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Carboxymethyl Cellulose–Polyaniline (CMC–PANI) Hydrogels as Multifunctional Binders for Sustainable Sodium-ion Batteries: Structure, Mechanism, and Performance Insights

Karunanidhi M.[1], Subhashini S.[2], Venckatesh R.[1], Rajeshwari Sivaraj[1]

[1]Department of Chemistry, Government Arts College, Udumalpet, Tamilnadu, India

[2]Arulmigu Palaniandavar Arts College for Women Palani,Tamilnadu, India

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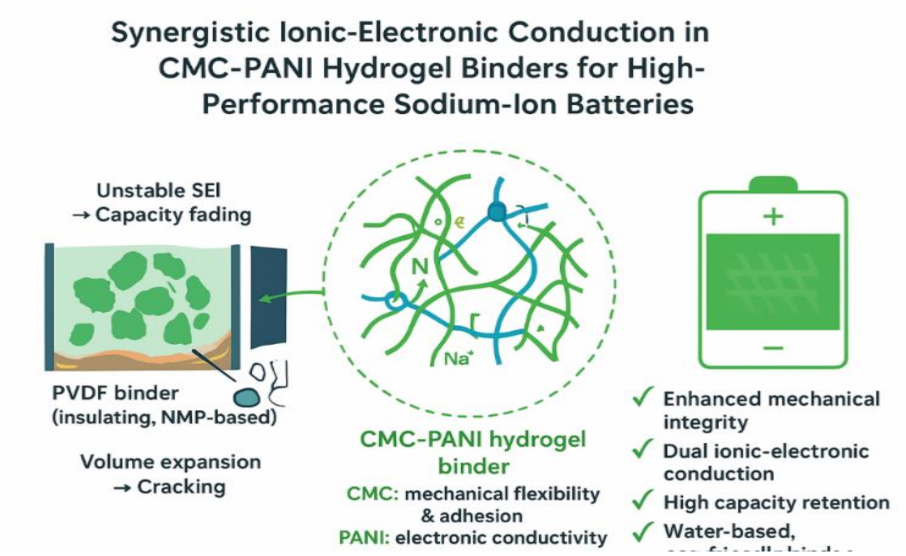
ABSTRACT

The growing global demand for energy and the urgent transition toward sustainable technologies have accelerated the search for efficient, low-cost, and environmentally benign energy storage systems. Sodium-ion batteries (SIBs) have emerged as a promising alternative to lithium-ion batteries (LIBs) due to the natural abundance and comparable electrochemical properties of sodium. However, the larger ionic radius of Na^+ results in significant volume fluctuations during cycling, leading to rapid capacity fading and poor mechanical stability of electrodes. In this context, the binder—though a minor component by weight—plays a crucial role in maintaining electrode integrity and electrochemical stability.

This review highlights the synthesis, structure, and function of carboxymethyl cellulose–polyaniline (CMC–PANI) hydrogel binders, a new class of multifunctional materials for SIBs. The CMC component offers strong adhesion, mechanical flexibility, and water processability, while PANI contributes electrical conductivity and electrochemical stability. The in-situ polymerization approach produces a semi-interpenetrating polymer network (semi-IPN) that synergistically integrates ionic and electronic conduction pathways. This composite binder enhances electrode cohesion, facilitates stable solid electrolyte interphase (SEI) formation, and improves rate capability and cycle life. The review also discusses structure–property relationships, mechanisms of performance enhancement, and the potential of CMC–PANI hydrogels as sustainable binders for next-generation sodium-ion batteries.

Keywords: Sodium-ion batteries; Hydrogel binders; Carboxymethyl cellulose; Polyaniline; Polymer network; Sustainable energy storage

Graphical Abstract



INTRODUCTION

The escalating global demand for energy and the pressing need to transition toward renewable energy technologies have driven intensive research into high-performance and sustainable energy storage systems. Among these, rechargeable batteries play a pivotal role in bridging the gap between intermittent renewable energy sources and consistent power supply. Lithium-ion batteries (LIBs) have dominated the portable electronics and electric vehicle markets for decades due to their high energy density, long cycle life, and excellent efficiency (Tarascon & Armand, 2001; Goodenough & Park, 2013). However, the limited and geographically concentrated lithium resources, coupled with increasing cost and safety concerns, pose significant challenges for their large-scale deployment, particularly in grid-level storage systems (Slater et al., 2013; Pan et al., 2020).

Sodium-ion batteries (SIBs) have emerged as a promising alternative owing to the natural abundance, low cost, and similar intercalation chemistry of sodium compared to lithium (Hwang et al., 2017; Fang et al., 2020). The redox potential of sodium (–2.71 V vs. SHE) is only slightly higher than that of lithium (–3.04 V vs. SHE), allowing comparable cell voltages and energy densities (Li et al., 2021). Moreover, the chemical similarity between Na and Li enables the adaptation of much of the existing LIB technology to SIB systems. Despite these advantages, SIBs face critical challenges that hinder their commercial viability—chief among them being the poor cycling stability and limited rate capability of electrode materials (Zhao et al., 2020; Yu et al., 2023).

The larger ionic radius of Na^+ (1.02 Å) relative to Li^+ (0.76 Å) induces substantial lattice strain, volume expansion, and mechanical degradation of the host material during sodiation and desodiation processes (Zhang et al., 2019). These structural stresses lead to pulverization of active particles, electrical disconnection, and continuous formation of an unstable solid–electrolyte interphase (SEI), resulting in severe capacity fading over repeated cycles (Li et al., 2020). While much attention has been devoted to developing advanced active materials, the binder—though often less than 5 wt% of the electrode composition—plays an equally critical role in ensuring electrode integrity and interfacial stability (Liu et al., 2016; Zhao et al., 2022).

Conventional binders such as polyvinylidene fluoride (PVdF), widely used in LIBs, rely on toxic and expensive organic solvents like N-methyl-2-pyrrolidone (NMP) for electrode processing (Lee et al., 2021). Moreover, PVdF exhibits poor mechanical flexibility and weak adhesion under large volume changes, leading to delamination and rapid capacity decay, especially in alloying or conversion-type anodes (Wang et al., 2022). These limitations have catalyzed the development of water-processable, mechanically robust, and multifunctional binders, particularly those based on natural polymers and hydrogels (Han et al., 2023).

Hydrogel-based binders, characterized by their three-dimensional cross-linked polymer networks and high water content, offer exceptional elasticity, strong adhesion, and excellent ionic permeability (Zhou et al., 2022). Among them, carboxymethyl cellulose (CMC), a derivative of cellulose, has gained considerable attention as a green, water-soluble, and biodegradable binder (Xu et al., 2021). Its abundant hydroxyl (–OH) and carboxyl (–COOH) groups enable strong hydrogen bonding and ionic interactions with active materials, ensuring good dispersion and adhesion (Yue et al., 2023). However, CMC is electronically insulating, which limits charge transport across the electrode.

To overcome this limitation, researchers have explored the integration of conductive polymers such as polyaniline (PANI) into the CMC framework. PANI is an intrinsically conducting polymer with high electrical conductivity, tunable oxidation states, and remarkable environmental stability (Zhang et al., 2020; Gao et al., 2022). When incorporated into CMC hydrogels, PANI forms an interconnected conductive network that enhances electron mobility while retaining the mechanical strength and adhesion provided by the CMC matrix (Li et al., 2022). The resulting CMC–PANI hydrogel composite thus serves as a multifunctional binder, providing ionic and electronic conductivity, flexibility, and chemical stability—key factors for long-cycle, high-rate sodium-ion batteries.

This review provides a comprehensive overview of CMC–PANI hydrogel binders, discussing their synthesis methods, structural characteristics, and electrochemical performance in SIBs. The synergistic effects between the CMC and PANI components, the underlying conduction mechanisms, and their influence on electrode

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stability and SEI formation are critically analyzed. Finally, the challenges and future directions for integrating such bio-derived, conductive hydrogel binders into next-generation energy storage systems are discussed.

Synthesis of CMC–PANI Hydrogels

The performance of CMC–PANI hydrogels depends critically on the uniform distribution of conductive PANI within the CMC matrix. In-situ oxidative polymerization is the most effective method to achieve this homogeneity, yielding a semi-interpenetrating polymer network (semi-IPN) structure. The process typically begins by dissolving sodium carboxymethyl cellulose in deionized water to form a viscous, three-dimensional hydrogel network, which acts as a scaffold. Aniline monomer is then introduced into the CMC hydrogel, where the porous structure facilitates its uniform diffusion and adsorption. Finally, polymerization is initiated using an oxidizing agent such as ammonium persulfate (APS), which converts aniline into polyaniline directly within the CMC network. During this step, the carboxylate groups ($–COO^-$) in CMC interact electrostatically with the positively charged aniline species, promoting uniform polymer growth.

The resulting composite exhibits intimate molecular-level interactions between the two polymers, forming a stable semi-IPN structure. Unlike simple blending methods, this approach prevents phase separation, ensures homogeneous conductivity, and enhances the mechanical and electrochemical properties of the binder.

Role and Performance in Sodium ion Battery Electrodes

CMC–PANI hydrogels function as multifunctional binders that simultaneously enhance electronic conductivity, mechanical durability, and interfacial stability. They have been successfully applied to both anode and cathode materials in SIBs.

Anode Applications

Anode materials such as SnO_2, Sb, and hard carbon experience severe volume changes during sodiation/desodiation. The CMC–PANI hydrogel binder effectively mitigates these challenges in several ways. First, its flexible hydrogel matrix provides mechanical buffering, accommodating volume fluctuations to prevent electrode cracking and maintain structural integrity over prolonged cycling. Second, the interconnected PANI network creates continuous electronic pathways, leading to enhanced conductivity that reduces charge-transfer resistance and improves rate performance. Third, the hydrogel's hydrophilic surface and chemical functionality promote the formation of a stable SEI layer, which reduces irreversible capacity loss and enhances long-term stability. Consequently, electrodes incorporating CMC–PANI binders have demonstrated higher reversible capacities and improved cycling performance compared to those with PVdF or pure CMC binders.

Cathode Applications

For cathodes, particularly Prussian blue analogues and layered oxides, CMC–PANI hydrogels offer several advantages. The PANI component enhances electronic conductivity, which supports high-rate charge–discharge processes for improved rate capability. Concurrently, the strong adhesion of the CMC provides structural reinforcement, preventing delamination and ensuring a cohesive electrode architecture during extended cycling. Furthermore, the water-based synthesis offers the benefit of **eco-friendly processing**, eliminating the need for toxic organic solvents and enabling sustainable electrode fabrication. As a result, cathodes using CMC–PANI binders exhibit higher specific capacities at high current densities and superior capacity retention over long-term operation.

Synergistic Mechanisms and Structure–Property Relationships

The exceptional performance of CMC–PANI hydrogels arises from synergistic effects between the two polymers, which together deliver properties unattainable individually. A key advantage is dual ionic–electronic conductivity; the hydrogel's hydrated matrix promotes ionic (Na^+) transport, while the PANI network provides continuous electronic pathways, a combination crucial for balanced charge transport in thick electrodes. The synthesis process also creates a hierarchical porosity, enhancing electrolyte infiltration, ion diffusion, and active site accessibility. At the nanoscale, molecular-level interactions such as hydrogen bonding and electrostatic forces between CMC and PANI ensure strong interfacial compatibility and mechanical stability, preventing phase separation during cycling. These interactions contribute to the composite's excellent mechanical flexibility and strength, allowing the electrode to accommodate repetitive expansion and contraction without structural failure.

CONCLUSION AND FUTURE PERSPECTIVES

CMC–PANI hydrogels represent a new generation of biopolymer-based, conductive, and eco-friendly binders for sodium-ion batteries. Their unique combination of ionic and electronic conduction, mechanical robustness, and aqueous processability provides a viable route to overcome the limitations of traditional PVdF-based binders.

Future work should focus on several key areas: optimizing the CMC-to-PANI ratio and polymerization parameters to tailor mechanical and electrical properties; investigating the interfacial chemistry between the binder and active materials using advanced spectroscopic techniques; scaling up aqueous processing methods for commercial electrode manufacturing; and exploring hybrid hydrogel systems that incorporate additional conductive or redox-active components.

Through such advancements, CMC–PANI hydrogels hold immense potential for realizing sustainable, high-performance, and low-cost sodium-ion batteries suitable for large-scale energy storage applications.

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