muhammad Waleed: data curation, formal analysis, visualization, and writing–original draft.
Muhammad Asif: data curation, formal analysis, visualization, and writing–original draft.
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Muhammad Asif Rafiq: investigation, validation, visualization, and writing–review and editing.
Ehsan Ul Haq: conceptualization, investigation, supervision, validation, and writing–review and editing.

Jung Heon Lee: conceptualization, funding acquisition, investigation, project administration, resources, supervision, validation, and writing–review and editing.

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Supplementary Information

Expanding the Frontiers of Flame-Retardant Hydrogels: Recent Advances in Applications, Design Strategies, and Multifunctional Performance

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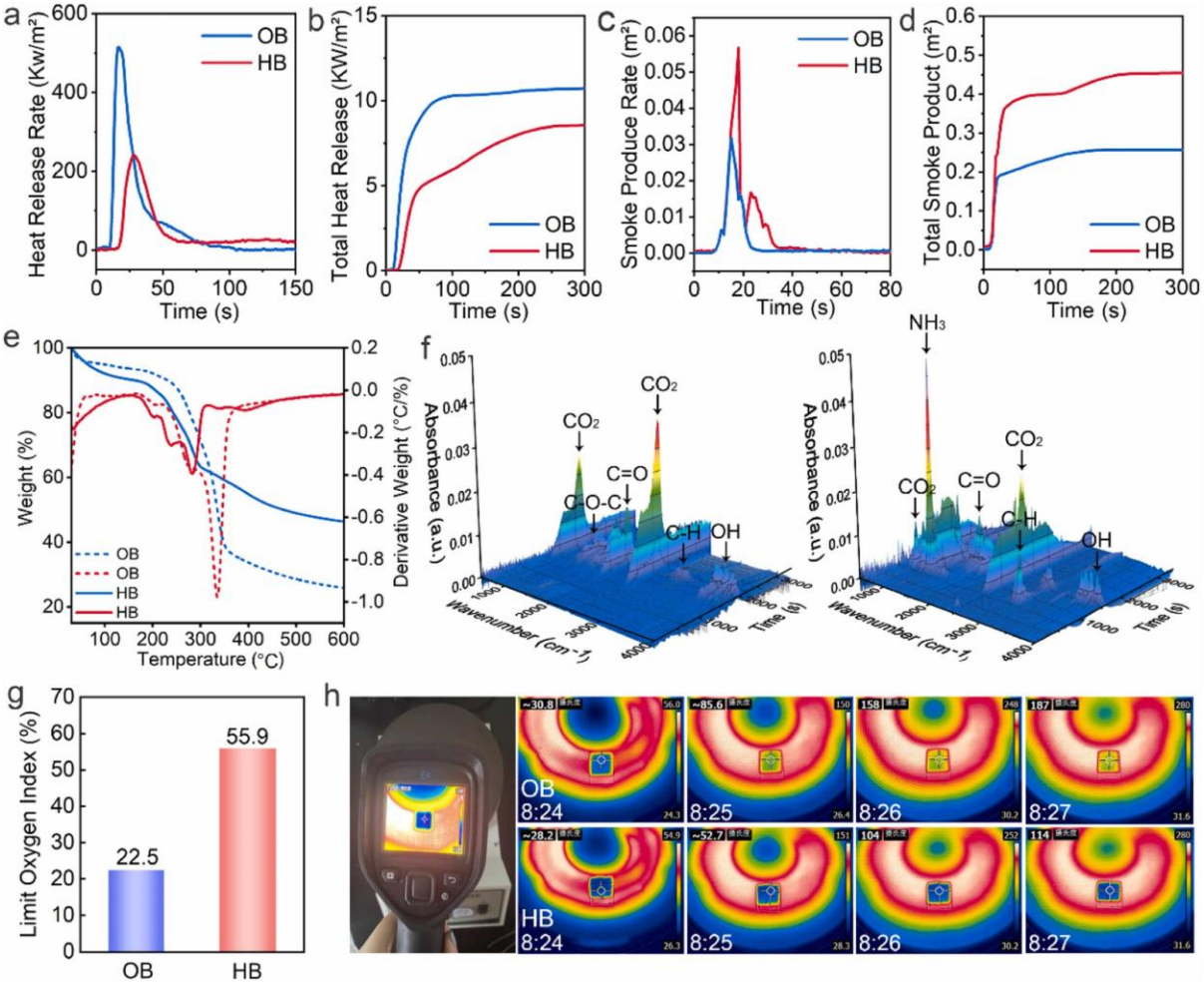

Figure S1. Thermal and Combustion Properties of OB and HB Hydrogels: (a) HRR, (b) THR, (c) Rate of smoke production, and (d) Total smoke production. (e) TG and DTG graphs for OB and HB in a N_2 environment. (f) TG-IR analysis of the gases released during the thermal breakdown of OB and HB. (g) The LOI. (h) Setup and findings from the thermal insulation experiment. Adapted with permission from Ref. [1] Copyright 2024, Elsevier B.V.

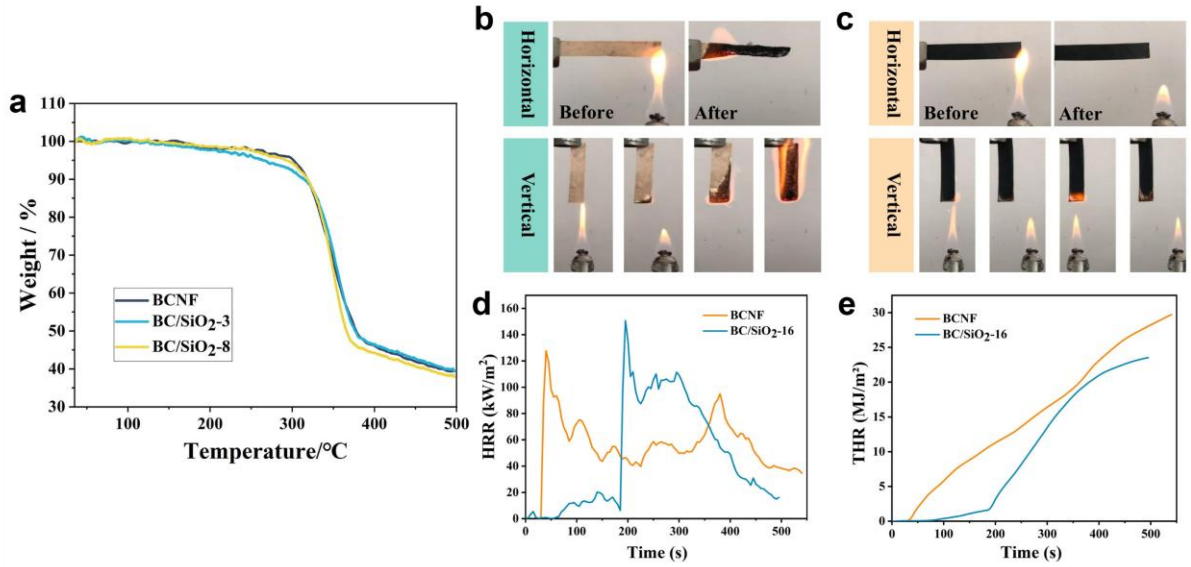

Figure S2. Thermal Stability and Combustion Behavior of BCNF and BC/SiO_2 Composites: (a) TG plots for BCNF and BC/SiO_2 composites. The combustion of BCNF with an alcohol lamp (b) and, (c) the combustion of BC/SiO_2 composites. Results for (d) HRR and, (e) THR of BCNF and BC/SiO_2 composites. Adapted with permission from Ref. [2] Copyright 2025, Elsevier B.V.

S1. Non-Flame-Retardant Performance Indicators of Hydrogel

S1.1. Water Retention, Freezing Tolerance, and Self-Healing

Considering water retention, for example, Yang et al. assessed the water retention capabilities of Gly-GAPL hydrogels [3]. At 80 °C, GAP0L2 quickly loses 66.7% of its weight within an hour and becomes fragile, whereas Gly-infused samples such as Gly-GAP4L2 maintain as much as 74.1% of their moisture after 7 hours and remain pliable (Figure S3a, S3b). At ambient temperature, GAP0L2 sheds 70.1% of its mass on the first day, but Gly-GAP4L2 only loses 14.2% throughout 7 days (Figure S3c, S3d). This improved retention can be attributed to hydrogen bonding from Gly, the interaction of PA-Na with polymer chains, and the ionic hydration effect of LiBr, which contributes to the long-term stability of the hydrogel in fire-warning applications [3].

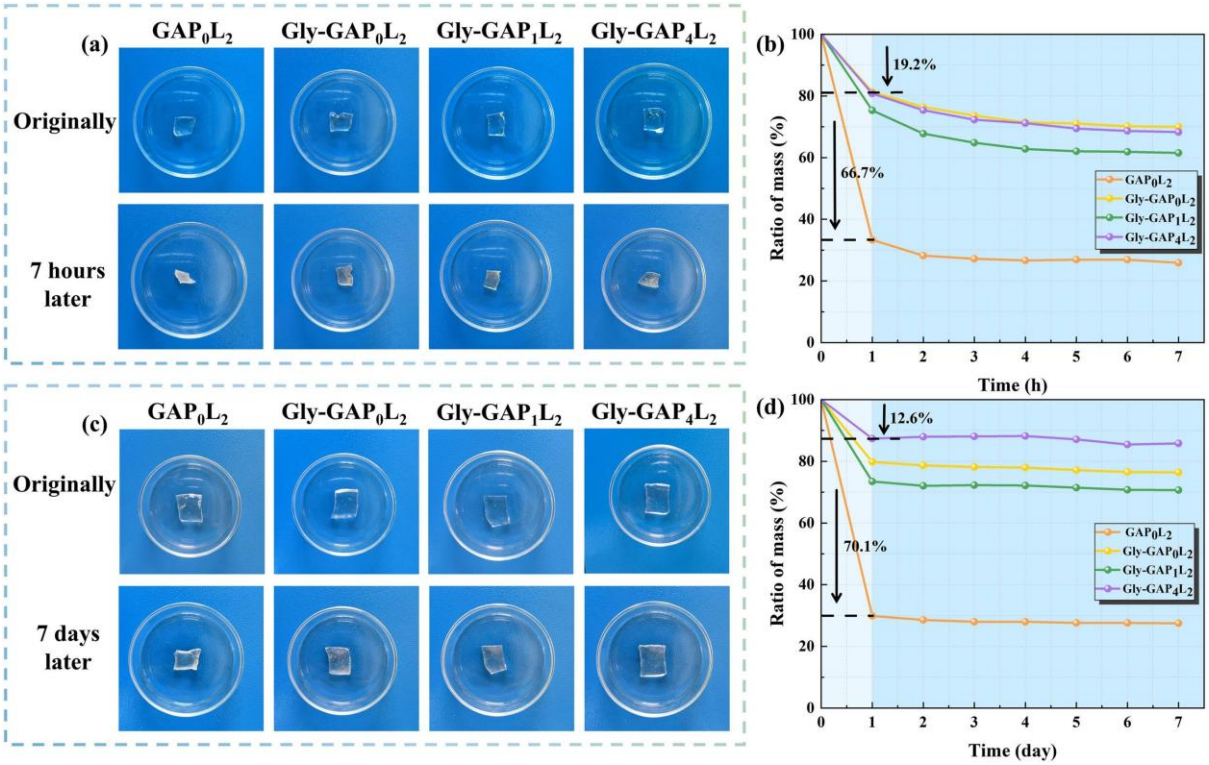

Figure S3. Water retention: Images and water retention measurements of the prepared GAPL hydrogels kept under varying conditions: (a)(b) 80 °C for 7 hours and (c)(d) 25 °C for 7 days. Adapted with permission from Ref. [3] Copyright 2025, Elsevier B.V.

In the realm of anti-freezing characteristics, Zuo et al. reported the anti-freezing capabilities of itaconic acid/cellulose-based hydrogels [4]. The anti-freezing performance of IAB and IAB/HPCS hydrogels, depicted in Figure S4, underscores their outstanding effectiveness in extremely cold conditions. Figure S4a illustrates that both hydrogels maintain high optical clarity after being frozen at −5 °C and −60 °C. Mechanical assessments (Figure S4b, S4c) indicate that the hydrogels preserve good flexibility and tensile strength post-freezing. The mechanism depicted in Figure S4d is due to the hydrogen bonding between hydroxyl groups and water, which prevents ice formation. SEM images (Figure S4e, S4f) validate the structural integrity of IAB/HPCS even after undergoing freezing at −60 °C for 12 hours. These findings demonstrate the hydrogels' exceptional cold resistance, making them suitable for applications in sub-zero conditions.

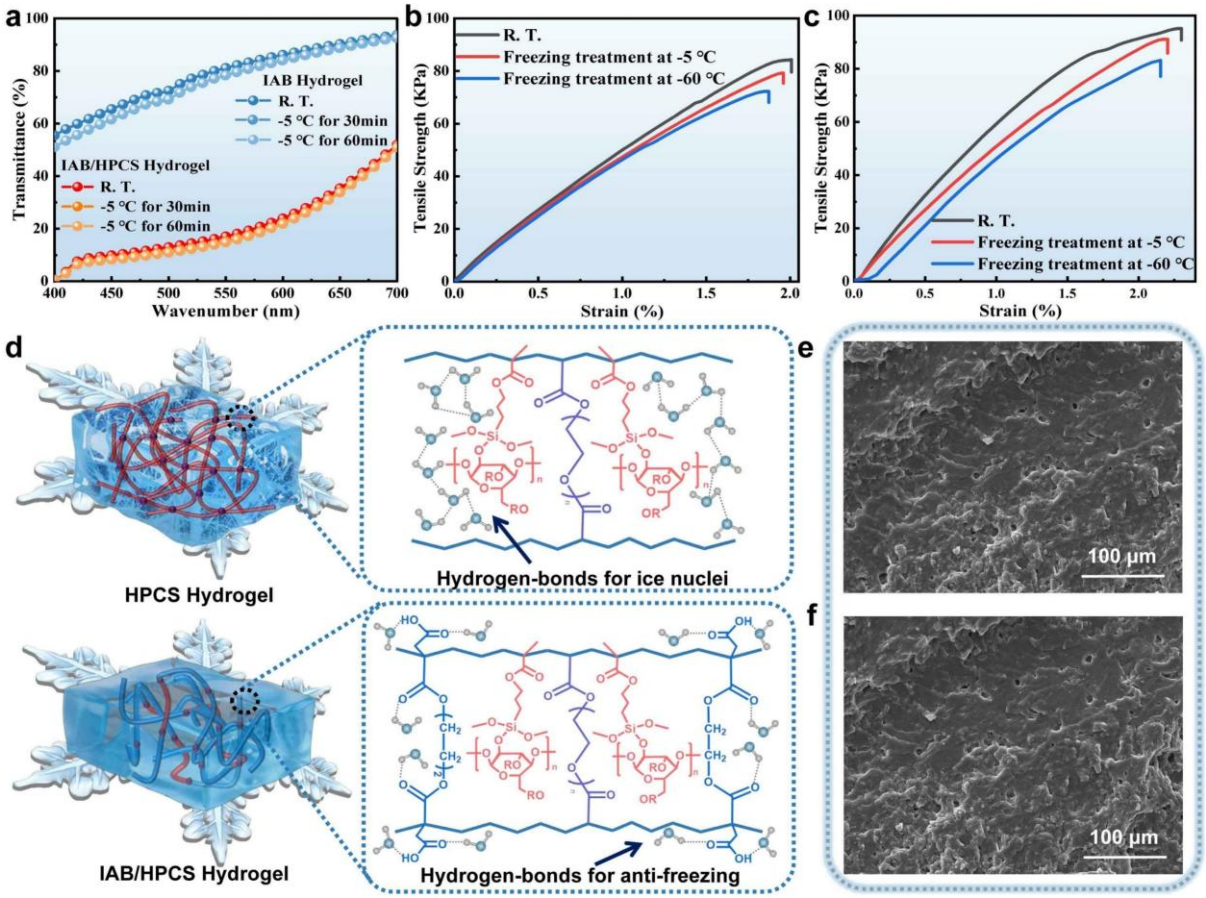

Figure S4. Anti-Freezing Performance and Mechanical Stability of IAB and IAB/HPCS Hydrogels: (a) Transmittance spectra of IAB and IAB/HPCS hydrogels at ambient temperature and at −5 °C after 30 and 60 minutes, respectively. Tensile strength-strain graphs for (b) IAB hydrogel and, (c) IAB/HPCS hydrogel prior to and following freezing treatment at −5 and −60 °C, respectively. (d) Schematic representation of the anti-freezing properties of IAB/HPCS hydrogel. SEM images showing the fracture morphology of IAB/HPCS hydrogel (e) before the freezing treatment and, (f) after the freezing treatment at −60 °C for 12 hours. Adapted with permission from Ref. [4] Copyright 2025, Elsevier B.V.

Figure S5a illustrates that the SPBG-$ZnSO_4$ hydrogel immediately self-repairs at room temperature, restoring conductivity as the LED bulb illuminates. Figure S5b elucidates this phenomenon through dynamic borate ester and hydrogen bonds that facilitate reversible connections. Figure S5c indicates a low freezing point of −24.24 °C, attributed to the presence

of glycerol and $ZnSO_4$, which inhibit ice formation. Even after being frozen for 2 hours at −20 °C, the hydrogel promptly repairs itself and regains conductivity, as depicted in Figure S5d, demonstrating its outstanding performance in cold conditions.

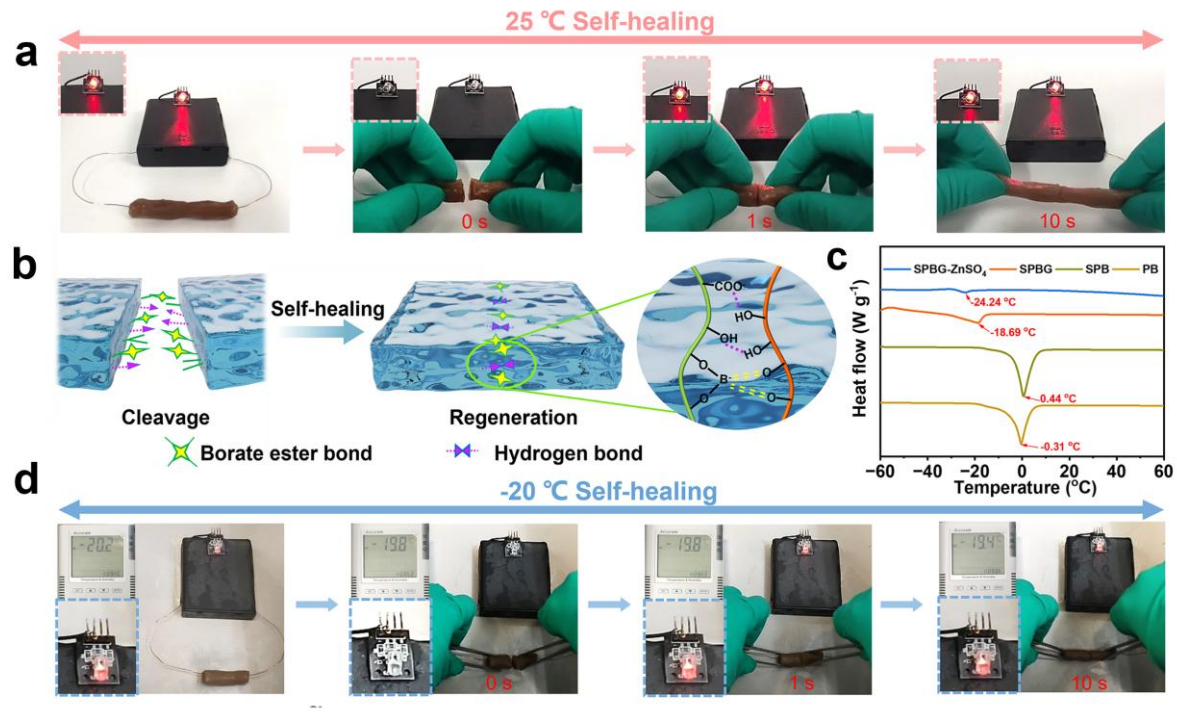

Figure S5. Self-Healing Conductivity and Thermal Behavior of SPBG-$ZnSO_4$ Hydrogels: (a) Images depicting the conductivity of SPBG-$ZnSO_4$ that has been cut, self-healed, and stretched at 25 °C. (b) Mechanism of self-healing for SPBG-$ZnSO_4$. (c) DSC curves for PB, SPB, SPBG, and SPBG-$ZnSO_4$. (d) Images illustrating the conductivity of SPBG-$ZnSO_4$ that has been cut, self-healed, and subsequently stretched at -20 °C. Adapted with permission from the reference [5]. Copyright 2024, American Chemical Society.

S1.2. Eco-friendliness, Biodegradability, and Recyclability

For instance, Zu et al. examined the recyclability of dual cross-linked CS-LA-$B(OH)_4Na$ GPEs [6]. As illustrated in Figure S6, the CS-LA-$B(OH)_4Na$ gel electrolyte demonstrates impressive recyclability. In Figure S6a, damaged gels completely dissolve in a levulinic acid (LA) solution and can be effectively reformed into functional gels after being immersed in a sodium borate solution. Figure S6b shows that the regenerated gels exhibit cyclic voltammetry (CV) curves

similar to those of the original gels, confirming that their electrochemical behavior is preserved. Additionally, Figure S6c reveals that supercapacitors using the recycled gels retain 88% of their capacity even after 35,000 cycles, highlighting their strong durability and reusability. This emphasizes the gel's sustainable and recyclable design, making it an excellent choice for green energy applications.

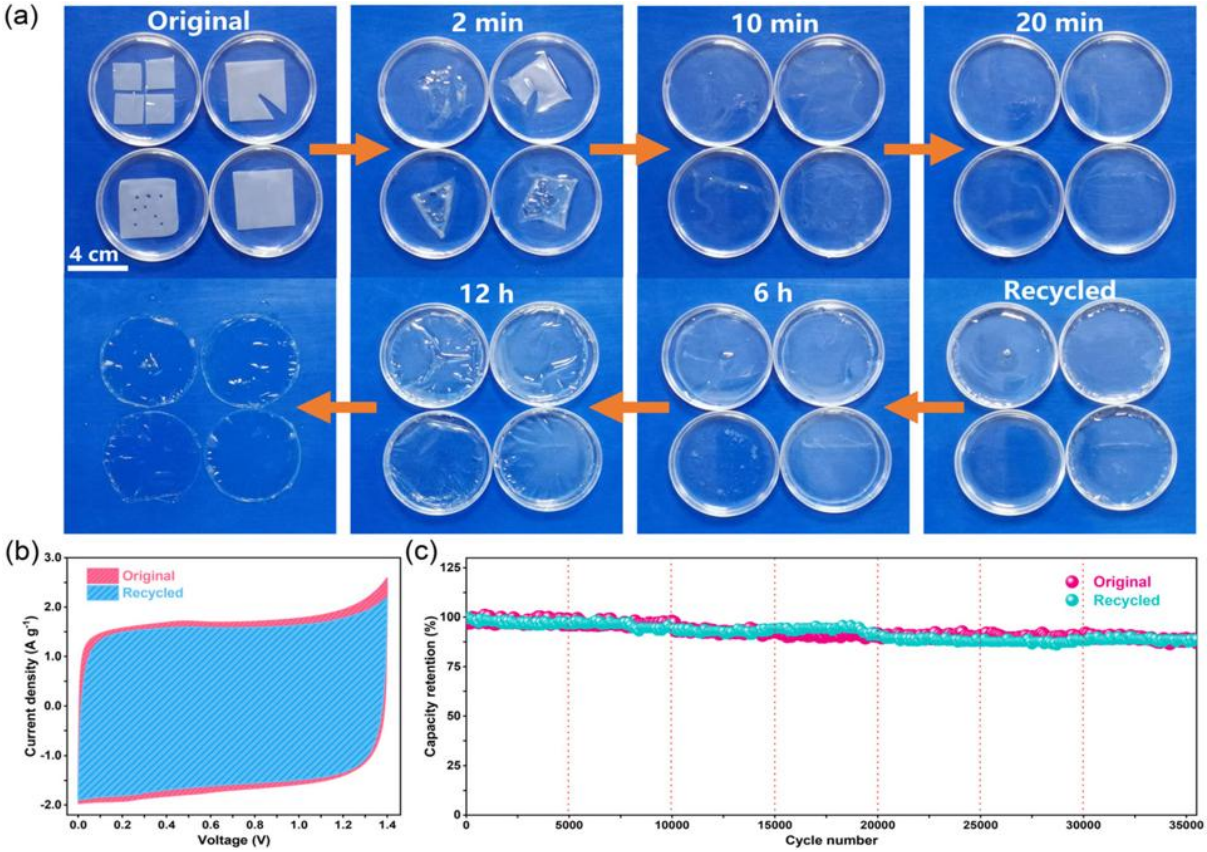

Figure S6. Recyclability and Electrochemical Performance of CS-LA-$B(OH)_4Na$ Gel Polymer Electrolytes (GPEs): (a) Digital photographs of CS-LA-$B(OH)_4Na$ GPEs slowly dissolving in an aqueous LA solution and being recycled in a sodium borate solution. (b) CV curves for both recycled GPEs and the original gel at a rate of 30 mV s^{-1}. (c) Cycling durability of recycled GPEs and the original GPE-based supercapacitors over 35,000 cycles at a current density of 1 A g^{-1}. Adapted with permission from Ref. [6]. Copyright 2024 Royal Society of Chemistry.

S1.3. Others (Anti-bacterial Activity and Biocompatibility)

Mahaninia et al. developed chitosan-vanillin hydrogels that offer flame-retardant properties, significant antibacterial effectiveness, and biocompatibility, making them suitable for applications that involve skin contact [7]. The research findings in Figure S7a and S7b demonstrates that the CSPA2 hydrogel displays impressive antimicrobial activity, eliminating 99% of Staphylococcus aureus and 80% of E. coli—13% more effective than pure chitosan. This enhanced performance is attributed to the imine and phenolic groups present in the vanillin-based cross-linker. Additionally, Figure S7c and S7d illustrates favorable biocompatibility, showing that HaCaT cells adhere to and grow better on CSPA2 compared to pure chitosan. Confocal imaging showed significantly improved cell proliferation on CSPA2 compared to pristine chitosan after 5 days, indicating better skin compatibility. This effect was linked to the imine and phenolic groups from vanillin that enhance cellular attachment. These findings suggested CSPA2's potential for direct skin-contact applications in flame-retardant coatings. These findings confirm that the hydrogel is both antimicrobial and skin-friendly, making it ideal for protective biomedical applications.

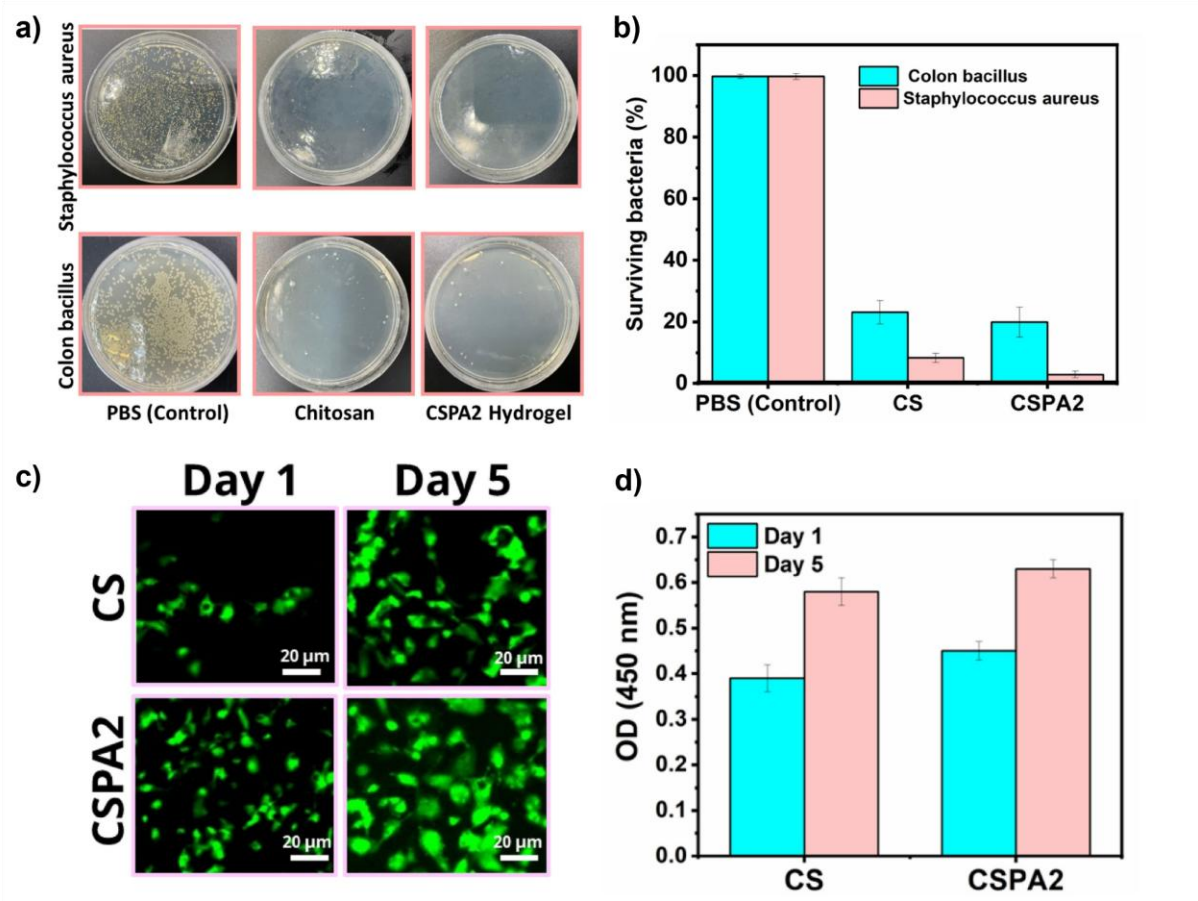

Figure S7. Antibacterial and Cytocompatibility Properties of CSPA2 Hydrogel: (a) Digital images showcasing the growth of Staphylococcus aureus (10^4 CFU/mL) and Colon bacillus (10^4 CFU/mL) bacteria on PBS (control), chitosan, and CSPA2 hydrogel. (b) A comparison of the survival rates of Staphylococcus aureus and Colon bacillus bacteria on PBS, chitosan, and CSPA2 hydrogel. (c) Observations of cell adhesion through confocal microscopy. (d) The number of cells counted on days 1 and 5. Adapted with permission from Ref. [7] Copyright 2023, Elsevier B.V.

S2. Flame-retardant hydrogel coatings specific

S2.1. Adhesion Performance

Xu et al. assessed the adhesive characteristics of a thermosensitive ionic hydrogel (Gel/PAM/GLY/TA/NaCl, PGTS) that exhibits flame-retardant properties [8]. Figure S8 illustrates the robust and diverse adhesion capabilities of the PGT_2S_2 hydrogel. As depicted in

Figure S8a, the hydrogel adheres strongly to a variety of surfaces, including metal, glass, rubber, wood, plastic, and paper. This showcases the broad adhesion potential of PGTS. The substantial adhesion is due to the many functional groups present in the hydrogel, including –OH, –COOH, and –CONH–, which create hydrogen bonds with the surfaces of the substrates. Furthermore, TA enhances adhesion through mechanisms such as π–π stacking, ligand bonding, and hydrophobic interactions. The adhesive strength was quantitatively assessed using the apparatus shown in Figure S8b, with results presented in Figure S8c revealing peak strengths of 48.3 kPa (paper), 45.3 kPa (wood), and 37.2 kPa (glass). These figures indicate strong bonding capabilities, even on low-energy surfaces like paper. In this aspect, the hydrogel exceeds the performance of many previously documented adhesive hydrogels. Repeatability experiments in Figure S8d demonstrate that the hydrogel maintains significant bonding strength even after five adhesion cycles, validating its fatigue-resistant properties. Together, these figures affirm the hydrogel's outstanding and reusable adhesion performance.

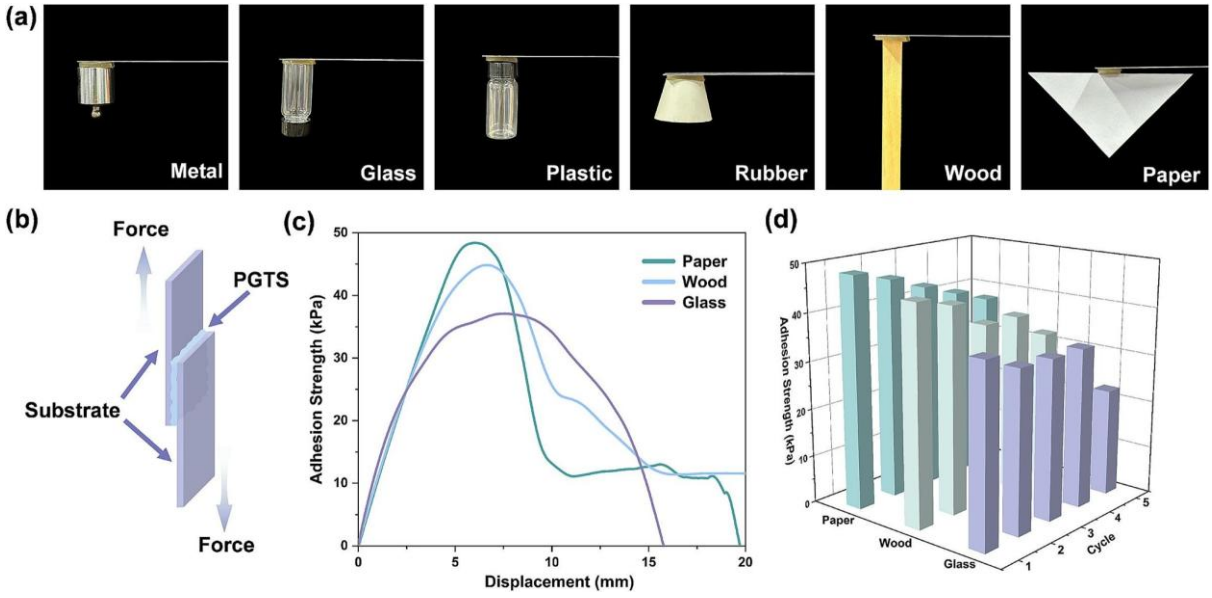

Figure S8. Adhesion Performance of PGT_2S_2 Hydrogel on Various Substrates: (a) The adhesion properties of the PGT_2S_2 hydrogel on different surfaces; (b) A schematic illustration of the experimental arrangements utilized to assess adhesive strength; (c) The adhesive strength

of the PGT_2S_2 hydrogel in relation to various substrates; (d) The adhesion strength of the PGT_2S_2 hydrogel to the respective substrates after 5 cycles. Adapted with permission from Ref. [8] Copyright 2025, Elsevier B.V.

S3. Bulk-Form Flame-Retardant Hydrogels Specific

S3.1. Mechanical Performance

The mechanical performance of flame-retardant hydrogels is crucial for their use in fire-safety systems. For instance, Zaidi et. al. evaluated the mechanical properties of chitosan-reinforced gelatin flame-retardant hydrogel for supercapacitor applications [9]. Figure S9 evaluates the mechanical properties of Gel/CS composite hydrogels. Figure S9a shows that increasing CS content enhances the strength and toughness of the hydrogels. Figure S9b illustrates the flexibility and durability of the $Gel_7CS_{0.6}$ hydrogel through various mechanical tests, confirming its resilience. Quantitative results in Figure S9c reveal the elastic modulus rising from 0.0627 ± 0.0179 MPa (no CS) to 1.96 ± 0.047 MPa (0.6 wt% CS). Figure S9d shows tensile strength increasing from 0.279 ± 0.038 MPa to 2.946 ± 0.19 MPa, while Figure S9e highlights toughness improvement from 0.35 ± 0.067 MJ/m^3 to 3.812 ± 0.345 MJ/m^3 at 0.6 wt% CS. Figure S9f notes a reduction in elongation at fracture from 346.1% to 254.6%, indicating a trade-off between flexibility and structural rigidity. Figure S9g compares stress–strain behaviors of normal vs. notched samples, showing that CS enhances crack resistance. Finally, Figure S9h quantifies a tenfold increase in fracture energy, from 2.5 kJ/m^2 to 26 kJ/m^2 for the composite hydrogel, attributed to the toughening effect of CS particles.

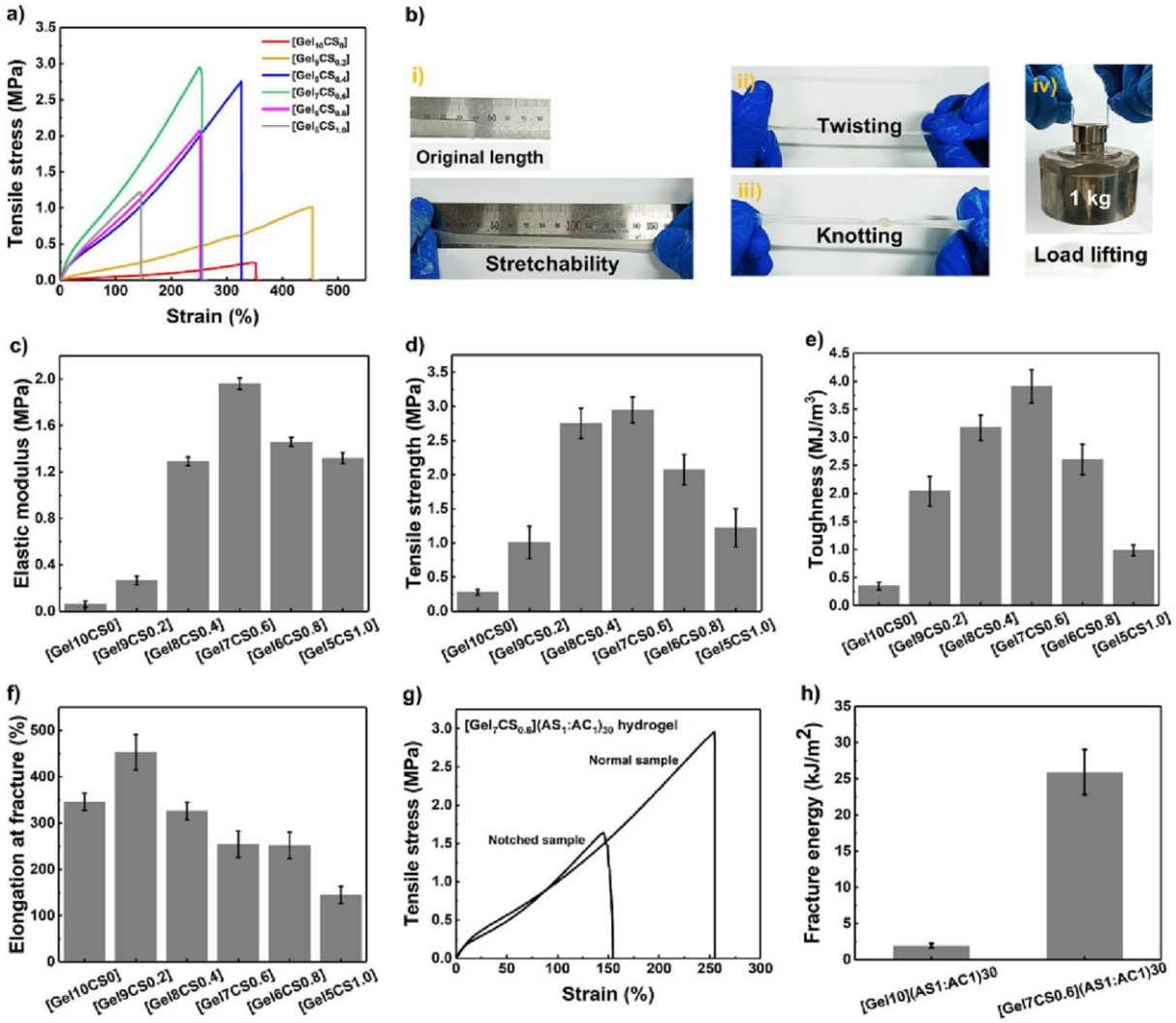

Figure S9. The mechanical characteristics of the [Gel/CS]$(AS_1:AC_1)_{30}$ composite hydrogels depend on the fraction of chitosan (CS): (a) A graphical depiction of the typical behavior of tensile stress versus strain; (b)- i)properties such as stretchability, ii) twisting, iii) knotting, and iv) load-bearing capacity; c elastic modulus; (d) tensile strength; (e) toughness; and (f) elongation at the point of fracture of the hydrogel as influenced by CS content; (g) tensile stress-strain curves for both normal and notched hydrogel samples; and (h) fracture energy for the $[Gel_{10}](AS_1:AC_1)_{30}$ hydrogel compared to the $[Gel_7CS_{0.6}](AS_1:AC_1)_{30}$ composite hydrogel. Adapted with permission from Ref. [9] Copyright 2023, Elsevier B.V.

S3.2. Self-Adhesion Performance

Zhang et. al. evaluated the self-adhesive performance of the bio-based hydrogel electrolytes (PSBGL) based on peach gum polysaccharide/sisal nanofibers for flexible supercapacitors [10]. The self-adhesive properties of the PSBGL bio-based hydrogel electrolyte are presented in Figure S10. Figure S10a shows the layered structure of the flexible supercapacitor, with the PSBGL hydrogel as the core electrolyte between two AC-symmetric electrodes on carbon cloth, relying on the hydrogel's adhesive strength for mechanical performance. Figure S10b quantitatively demonstrates the self-adhesive performance, revealing a lap shear strength of 1.15 MPa, indicating a strong interfacial bond essential for electrochemical stability. Photographic evidence in the insets highlights firm contact retention during tensile testing. Figure S10c provides optical microscope images of the hydrogel-electrode interface before and after mechanical bending, showing no visible detachment or deformation, confirming excellent flexibility and adhesion even under stress. Figure S10d illustrates practical implications, where a supercapacitor powered a digital stopwatch while being compressed and bent, thanks to the hydrogel's strong adhesion. Finally, Figure S10e highlights sustainability, showing biodegradation of the PSBGL hydrogel in soil over one, three, and five months, emphasizing its skin-safety and biodegradability while adhering well to human joints.

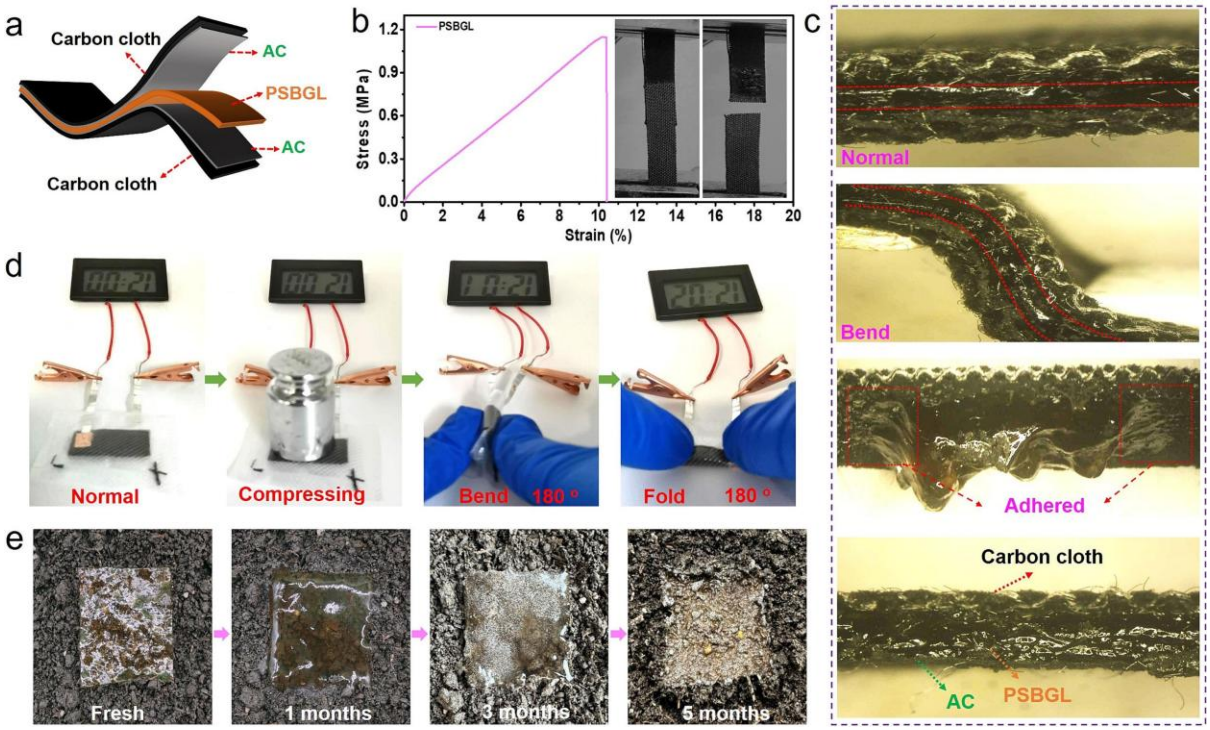

Figure S10. Mechanical Integration, Functionality, and Biodegradability of PSBGL Bio-Based Hydrogel Electrolyte in Flexible Supercapacitors: (a) Diagram depicting the configuration of the flexible supercapacitor. (b) Strength of adhesion between the PSBGL bio-based hydrogel electrolyte and the electrode (the insets display images of the tensile testing process). (c) Visual observations of the interface between the PSBGL bio-based hydrogel electrolyte and the electrode. (d) Demonstration of power supply using the flexible supercapacitors under different mechanical stimuli. (e) Changes observed in the fresh PSBGL bio-based hydrogel electrolyte after being buried in soil for one, three, and five months. Adapted with permission from Ref. [10] Copyright 2024, Elsevier B.V.

S3.3. Resistive Strain Sensing Performance

Most flame-retardant strain sensor hydrogels also work on resistive strain sensing mechanisms [11–14]. For example, Li et al. assessed the strain sensing capabilities of a multifunctional hydrogel based on carbon nanotubes, referred to as GEL-CNT-ILS, which is a novel copolymer gel made from CNT with acrylamide and acrylic acid [11]. Figure S11a illustrates the response of the GEL-CNT-ILS hydrogel under different levels of motion, categorized into three

intensities—basic (30%), mild (50%), and intense (70%) elongation. The figure depicts the ratio of resistance to initial resistance (R/R_0), which rises with strain: around 160% for basic movement, 225% for mild movement, and 275% for intense movement. This linear and differentiated response emphasizes the material's significant sensitivity and its ability to differentiate between various levels of physical activity. Figure S11b shows an experiment involving finger bending, where the hydrogel is directly affixed to a human finger. As the finger bends and stretches, the hydrogel undergoes deformation, resulting in real-time variations in resistance. This immediate response validates the hydrogel's appropriateness for applications requiring wearable and flexible motion sensing. The hydrogel demonstrated consistent performance even after multiple motions, showcasing its durability and flexibility. Figure S11c illustrates how the GEL-CNT-ILS hydrogel acts as a conductor to light a small bulb, showcasing its electrical conductivity. Figure S11d provides a wiring diagram for connecting the GEL-CNT-ILS sample to a power source and load, clarifying the experimental setup shown in Figure S11c. Figure S11e illustrates how the resistance of GEL-CNT-ILS_1 changes with stretching. As the material undergoes elongation, its resistance increases, which corresponds with typical behaviors found in strain sensors. This mechanical-electrical interaction enhances its potential for use in strain or deformation sensing. Figure S11f shows the results of Electrochemical Impedance Spectroscopy (EIS), highlighting the impact of ionic liquid amounts on the impedance spectrum. The Nyquist plot's intersection with the real axis indicates the material's resistance, demonstrating that ionic liquid content significantly affects both resistance and conductivity of the gel. Figure S11g plots the conductivity measurements of various hydrogels. The results show that carbon nanotubes by themselves do not significantly enhance conductivity, but increasing the concentration of ionic liquid leads to a marked rise in conductivity. The GEL-CNT-ILS composite hydrogel achieves an impressive conductivity of 3.67 $mS \cdot cm^{-1}$, making it suitable for applications that require effective charge transport.

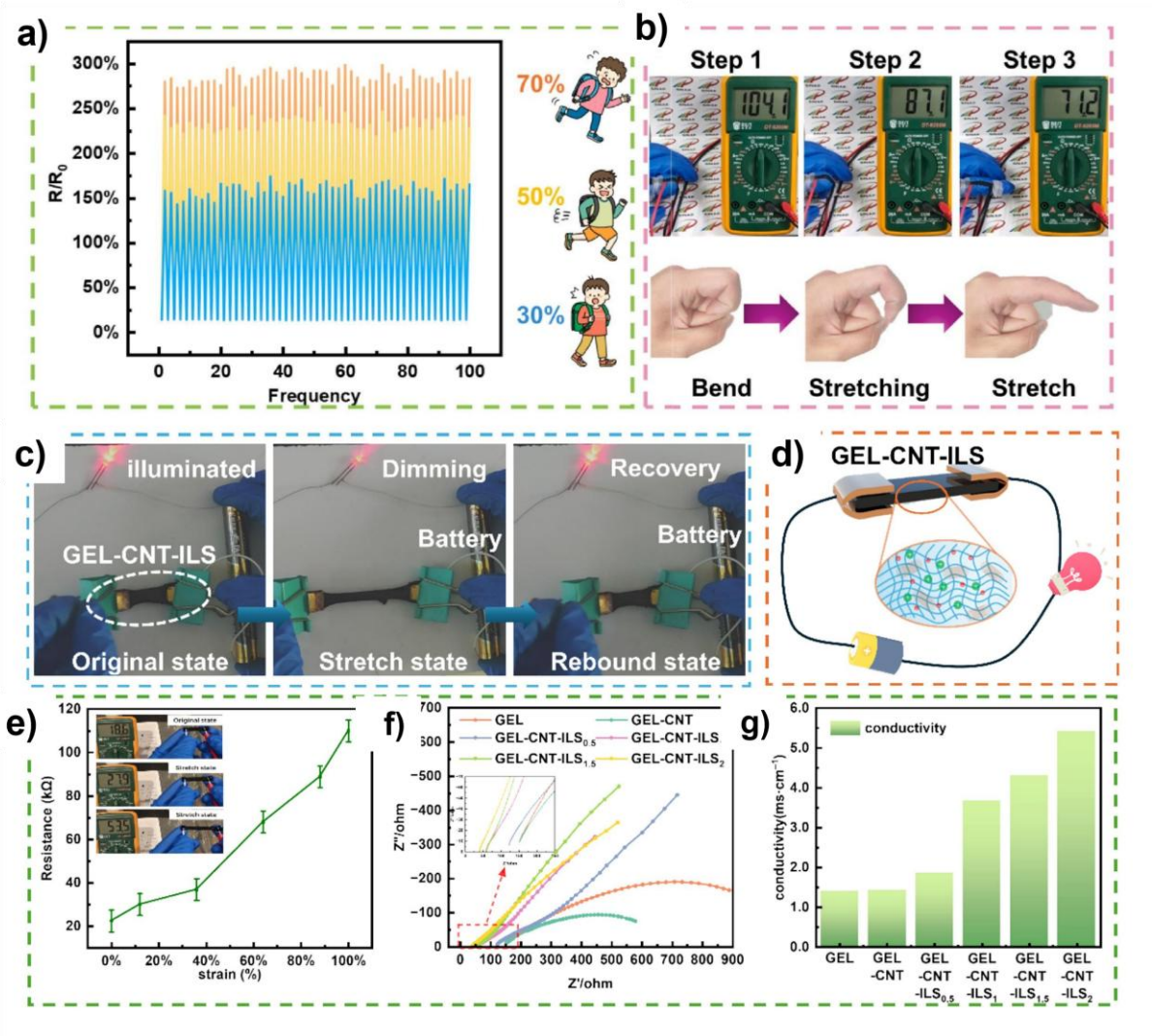

Figure S11. Strain sensing performance of GEL-CNT-ILS$_1$ hydrogel: (a) The behavior of the hydrogel when subjected to varying levels of motion. (b) Demonstration of finger bending sensing. (c) Utilizing GEL-CNT-ILS as conductors to illuminate small light bulbs. (d) Wiring schematic of GEL-CNT-ILS functioning as a conductor. (e) The change in resistance of GEL-CNT-ILS$_1$ with stretching deformation. (f) EIS impedance spectrum analysis of materials. (g) Electrical conductivity of materials. Adapted with permission from Ref. [11] Copyright 2025, Elsevier Inc.

S3.4. Energy Storage Performance

Recent studies have highlighted flame-retardant hydrogels as promising electrolytes for safe supercapacitors [5, 6, 9, 15–23], particularly in ZICs [24], as presented in Figure S12. The

PAM/PUL/PA hydrogel showed impressive electrochemical stability, mechanical flexibility, and an ionic conductivity of 22.54 mS cm^{-1}, surpassing PAM-only hydrogels at 10.34 mS cm^{-1}. In ZICs, it achieves a specific capacity of 111.5 mAh g^{-1} at 0.1 A g^{-1} and an energy density of 97.57 Wh kg^{-1} at 49.99 W kg^{-1} power density. It maintains 74.6% capacitance retention after 10,000 cycles and remains stable for 150 hours in Zn//Zn symmetric cells. Furthermore, it retains 98% capacitance under 180° bending and 83.8% after 1000 bending cycles, showcasing its potential for wearable devices and safe energy storage systems [24].

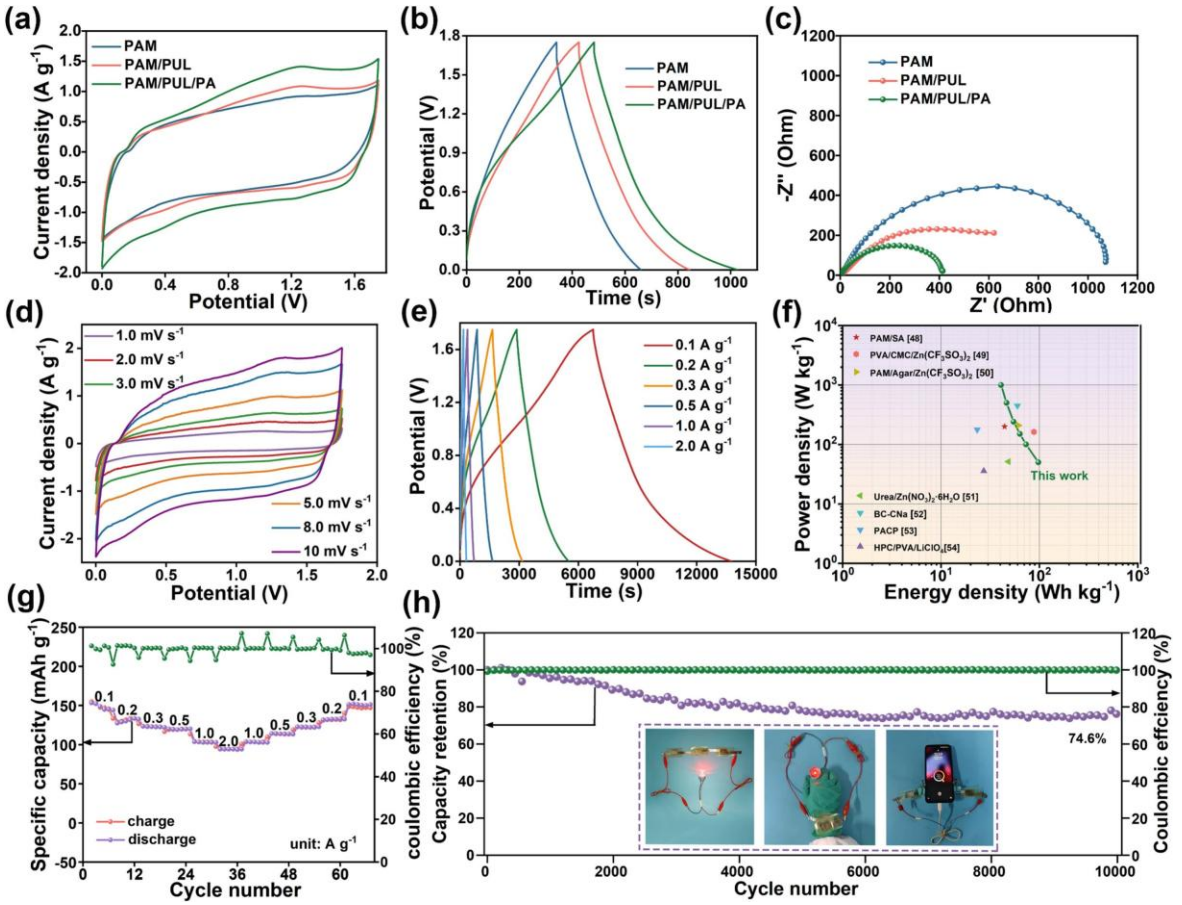

Figure S12. Electrochemical performance characteristics of the ZICs utilizing PAM, PAM/PUL, and PAM/PUL/PA hydrogel electrolytes include: (a) CV curves, (b) GCD curves, and (c) Nyquist plots. The electrochemical performance of devices based on PAM/PUL/PA hydrogel is represented by: (d) CV curves, (e) GCD curves, (f) Ragone plots, g rate performance, and (h) cyclic performance of the ZICs (Insets: images of a flexible wearable device). Adapted with permission from Ref. [24] Copyright 2024, Elsevier B.V.

Furthermore, hydrogel-based battery components with flame-retardant properties are promising for energy storage, especially as separators [25], electrodes [26], and electrolytes [19, 25, 27] in lithium-ion batteries (LIBs). For example, Wang et al. investigated the feasibility of using flame retardant carrageenan (CG)-PAM@polyacrylonitrile (PAN)-SiO_2 quasi-solid-state hydrogel electrolyte (QSE) in Li||Li symmetric batteries [17]. Figure S13 offers a detailed evaluation of the electrochemical performance of Li||QSE||LFP and symmetric cells with the CG-PAM@PAN-SiO_2 quasi-solid-state electrolyte, particularly under low-temperature conditions. In Figure S13a and S13b, Li||Li symmetric cells achieved stable cycling for approximately 157 hours at a high current density of 10 mA/cm² before experiencing a voltage drop due to a dendritic short circuit, with a modest overpotential of 2.5 mV indicating strong initial stability. Figure S13c shows that the voltage was stable at room temperature (20 °C) but significantly decreased at –20 °C and –60 °C, dropping to 1.67 V due to reduced ion conductivity caused by electrolyte freezing, although some recovery was observed. Figure S13d displays cyclic voltammetry (CV) performed at –30 °C, revealing distinct redox peaks at 1.7 V, 3.0 V, and 3.7 V, validating the system's functionality at sub-zero temperatures. Electrochemical impedance spectroscopy presented in Figure S13e indicates that the addition of PAN-SiO_2 reduces overall impedance and facilitates Li^+ diffusion, while Figure S13f illustrates an increase in impedance at –40 °C due to limited ion mobility. In Figure S13g and S13h, CV conducted at various scan rates (2–8 mV/s) shows increasing current and b-values of 0.75 and 0.78, suggesting pseudocapacitive behavior with rapid charge storage. Figure S13i emphasizes capacity loss in CV curves at –30 °C, likely due to degradation of the solid electrolyte interphase (SEI) and frozen interfaces. Figure S13j exhibits charge–discharge profiles at 20 °C, indicating low polarization voltages (0.06 V and 0.05 V), which reflect effective Li^+ intercalation. Long-term cycling stability is confirmed in Figure S13k, demonstrating the durability of the QSE. Finally, Figure S13l showcases the synergistic effect

of the composite structure—integrating CG-PAM gel and PAN-SiO_2 nanofibers—to enhance flame retardancy, mechanical strength, and prevention of dendrite formation.

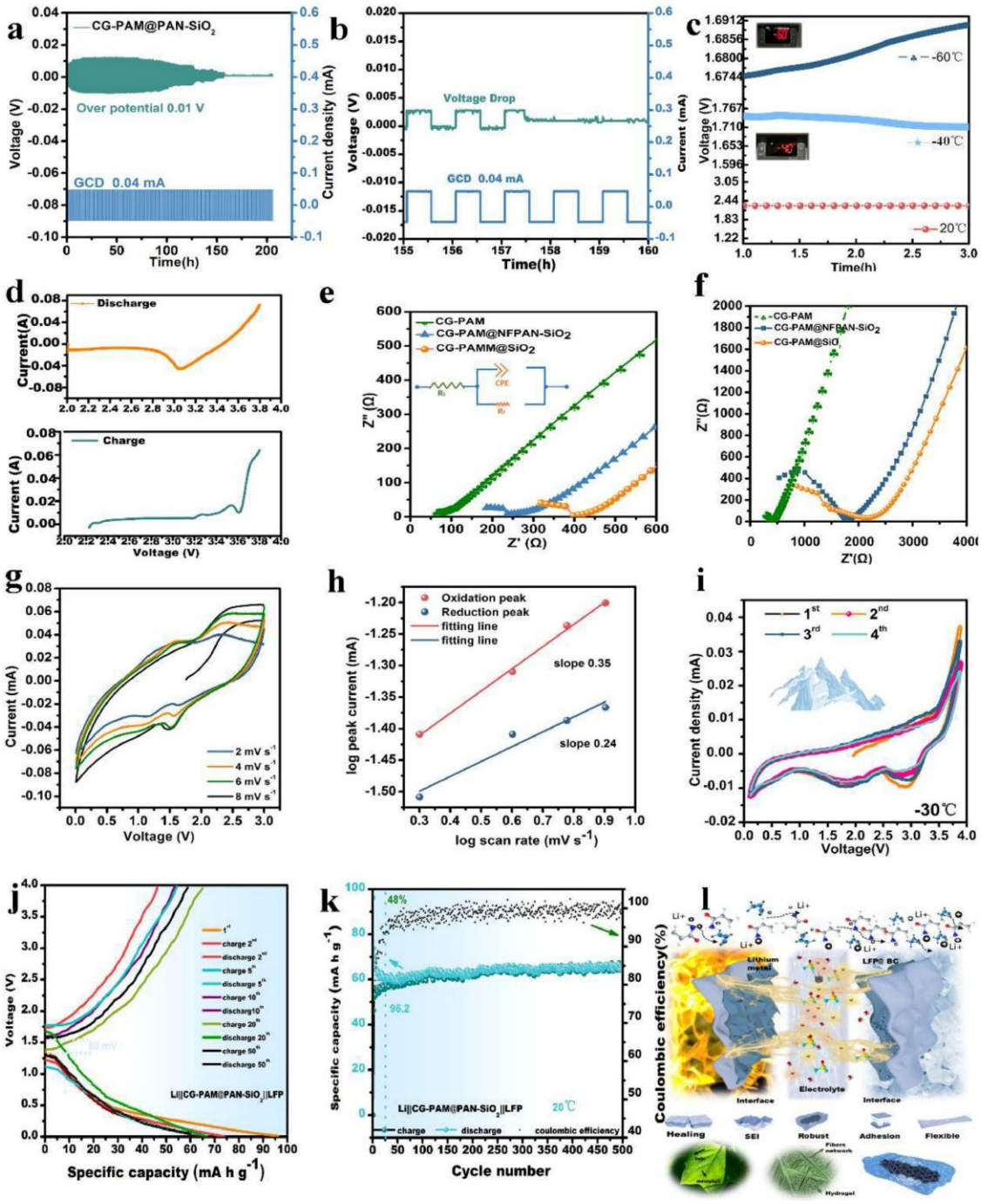

Figure S13. Electrochemical Performance and Low-Temperature Stability of CG-PAM@PAN-SiO_2 QSE in LIBs: (a)(b) illustrate the extended cycling test results of the Li||Li symmetric battery. (c) shows the voltage retention assessment of a completely assembled battery utilizing freeze-resistant low-temperature electrolyte materials under low-temperature

conditions. (d) displays the cyclic voltammetry curves of a fully assembled battery (Li| GE |LFP) incorporating freeze-resistant low-temperature electrolyte materials, examining the occurrence of electrochemical reactions. (e) represents the EIS test results of the electrolyte material at room temperature, along with the corresponding fitted circuit diagram. (f) shows the Bode plot from the EIS impedance test of the electrolyte material at low temperatures (-40 °C). (g) presents the cyclic voltammetry curves of Li| CG-PAM@PAM-SiO_2|LFP at various current densities (2∼8 mV/s). (h) provides the fitted analysis of the cyclic voltammetry curves across different current densities. (i) depicts the cyclic voltammetry curve of the complete battery of Li| CG-PAM@PAN-SiO_2|LFP at −30 °C. (j) illustrates the charge-discharge voltage profiles of Li| CG-PAM@PAN-SiO_2|LFP at a temperature of 20 °C. (k) outlines the cycling performance of Li||LFP cells with and without CG-PAM@-PAN-SiO_2 electrolyte at 20 °C. (l) shows the mechanism diagram depicting how individual components influence the electrolyte's electrochemical performance. Adapted with permission from Ref. [17] Copyright 2024, Elsevier B.V.

Moreover, flame-retardant hydrogels serve as safe alternatives to flammable commercial polyolefin-based separators during thermal runaway or short circuits. They enhance electrochemical performance with high porosity, wettability, and ionic conductivity [25, 26]. For instance, the flame-retardant hydrogel (Melamine–PA Supramolecular Polymer Assembly, MAPA) was used to construct the anode of a lithium-ion battery, enhancing safety by reducing flammability [26] (Figure S14). As an anode material, it demonstrated impressive electrochemical performance, achieving an initial discharge capacity of 570 mAh g^{-1} at 0.1 A g^{-1} and retaining 397 mAh g^{-1} after 600 cycles, indicating excellent cycling stability. The electrode also exhibited capacity recovery, maintaining 392.3 mAh g^{-1} at the 600th cycle due to improved electrolyte infiltration and porosity. Additionally, the MAPA-based electrode

facilitated fast Li^+ adsorption/desorption kinetics, achieving high performance across current densities from 0.1 to 5 A g^{-1} [26].

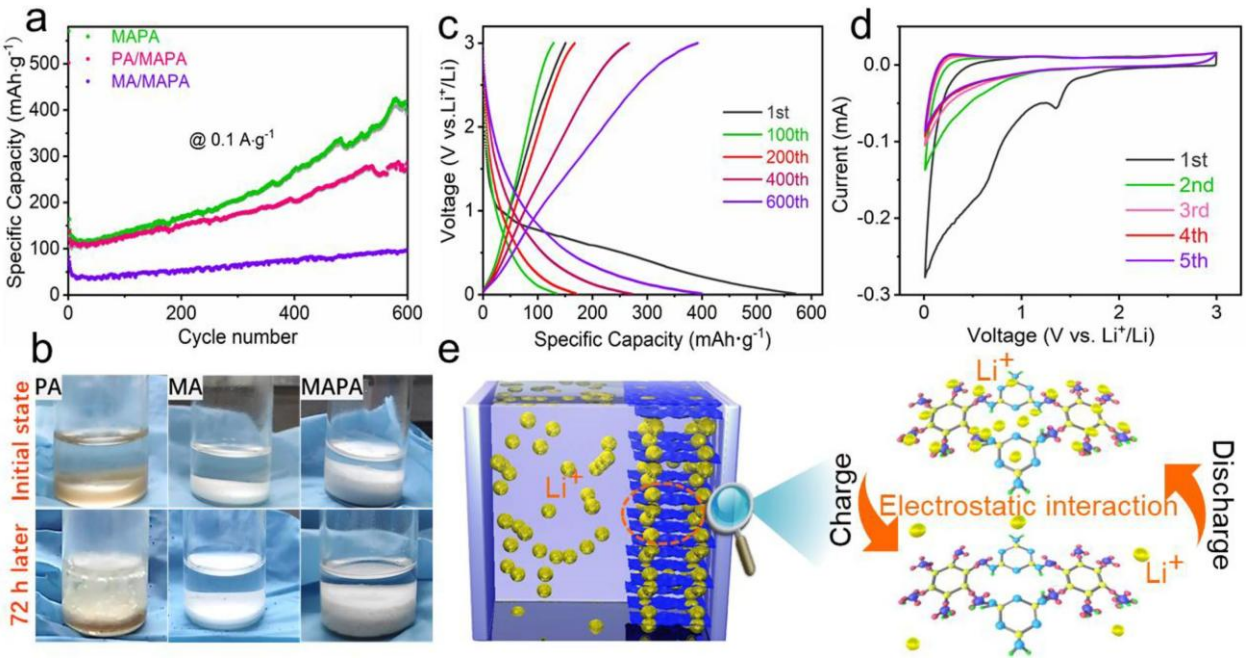

Figure S14. Electrochemical Properties and Charge/Discharge Mechanism of MAPA-Based Anodes for Lithium-Ion Batteries: (a) The cycling performance of MA/MAPA, MAPA, and PA/MAPA electrodes at a current density of 0.1 A g−1. (b) The visible solubility assessment of MA, PA, and MAPA in the electrolyte (EC: DMC = 1: 1). (c) The charge/discharge profiles of the MAPA electrode over various cycles from (a). (d) Cyclic voltammetry results of MAPA at a scanning rate of 0.2 mV s−1. (e) A schematic representation of the MAPA anode's charge/discharge process for lithium-ion batteries (LIBs). Adapted with permission from Ref. [26] Copyright 2021, Elsevier B.V.

S3.5. Thermoelectric Performance

Li et al. developed a multifunctional carbon nanotube-based hydrogel (Carbon Nanotube and Ionic Liquid-doped Gel, GEL-CNT-ILS) with excellent thermoelectric properties via radiation polymerization, combining multi-walled carbon nanotubes and ionic liquids in an acrylamide-acrylic acid copolymer gel [11]. Figure S15 depicts the thermoelectric characteristics of the

GEL-CNT-ILS hydrogel, showcasing its sensitivity to temperature variations due to the Soret effect. In Figure S15a, voltage generation occurs solely in samples containing ionic liquids when one end is heated, confirming the thermoelectric behavior based on ions. Figure S15b indicates that the voltage output rises with the increase in absorbed radiation dose, while Figure S15c and S15d demonstrate that greater doses lead to increased resistance and reduced conductivity as a result of denser cross-linking. Figure S15e shows a variable Seebeck coefficient ranging from 1.68 to 5.62 mV/K, which improves with the dose. The Seebeck coefficient is a crucial parameter in thermoelectric materials, including hydrogels, measuring the voltage generated per unit temperature difference. Its unit is microvolts per Kelvin (μV/K) [3, 28, 29]. Figure S15f illustrates that limited cation movement caused by high cross-linking improves ion separation. Lastly, Figure S15g reveals the hydrogel's potential application as a flame sensor, where heat triggers current through thermoelectric generation [11].

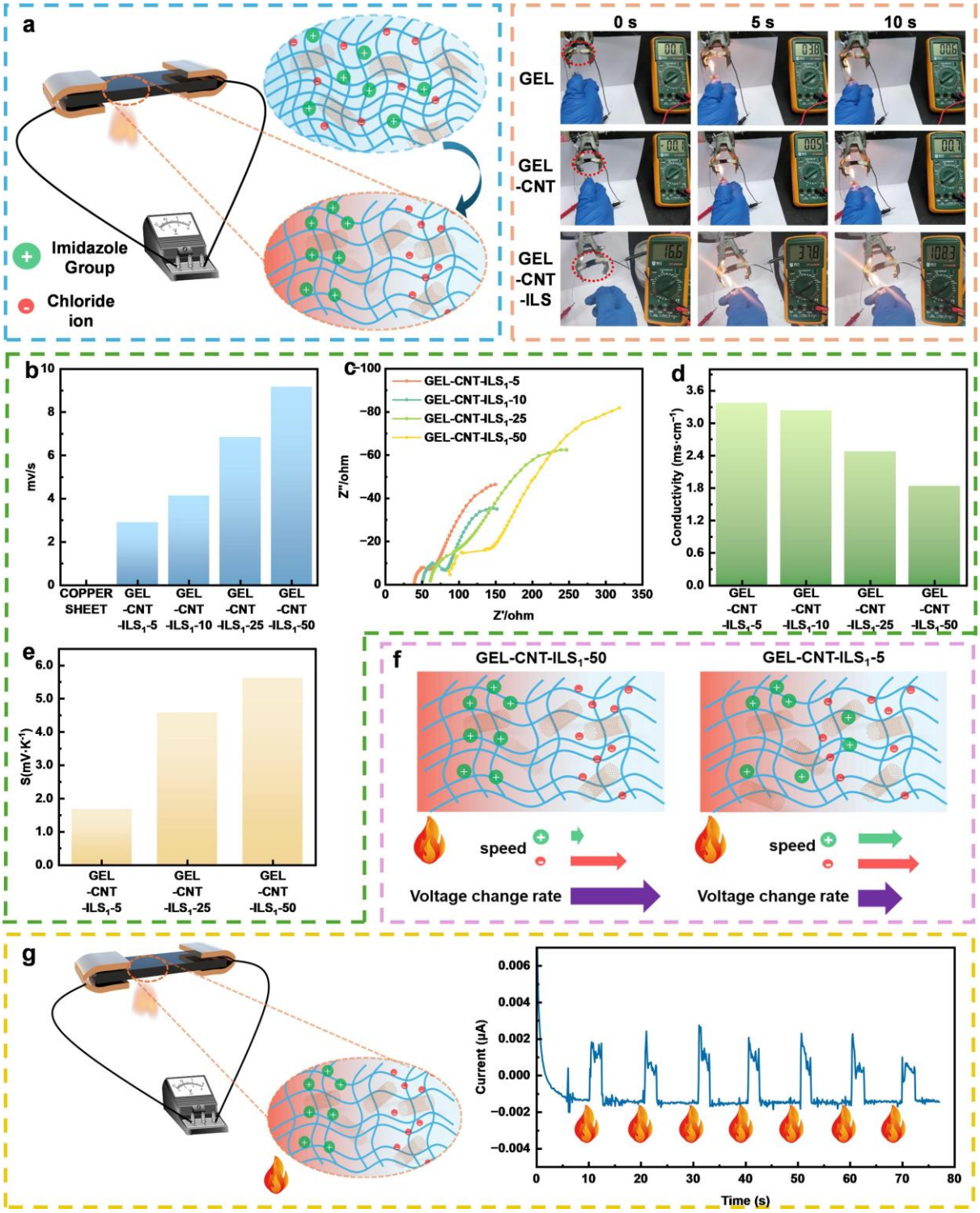

Figure S15. Thermoelectric and Flame-Sensing Performance of GEL-Based Composite Materials: (a) A schematic illustration of the experimental arrangement along with the results of GEL, GEL-CNT, and GEL-CET-ILS. (b) Change in voltage over time. (c) Electrochemical impedance spectroscopy (EIS) of the materials. (d) Conductivity measurements of the materials.

(e) Seebeck coefficients of the materials. (f) A schematic representation of the thermoelectric mechanism. (g) Illustration of the materials functioning as flame sensors. Adapted with permission from Ref. [11] Copyright 2025, Elsevier B.V.

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