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Review

Physical Chemistry of Conductive Core–Shell Superabsorbent Polymers: Mechanisms, Interfacial Phenomena, and Implications for Construction Materials

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Featured Application

This research provides a foundation for sustainable packaging of smart additives within cementitious matrices to create self-monitoring concrete. The success of these conductive core–shell SAPs relies on complex interfacial phenomena, where the polymer shell's interaction with the cement paste ensures stable charge transport and efficient moisture exchange. While the swellable core drives internal curing and self-sealing to mitigate shrinkage, the conductive interface allows the entire structure to act as a distributed sensor. This synergy enables bridges and tunnels to monitor strain and humidity in real-time, resulting in resilient, low-maintenance, and multifunctional infrastructure.

Abstract

Conductive core–shell superabsorbent polymers (SAPs) are emerging as multifunctional additives for cementitious materials, combining moisture management with electrical functionality. In cement-based systems, a swellable polymeric core enables internal curing and crack-sealing through controlled water uptake and release, while a conductive shell introduces ionic and/or electronic charge transport, addressing key limitations of conventional non-conductive SAPs. This dual functionality provides a pathway toward smart cementitious composites with enhanced durability, self-sensing capability, and moisture-responsive behavior. This review focuses on the physical chemistry mechanisms governing conductive core–shell SAPs in cementitious environments, with emphasis on swelling thermodynamics, water transport kinetics, interfacial phenomena, and charge transport mechanisms. The roles of osmotic pressure, elastic network constraints, ionic effects, and pore solution chemistry are critically discussed, together with their impact on conductivity, hydration processes, microstructure development, and long-term performance. The relative contributions of ionic and electronic conduction are examined in relation to hydration state, shell morphology, and percolation of conductive networks. In addition, the relevance of core–shell SAP architectures to sustainable packaging is briefly discussed as a secondary application, illustrating how similar physicochemical principles—such as moisture buffering and functional coatings—apply beyond construction materials. Finally, key knowledge gaps are identified, including long-term stability in highly alkaline environments, trade-offs between swelling capacity and conductivity, environmental impacts of conductive phases, and the need for integrated experimental and modeling approaches. Addressing these challenges is essential for the rational design and practical implementation of conductive core–shell SAPs in next-generation cementitious materials.

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1. Introduction

1.1. Relevance of Conductive Core–Shell Superabsorbent Polymers to Sustainable Packaging Technologies

Multifunctional additives that combine mechanical enhancement with autonomous functionality have attracted increasing attention in recent cementitious materials research, reflecting a shift toward resilient and self-responsive infrastructure systems [1–6]. In particular, recent studies emphasize the integration of internal curing, crack mitigation and adaptive response mechanisms within a single material platform, driven by durability and sustainability requirements in modern construction [7]. Superabsorbent polymers (SAPs) are among the most extensively investigated systems in this context, owing to their ability to absorb and release large quantities of water under varying environmental conditions. Contemporary research highlights that SAP-induced swelling not only enables autonomous crack-sealing but also regulates internal relative humidity and hydration kinetics, contributing to enhanced durability under aggressive exposure conditions such as freeze–thraw and cyclic drying–wetting. Moreover, recent advances focus on tailoring SAP chemistry and structure to optimize water release timing and minimize adverse effects such as porosity increase, indicating a transition from empirical use toward design-driven SAP engineering [8].

In parallel with their increasing adoption in construction, SAP-based materials—especially those engineered with conductive or core–shell architectures—are also receiving growing attention in domains outside traditional civil engineering. One emerging area is sustainable packaging, where innovative polymer systems are actively sought to provide low-environmental-impact solutions with multifunctional performance. The mechanisms that make SAPs valuable in cementitious matrices, such as controlled aqueous uptake, interfacial transport, and tunable swelling kinetics, are directly relevant to modern packaging systems that require moisture buffering, environmental adaptability, and improved material reliability [9–11].

Conductive core–shell SAPs introduce functionalities such as antistatic behavior, adaptive moisture regulation, and compatibility with low-energy electronic components, all of which align with current trends toward intelligent and environmentally responsible packaging technologies [11–13]. For example, conductive hydrogel systems have been shown to exhibit humidity-dependent electrical responses, where swelling-induced structural changes directly influence conductivity, enabling their use in sensing and responsive packaging applications. Similarly, polymer composites incorporating conductive fillers such as carbon-based nanomaterials demonstrate enhanced electrical conductivity and antistatic properties, which are already exploited in electronic and protective packaging materials. These studies provide concrete evidence that the coupling between moisture responsiveness and electrical functionality, central to conductive SAPs, has been successfully utilized in related systems, supporting their relevance for advanced packaging applications. In the work of Wang et al., conductive hydrogels are categorized based on their composition and conduction pathways, while their key physicochemical properties are correlated with a wide range of functional applications. Figure 1 emphasizes the relationship between structural design, electrical behavior and performance, offering a general framework for understanding multifunctional hydrogel systems. This perspective supports the development of advanced materials that combine mechanical integrity with conductivity [11–13].

Accordingly, the investigation of these polymers' swelling thermodynamics, interfacial electrochemistry, and conductive behavior is not only critical for advancing cement-based systems but also directly supports the objectives of the Applied Sciences Special Issue "Innovative Materials and Technologies for Sustainable Packaging" [14]. The insights gained here can therefore contribute to the rational design of greener, multifunctional materials capable of reducing environmental impact while enhancing performance across multiple industries. Although conductive core–shell SAPs are relevant to a range of polymer-based technologies, including sustainable packaging, the present review primarily focuses on cementitious materials as the core application domain. Packaging-related examples are discussed in a secondary and illustrative context, with the aim of highlighting transferable physicochemical mechanisms rather than establishing parallel application areas.

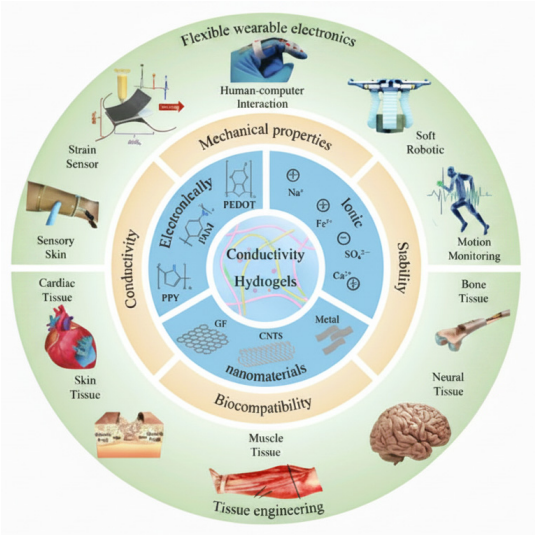


Figure 1. Classification, properties and applications of conductive hydrogels [12].

Despite the increasing work on superabsorbent polymers and conductive composites, there remains a clear research gap in understanding how swelling behavior, interfacial phenomena, and charge transport mechanisms are coupled within core–shell architectures. Existing studies typically address moisture regulation and electrical functionality separately, with limited attempts to integrate these processes into a single physicochemical framework.

The novelty of this review lies in the systematic analysis of conductive core–shell SAPs from a physicochemical perspective, explicitly linking swelling thermodynamics, interfacial electrochemistry, and transport mechanisms. By bridging these aspects, the work aims to provide design guidelines for multifunctional materials that simultaneously achieve moisture control and electrical functionality in cementitious systems.

1.2. Background and Motivation

In recent years, the construction materials field has seen a marked shift toward multifunctional additives that provide not only traditional mechanical enhancements like strength and durability but also added functional capabilities such as self-sealing, self-healing, and environmental adaptability. This trend reflects an industry's need for smarter materials that can autonomously respond to damage and extend service life, rather than relying solely on external repairs [15]. Superabsorbent polymers (SAPs) have emerged as a particularly promising class of multifunctional additives for cementitious systems. SAPs, being highly hydrophilic crosslinked polymers, can absorb many times their weight in water and trigger autonomous crack-sealing through volumetric swelling and controlled moisture release, which promotes ongoing hydration and localized self-healing processes,

effectively enhancing concrete durability under environmental stresses such as freeze–thaw and dry–wet cycles [16].

Conventional superabsorbent polymers (SAPs) in cementitious systems are highly effective at mitigating autogenous shrinkage and promoting self-sealing through water absorption and swelling, yet they also exhibit important limitations related to their single-function behavior and lack of multifunctionality. Studies on SAP-modified concrete demonstrate that while SAPs can absorb large amounts of mixing water and provide controlled internal curing, this same mechanism leads to the formation of macro voids and increased porosity after water release, changes that can reduce mechanical strength and negatively influence material performance if not carefully optimized [17]. Furthermore, SAPs function primarily as moisture regulators and do not contribute additional functional properties such as electrical conductivity or active sensing capabilities that are sought in emerging multifunctional construction materials; their role remains largely physical and single purpose rather than multifunctional in contrast to additives designed for smart structural functions [16]. These limitations underline why conventional SAPs, despite valuable internal curing and crack-sealing benefits, are not inherently suitable for advanced multifunctional applications without further modification or combination with other functional fillers [16,17].

Electrical conductivity is a key property for advancing concrete from a passive structural material to a truly smart and multifunctional composite. Conductive pathways within cementitious matrices enable the material to exhibit self-sensing capabilities, where changes in electrical resistivity or conductivity correlate directly with strain, stress, or damage, allowing concrete to autonomously monitor its structural health without external sensors. Research indicates that conductive fillers or aggregates must be incorporated to form a continuous or semi-continuous network capable of transmitting electrical signals. Conductivity mechanisms, percolation, electron hopping, and ionic conduction, enable detection of mechanical or environmental stimuli within the concrete matrix [18]. Beyond sensing, conductive composites play a crucial role in electromagnetic interference (EMI) shielding, where they block or attenuate harmful electromagnetic radiation. Integrating conductive aggregates creates cement materials capable of both piezo resistivity and effective EMI attenuation, yielding multifunctional protective structures suited for sensitive or high-tech infrastructure [19].

However, the integration of conductive functionality into SAP-based systems remains underdeveloped, particularly with respect to maintaining mechanical integrity while enabling stable transport pathways. This limitation highlights the need for structured material designs, such as core–shell architectures, that can decouple and optimize these competing requirements.

1.3. Core–Shell Design as a Multifunctional Approach

Core–shell structured materials offer enhanced mechanical stability and robustness compared with single-component systems because the core–shell configuration amplifies the material's resistance to external forces and stress, making the structure more enduring and dependable in demanding environments. This architecture also provides tunable interfacial properties by adjusting the composition, thickness, and configuration of the core and shell layers, allowing selective adsorption, optimized surface interactions, and precise control over how the core and shell interact with external species [20]. Additionally, core–shell particles can be synthesized with structural features that are adjustable at the nanoscale, such as tunable pore sizes in silica core–shell particles, enabling controlled structural expansion or accessibility of the internal domains for specific functions like separation or transport processes [21].

In core–shell systems designed for cementitious composites, the rationale for combining swelling behavior with electronic or ionic conduction is rooted in the complementary roles these functions play under moisture variation and mechanical stress. The swellable component, such as superabsorbent polymers, absorbs and releases water through osmotic and ionic mechanisms, regulating internal moisture and mitigating shrinkage while the associated swelling effect can promote crack self-sealing, reduced permeability, and in some cases a self-healing response by restoring continuity within the cement matrix [22]. When this moisture-responsive behavior is combined with a conductive core or phase, material can provide a continuous electronic or ionic conduction network and maintain stable conductive pathways despite volumetric changes induced by swelling and deswelling, as constrained swelling allows the conductive network to remain functional under deformation and environmental fluctuations [23].

Therefore, the present review specifically focuses on elucidating how the core–shell design allows for the controlled integration of swelling and conductivity, with an emphasis on interfacial coupling and transport phenomena as key determinants of multifunctional performance.

1.4. Scope and Structure of the Review

The physical chemistry section of the review aims to synthesize contemporary theoretical and computational approaches describing the control of swelling and ionic conductivity in hydrogels and hydrophilic polymer matrices. From a theoretical standpoint, mixing/elasticity models and chemical-equilibrium concepts explain how variables such as crosslinking density, polyelectrolyte charge, and ionic strength regulate the equilibrium of water and ion uptake, directly affecting ionic mobility [24]. In parallel, recent studies employing molecular dynamics simulations and microstructural modeling demonstrate how nanostructure (e.g., embedded conductive nanomaterials) correlates with the formation of continuous ionic or electronic pathways, and how micro-electrical processes (such as dipole reorientation or interfacial potential barriers) govern overall conductivity. This section also emphasizes the importance of combined experimental techniques (electrical spectroscopy, pulse-response methods, structural microscopy) together with computational tools for quantifying charge transport mechanisms [25]. By establishing these fundamentals, the review provides a framework for interpreting how design variables in core–shell SAPs influence swelling capacity, conductivity, and mechanical stability [24,25].

The applications section anchors the theoretical discussion in the modern literature on polymer-modified cements and “smart” construction materials [26,27]. It describes how the swelling and conductive properties of core–shell SAPs can influence critical concrete parameters, ranging from workability and curing behavior to microstructural integration and long-term durability [26]. Reviews on polymer reinforcement and conductive composites show that incorporating tunable, electro-active additives yields dual benefits: internal moisture control and shrinkage mitigation (through SAP action), alongside the creation of electrical/EM pathways (through conductive fillers or ionic transport) [26,27]. At the same time, the studies highlight challenges such as chemical compatibility with the alkaline cement environment, impacts on mechanical strength, and long-term conductivity stability, and they propose experimental protocols that integrate in situ electrical measurements with mechanical and durability testing. The review therefore evaluates how these empirical insights can support the adoption of conductive core–shell SAPs in next-generation concrete technologies [27] (Figure 2). The integration of cementitious and packaging-related literature in this review is achieved through their shared physical chemistry foundations, including swelling thermodynamics, moisture transport, interfacial interactions, and con-

ductivity control. These mechanisms are discussed within a unified framework, rather than through a parallel application-based comparison.



Figure 2. The framework of this review article.

2. Chemical and Structural Design of Conductive Core–Shell SAPs

2.1. Core Composition

Superabsorbent polymers (SAPs) and particularly core–shell hydrogel systems are typically composed of three main elements: (i) the polymer network forming the core structure (e.g., acrylic-based, polysaccharide-based or hybrid networks), (ii) crosslinking agents controlling network density and mechanical integrity and (iii) functional additives or shell layers (e.g., conductive fillers, nanoparticles or ionic species) that tailor physicochemical and transport properties. Recent studies have increasingly focused on the design of multifunctional systems, where composition and architecture are engineered to simultaneously optimize swelling capacity, mechanical stability and electrical conductivity.

Hydrophilic networks composed of acrylamide (AAm), acrylic acid (AA), and related acrylate copolymers form the shell of core–shell superabsorbent polymers, providing a water-absorbing matrix that surrounds the core and can be tailored in composition, structure, and mechanical properties [28,29]. Copolymerizing AAm with AA or acrylates produces three-dimensional polymer networks whose equilibrium swelling and mechanical behavior can be tailored through comonomer ratios, crosslinking density, and polymerization conditions. Acrylic acid introduces ionizable carboxyl groups that enhance osmotic swelling in low-ionic-strength media, while acrylamide units contribute to network cohesion and help moderate brittleness [29]. The microstructure of the network, including porosity, pore size distribution, and homogeneity, which are set during polymerization, strongly affects swelling kinetics, with more open networks absorbing water more rapidly. Many practical hydrogel formulations exploit blends or grafts to combine enhanced mechanical resilience and high swelling capacity [28]. Finally, selecting appropriate comonomer ratios and polymerization methods also influences chemical stability in challenging environments such as varying pH or salt concentration [29].

Crosslinking density is a key structural parameter controlling the equilibrium swelling and mechanical properties of hydrogels. Increasing crosslinker concentration reduces the network mesh size, which limits the maximum water uptake while increasing the elastic modulus of the material [30]. Experimental studies on chemically crosslinked hydrogels confirm that denser networks swell less, reflecting the balance between polymer–solvent interactions and the elastic restoring forces imposed by the crosslinked strands. Both macroscopic and nanoscale analyses indicate that adjusting crosslink density provides a straightforward route to tailor swelling behavior and mechanical stiffness, enabling rational design of hydrogels for a wide range of applications [31]. A comparison of recent studies reveals several consistent trends. Increasing crosslinking density generally reduces swelling capacity but improves mechanical stability, while the incorporation of conductive fillers enhances electrical performance at the expense of network flexibility. Core–shell architectures enable partial decoupling of these effects, allowing independent tuning of swelling and conductivity. Additionally, systems with hierarchical or porous structures tend to exhibit improved ion transport due to enhanced connectivity to conductive pathways.

2.2. Shell Composition

Conductive hydrogels can be endowed with electrical conductivity by incorporating conductive components into their shell. Intrinsically conducting polymers such as polyaniline (PANI), polypyrrole (PPy), and PEDOT facilitate charge transport through their π conjugated backbones [32]. Carbon-based nanostructures, including carbon nanotubes and graphene, form interconnected networks within the polymer matrix, enhancing both electrical conductivity and mechanical properties [33]. The effectiveness of these conductive fillers depends on their dispersion and interaction with the polymer network. In addition, metal oxides and hybrid fillers are being explored in the literature to provide extra functionalities, though the dominant approaches in these reviews remain conductive polymers and carbon nanostructures [32].

In solid and composite materials, electrical conductivity can arise from electronic carriers (electrons or holes) and/or ionic species under an applied field. In mixed systems, such as structural battery electrodes, electronic transport occurs primarily through percolating conductive networks, while ionic conduction takes place via mobile ions in the electrolyte phase, with the relative contributions of the two mechanisms depending on material composition and microstructure [34]. In polymer-based mixed conductors, the balance between ionic and electronic conductivity can be tuned by additives or compositional modifications, allowing control over overall transport properties and functional performance [35]. Understanding and managing these mechanisms is critical for designing materials with tailored conductivity for energy, sensing, or multifunctional applications [34,35].

2.3. Synthesis Strategies

Synthesis strategies like graft polymerization, in situ polymer coating, and layer-by-layer (LbL) assembly are crucial for tailoring the morphology of conductive polymer composites. Grafting is a technique used to modify or functionalize polymers by incorporating side chains or branches that are covalently bonded to the primary polymer backbone [36]. In situ polymer coating involves polymerizing monomers directly in the presence of substrates or nanofillers, forming conformal polymer layers on their surfaces that enhance interfacial contact and uniformity within bio-nanocomposite systems. The layer-by-layer assembly technique builds multilayer structures through alternating deposition of oppositely charged components, offering nanoscale control of thickness and composition, which directly impacts how porous or compact the final structure is and how well the conductive phase forms uninterrupted channels [37].

The morphology of conductive polymer composites, including features such as thickness, uniformity, porosity, and the continuity of conductive networks, is critical for determining the electrical percolation threshold and overall performance in applications like sensors, coatings, and energy devices [38]. The way composites are synthesized strongly affects these morphological characteristics, as different processing parameters influence how fillers and polymer chains arrange within the matrix [38,39]. Careful control of synthesis conditions, such as in situ polymerization or solution processing, can promote a well-dispersed conductive network with minimal agglomeration, leading to more uniform coatings and continuous conductive pathways that enhance overall conductivity [39]. Conversely, poor dispersion or non-uniform growth of conductive phases can create voids or discontinuities, increase resistance and reduce connectivity. In general, synthesis strategies that favor aligned or interconnected filler networks reduce the percolation threshold and improve electrical performance [38]. Overall, tailoring the synthesis method directly influences the morphology of the conductive phase, which in turn governs the electrical, mechanical, and functional properties of the composite [38,39].

Reported differences in swelling and conductivity are often attributed to material design, yet in many cases they primarily reflect variations in synthesis parameters, such as initiator concentration, polymerization temperature and stirring rates. These conditions fundamentally dictate the crosslinking density and the homogeneity of the polymer network, directly impacting water release kinetics in cementitious matrices. For instance, high crosslinking densities systematically reduce swelling but may artificially enhance apparent conductive stability by limiting network deformation under hydration. Furthermore, the synthesis route is decisive; in situ polymerization tends to produce more homogeneous conductive pathways compared to post-functionalization, which often leads to phase segregation and disrupts the continuity of the conductive network.

To provide a clearer overview of the structural diversity and functional specialization of these systems, Table 1 synthesizes representative material architectures discussed in the recent literature. By categorizing the core–shell components alongside their dominant mechanisms and primary applications, the table highlights how precise chemical engineering of the polymer network dictates the macro-scale performance in both construction and smart packaging sectors.

Table 1. Comparison of material composition, mechanisms and applications for core–shell SAP systems.

Material Architecture	Key Core–Shell Components	Dominant Mechanisms	Primary Applications
Conventional SAP	AA, AAm	Osmotic pressure and Elastic network constraints	Internal curing and mitigation of autogenous shrinkage
Conductive Core–Shell	SAP core/PANI, PPy or PEDOT shell	Electronic transport via π-conjugated backbones	Self-sensing concrete and real-time monitoring
Nanocomposite SAP	SAP core/CNTs or Graphene fillers	Percolation network formation and charge transport	Electromagnetic interference (EMI) shielding
Smart Packaging SAP	Hydrophilic polymeric core/conductive shell	Controlled aqueous uptake and adaptive moisture regulation	Humidity buffering and smart sensing of spoilage

2.4. Physicochemical Characterization

Physicochemical characterization of polymer composites and hydrogels relies on a combination of analytical techniques to provide a comprehensive understanding of ma-

terial structure, composition, and morphology. Swelling tests are employed to quantify solvent uptake and network expansion, which reflect the mesh size, porosity, and water retention capacity of hydrogels; these measurements are often combined with thermal and spectroscopic techniques to probe hydration and polymer chain mobility. Differential scanning calorimetry (DSC) examines thermal transitions such as glass transition and melting points, providing insights into polymer phase behavior and structural changes upon hydration. Fourier transform infrared spectroscopy (FTIR) enables identification of specific chemical bonds and interactions, revealing molecular-level changes associated with swelling and network dynamics [40]. X-ray photoelectron spectroscopy (XPS) complements these analyses by determining the elemental composition and chemical states at material surfaces, which is particularly useful for assessing surface modifications and interactions in nanocomposite hydrogels [41,42]. Finally, scanning and transmission electron microscopy (SEM/TEM) provide high-resolution imaging of micro- and nanostructural features, including particle dispersion, network continuity, and morphology of conductive or reinforcing phases; this has been demonstrated both in polysaccharide-based hydrogels with embedded nanoparticles and in cellulose-based conductive paper composites [41,43].

Conductivity measurements are a key part of evaluating how well conductive polymers and their composites transport charge, and impedance spectroscopy (often called electrochemical impedance spectroscopy, EIS) and the four-point probe method are two widely used techniques. Impedance spectroscopy measures the material's response to an alternating electrical signal over a wide frequency range, allowing separation of different resistive and capacitive contributions such as bulk conductivity, interfacial effects, and charge transport mechanisms. This makes EIS particularly useful for identifying ionic and electronic transport processes in polymer-based systems [44]. The four-point probe method, by contrast, directly measures steady-state electrical resistance by applying current through two outer probes and measuring voltage between two inner probes; this configuration minimizes artifacts from contact resistance and is well suited for determining the bulk conductivity of thin films or bulk samples with higher uniformity [45]. Together, these methods enable both mechanistic understanding and accurate quantification of conductivity in conductive polymer materials [44,45].

3. Swelling Thermodynamics and Water Transport Mechanisms

3.1. Flory–Rehner and Osmotic Pressure Fundamentals

Swelling of crosslinked polymer networks is fundamentally driven by a balance between osmotic pressure and elastic resistance, as described by the Flory–Rehner theory. In this framework, the thermodynamic force promoting solvent uptake arises from the increase in entropy when solvent mixes with the polymer network, which creates an osmotic pressure that draws solvent into the gel [46]. This mixing contribution is counteracted by the elastic restoring force due to stretching of the polymer chains as the network expands, leading to an equilibrium swelling state when these opposing forces balance. The Flory–Rehner formalism combines the Flory–Huggins solution theory for polymer–solvent interactions with rubber elasticity theory for the network, producing an expression that predicts how swelling depends on crosslink density and polymer–solvent affinity [47]. Modifications of this model also incorporate effects such as Donnan equilibrium in ionic gels, where the difference in mobile ion concentration between the polymer interior and external solution further contributes to the osmotic driving force for swelling [48].

In polymeric networks, fixed charges along the polymer backbone and ionic strength of the surrounding solution strongly influence swelling through additional osmotic contributions, because ionized groups within the gel attract counterions and generate an internal osmotic pressure [46]. Charged groups within the network attract counterions,

creating a Donnan osmotic pressure that increases solvent uptake to balance mobile ion differences across the gel boundary; this can greatly enhance swelling compared to neutral networks [48]. The degree of crosslinking controls the elastic resistance to swelling higher crosslink density restrains network expansion and reduces equilibrium swelling, whereas lower crosslinking increases swelling capacity [47]. Ionic strength of the external solution modulates counterion distribution—higher ionic strength compresses the Donnan layer, reduces osmotic pressure differences, and thus decreases swelling, while lower ionic strength enlarges the osmotic driving force [46,48]. Overall, swelling equilibrium results from a complex interplay of mixing, elastic, and ionic osmotic pressures, with fixed charges and solution conditions tuning the balance and thus the swelling response of polymer gels [47,48]. It should be emphasized that the Flory–Rehner framework is employed here as a conceptual thermodynamic reference rather than as a fully predictive model for conductive core–shell SAP architectures. The classical theory assumes a homogeneous, isotropic polymer network undergoing uniform swelling, whereas core–shell SAPs exhibit pronounced spatial heterogeneity, constrained swelling, and interfacial coupling between chemically and mechanically distinct regions. As a result, deviations from Flory–Rehner predictions are expected when swelling is locally restricted by a shell layer or when conductive phases alter network connectivity and elasticity.

3.2. Water Uptake Kinetics in Core–Shell Systems

In core–shell systems, water uptake proceeds through layered diffusion, where solvent must first penetrate the shell before reaching the core, making the shell a primary diffusion barrier [49]. Experimental studies show a two-stage kinetic behavior: initial penetration into the outer shell and subsequent diffusion into the core, with each layer's structure and composition affecting the overall uptake rate [50]. Factors such as cross-link density and filler content in the shell can hinder or facilitate diffusion, altering how quickly equilibrium swelling is reached, due to how water molecules interact and diffuse through the polymer network. Because the shell typically acts as the primary barrier to solvent entry, its properties largely determine the early kinetics of water transport, while diffusion into the core becomes significant over longer time scales as the solvent front progresses inward [49]. These layered diffusion effects are crucial for designing hydrogel systems where controlled uptake, responsiveness, or release profiles are needed [49,50].

In the work of Zhou et al., the swelling mechanism is illustrated as a three-stage process involving water penetration, network expansion and partial release/retention. Figure 3 highlights how polymer chains with ionic groups attract water molecules, leading to osmotic swelling and structural rearrangement. It also shows that molecular weight and crosslinking density influence the extent of expansion and the ability of the network to retain water. Higher molecular weight networks exhibit greater entanglement and retention, whereas lower molecular weight systems display reduced swelling capacity and faster release [8].

In core–shell architectures, swelling kinetics can be regulated either by the shell or by the core, depending on relative diffusivity and network structure. A shell-controlled regime occurs when the outer layer is more restrictive, due to higher crosslink density or additional fillers, slowing water ingress and dominating the overall swelling behavior [51]. Conversely, a core-controlled regime arises when the shell is more permeable than the core, causing the diffusion rate inside the core to limit swelling. This interplay means that even with identical external conditions, the core's properties (e.g., polymer density, crosslinking) can become rate-limiting if the shell allows fast penetration [52]. Core–shell systems engineered with this perspective can tailor the sequence and rate of swelling across layers,

which is particularly relevant for applications like controlled release or shape-changing materials [51,52].

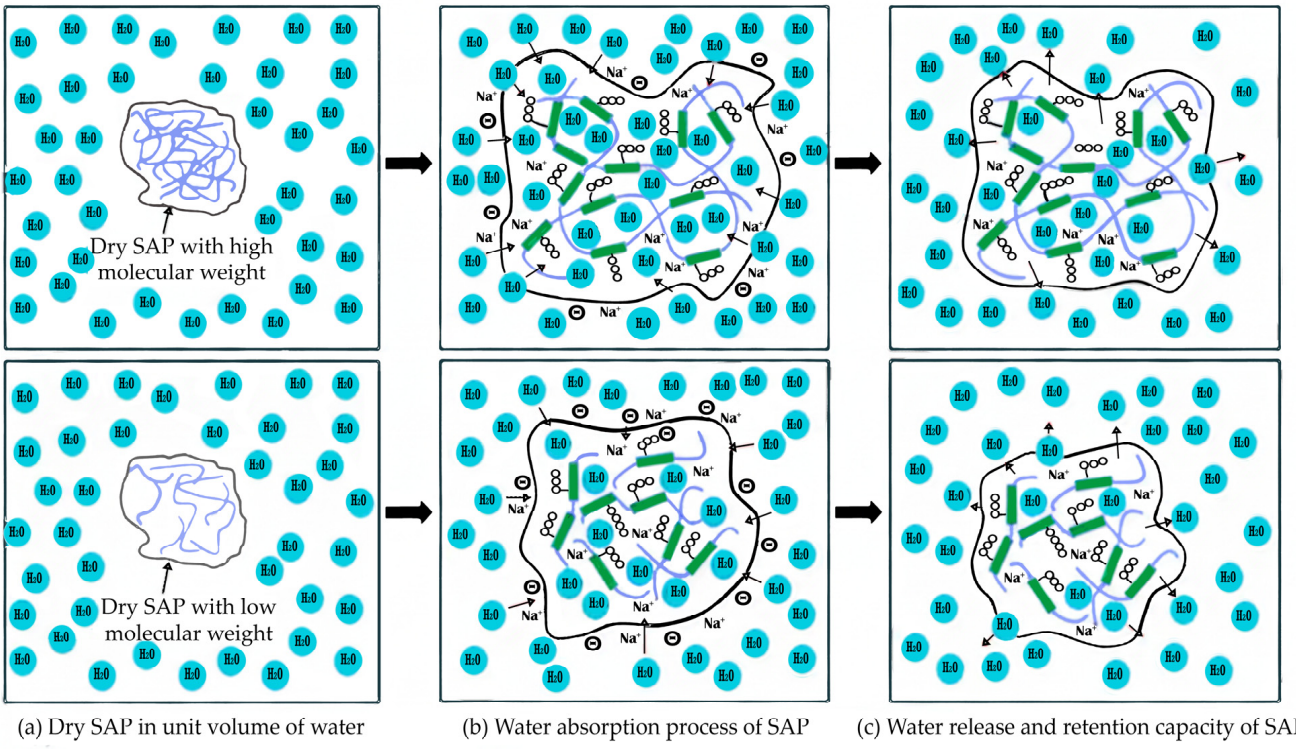


Figure 3. Absorption and desorption behaviors of SAP with high or low molecular weights [8].

Swelling in core–shell hydrogels directly influence the conductivity of the shell layer through coupled structural and transport effects. As water penetrates and expands the shell, polymer chains become more mobile and pore sizes increase, facilitating ionic transport and enhancing conductivity; conversely, restricted swelling or network densification reduces conductive pathways, lowering ion mobility [53,54]. Furthermore, ionic conductivity correlates strongly with mesoscopic pore structure and crosslinking density, so that larger pores and reduced crosslinking, often associated with swelling, increase ionic transport, whereas compact networks hinder it, linking morphological changes directly to charge transport behavior [55]. This coupling demonstrates that swelling not only alters hydrogel volume but actively modulates shell conductivity, enabling stimuli responsive behavior in conductive hydrogel systems [54,55].

3.3. Influence of Cementitious Pore Solution Chemistry

In cementitious pore solutions with very high alkalinity (pH ≈ 12–13), the ionization behavior of superabsorbent polymers (SAPs) is strongly affected. Specifically, increased pH promotes the dissociation of acidic functional groups in SAPs, enhancing electrostatic repulsion and water uptake, while simultaneously modifying ion-exchange processes within the polymer network due to the uptake of hydroxide ions [56]. Under strong alkaline conditions, interactions between ionized polymer chains and dissolved ions alter the contribution of ionized sites to osmotic pressure, affecting both swelling magnitude and kinetics [57]. Further, swelling exhibits a pH-dependent maximum, indicating that although higher pH initially enhances polymer ionization and swelling, extreme alkalinity leads to changes in network response within cementitious environments [58].

In cementitious pore solutions, divalent calcium ions (Ca^{2+}) strongly interact with negatively charged functional groups of superabsorbent polymers (SAPs), leading to charge

screening and partial neutralization of the polymer network, which reduces electrostatic repulsion between chains and significantly suppresses swelling capacity compared to monovalent ionic environments [56]. In the presence of trivalent aluminum ions (Al^{3+}), we observed even stronger effects, as Al^{3+} ions form ionic complexes and chelation bonds with carboxylate groups, effectively acting as additional physical cross-links within the SAP network and causing pronounced polymer contraction and loss of water absorption capacity [59]. A broader polyelectrolyte perspective showed that multivalent counterions promote counterion condensation, where ions accumulate in close proximity to charged polymer backbones, thereby reducing the effective charge density, decreasing osmotic pressure, and limiting network expansion, providing a theoretical and physicochemical framework that explains the experimentally observed swelling suppression of SAPs in Ca^{2+}- and Al^{3+}-rich cementitious environments [60].

Swelling capacities reported in deionized water are not representative of cementitious environments and tend to significantly overestimate SAP performance. In realistic pore solutions, high ionic strength and the presence of Ca^{+2} ions suppress swelling through charge screening and ionic crosslinking, often reducing absorption by several-fold. Consequently, studies based solely on idealized media may lead to misleading conclusions regarding internal curing efficiency in concrete.

4. Charge, Transport and Conductive Mechanisms

4.1. Electronic vs. Ionic Conductivity

In conjugated conducting polymers, the electronic conductivity fundamentally arises from the extended π-conjugation along the polymer backbone, where alternating single and double bonds create delocalized π-electron systems that enable charge transport once doped. In their undoped state, these polymers behave like semiconductors, but upon oxidation or reduction (chemical or electrochemical doping), localized charge carriers such as polarons and bipolarons form along the chain and introduce energy states within the bandgap that significantly enhance conductivity [61]. At increasing doping levels, these charged defects (polarons/bipolarons) can overlap and interact both along a single polymer chain (intrachain) and between adjacent chains (interchain) through π–π stacking, leading to improved electronic percolation pathways and higher carrier mobility [62].

In typical swollen polymer gel electrolytes, such as slide-ring gels swollen with ionic liquids, the ionic conductivity primarily arises from the movement of solvated ions through the liquid phase contained within the network pores rather than conduction along polymer chains, and conductivity changes with the degree of swelling because larger solvent volumes and interconnected liquid pathways facilitate higher ion mobility [63]. Additionally, in gel polymer electrolytes based on ionic liquids, the enhanced mobility of ions is attributed to the structure and dynamics of the IL-based gel network, where proper dispersion and interaction of ionic liquid molecules within the polymer matrix create more continuous ionic pathways and facilitate ion transport through liquid-like environments within the swollen polymer network [64].

In the proposed core–shell SAP system, both conduction mechanisms coexist, with their contribution depending on the structural design and operating conditions. The conductive shell constitutes the main electron transport pathway, the efficiency of which is determined by the thickness, morphological continuity and the degree of contact between the particles that allows the creation of extensive conductive networks. In contrast, the swollen core creates cohesive liquid phases that facilitate ionic transport, which often prevails under conditions of high hydration. At the same time, the interfacial core–shell interaction can enhance synergistic phenomena, where the ionic flow affects the local charge distribution in the shell, while the shell contributes to the partial electronic interconnec-

tion of the system. Therefore, the overall conductivity results from a dynamic balance between ionic and electronic components, which is controlled by the degree of hydration, microstructure and continuity of the conductive phase.

4.2. Percolation Networks in Core–Shell Designs

In percolation networks with core–shell designs, the conductive shell around particles significantly influences the formation of continuous pathways by controlling how conductive regions connect within the polymer matrix. In core–shell structured fillers, the shell can enhance compatibility with the polymer, increase interfacial contact, and facilitate charge transport along interconnected networks, which promotes percolation behavior at lower filler loadings than would occur without a well-engineered shell [65]. At the same time, the addition of conductive fillers of specific morphology and distribution supports the establishment of percolation networks by forming interconnected conductive pathways throughout the composite, improving overall electrical connectivity and transport properties [66]. These combined effects, conductive shells improving interface contact and well-dispersed fillers creating an interconnected network, are essential for achieving efficient percolation in core–shell composite systems [65,66].

In hydrated polymer electrolytes, water molecules play a central role in forming continuous ionic conduction pathways by creating interconnected channels within the polymer network that facilitate ion transport. Such water-filled pathways arise from the polymer's pore structure and water uptake, where the connectivity and size of these pores determine how far and how easily ions can migrate through the network, effectively reducing the energy barrier for ionic motion as water content increases and pore channels become more continuous through the gel matrix [24]. Moreover, structuring hydrogels to develop designed water channels enhances this effect; lamellar water channels form upon hydration and correlate strongly with enhanced proton conduction, indicating that ordered water networks at the nanoscale significantly mediate ionic transport in polymer electrolytes [67].

4.3. Dynamic Coupling of Swelling and Conductivity

Hydration plays a critical role in determining the ionic conductivity of swelling materials, as the uptake of water directly facilitates charge transport pathways. Conductivity is mainly protonic and exhibits protonic character related to ion hydration shell acidity. In swelling clays, protonic conductivity increases with water content due to enhanced proton mobility within the interlayer water, where absorbed water molecules enable proton diffusion through hydrogen-bonded networks [68]. Similarly, in polyacrylamide hydrogels, a critical hydration value (hc $\approx$ 0.15 g water/g dry polymer) has been identified, below which water molecules are irrotationally bound to primary hydration sites and do not contribute to the dielectric response, corresponding to the completion of the first hydration layer. Above this threshold, additional water forms continuous water-filled pathways that dominate proton transport, providing direct evidence that conductivity proceeds primarily through the aqueous phase rather than along the polymer backbone [69].

The effects of drying and repeated wet–dry cycles on polymer electrolyte systems are significant because alternating hydration and dehydration lead to structural changes that influence conductivity and mechanical integrity. In polymer electrolyte fuel cells, repeated cycles of membrane swelling and shrinkage caused by humidity fluctuations can produce mechanical stress, altering pore structures and water management properties that in turn affect ionic resistance; dehydration increases resistance and reduces water-mediated transport pathways, while rehydration lowers resistance, but repeated structural changes can degrade performance over time [70]. Although most detailed studies focus on fuel

cell components rather than isolated electrolyte films, these dynamics illustrate how cyclic hydration/dehydration leads to changes in pore network connectivity and ionic transport resistance, thereby impacting the coupling between swelling and ionic conductivity when materials repeatedly gain and lose water [71].

4.4. Environmental and Mechanical Influences

Mechanical strain and microstructural evolution strongly influence charge transport by altering the continuity of ionic and electronic pathways. In all-solid-state battery cathodes, volume changes in active materials generate significant local stresses during cycling, which promote crack initiation and propagation, leading to degradation of conductive pathways [72]. The role of microstructure is further highlighted in tape-cast solid electrolytes, where materials with very high relative density (99.8%) exhibit superior mechanical properties; this densest material was deliberately selected for mechanical testing due to the pronounced effect of porosity on mechanical behavior. The same study shows that bimodal grain size distribution negatively affects mechanical performance, as smaller grains increase grain boundary density, while pores within large grains or the presence of large grains themselves reduce fracture stresses [73]. These mechanically induced defects directly impact charge transport, as applied mechanical stress, particularly aggressive uniaxial compression during electrode preparation and cycling, can initially enhance conductivity by improving interfacial contact, but simultaneously increases the risk of cracking and material deformation, which ultimately disrupts homogeneous ionic and electronic transport pathways [74].

5. Polymer–Cement Chemical Interactions

5.1. Polymer–Cement Chemical Interactions

Polymer–cement interfaces are dominated by specific molecular interactions that include hydrogen bonding, electrostatic (ionic) binding, and surface complexation-like adsorption on charged cementitious phases. Polar functional groups such as hydroxyl and carboxylate groups on polymer chains can form hydrogen bonds with surface sites on inorganic materials, forming an interfacial polymer layer that enhances compatibility and cohesion in cement-based systems [75–77]. Molecular dynamics simulations and experimental analyses further demonstrate that polymer functional groups can participate in ionic binding through salt bridge-type interactions mediated by alkaline metal cations, which promote electrostatic attraction between charged polymer segments and ionic sites on mineral surfaces, thereby reinforcing interfacial attachment beyond purely physical adsorption [78]. Beyond these interactions, adsorption processes on cementitious surfaces, especially those involving charged polymers with specific functional groups, exhibit characteristics consistent with surface complexation phenomena, whereby polymer functional groups anchor onto surface sites with different affinities depending on their charge and chemical nature, influencing both fluidity and the interfacial properties of the system [79].

Reactions of polymers with cement hydration products, specifically $Ca(OH)_2$ and calcium silicate hydrate (C–S–H), play a significant role in modifying hydration kinetics and microstructural development in polymer-modified cementitious systems. Highly carboxylate colloidal polymers interact with cement primarily through surface adsorption rather than calcium complexation, where negatively charged polymer particles adsorb onto cement grain surfaces via electrostatic interactions, forming a polymer covering layer that inhibits C–S–H nucleation due to unfavorable interfacial energy between the polymer and C–S–H phases. The same study further shows that enrichment of polymer surfaces with Ca^{2+} and silicate species can transform the polymer layer into favorable nucleation sites for C–S–H, potentially accelerating hydration under specific chemical conditions [80]. In

polymer–cement waterproof coatings and similar composite materials, polymer emulsions containing functional groups such as carboxyl moieties react with Ca^{2+} in the liquid phase, effectively reducing the yield of $Ca(OH)_2$ and altering the phase distribution of hydration products, which in turn affects both the quantity and morphology of C–S–H formed [81]. At the micro- to nanoscale, chemical modification of C–S–H by polymers such as hydroxyl-terminated polydimethylsiloxane shows that polymer chains can connect with C–S–H gel particles through covalent or coordination bonds, leading to changes in C–S–H morphology, connectivity, and interlayer structure, demonstrating a direct chemical interaction between the polymer and the main cement hydration product [82].

5.2. Interfacial Water Dynamics

SAPs act as internal reservoirs that drive internal curing by absorbing a large fraction of the mixing water and releasing it into the cementitious matrix throughout early hydration. Both retentive and non-retentive SAPs initially saturate with water and subsequently redistribute that entrained water into the surrounding paste during hydration, mitigating autogenous shrinkage and maintaining internal relative humidity as the cement matrix densifies [83]. Neutron tomography measurements demonstrate that the effectiveness of SAP-driven internal curing is strongly governed by the timing of water release, where SAPs releasing stored water predominantly after final setting provide effective mitigation of autogenous shrinkage, while SAPs that release most of their water prematurely shortly after final setting show only partial shrinkage mitigation due to insufficient water availability at later hydration stages [84]. The chemical structure of SAPs also influences internal curing efficiency, polymers with higher densities of anionic functional groups absorb pore solution more rapidly and release water in a manner that better counteracts self-desiccation and supports ongoing hydration compared to SAPs with fewer anionic sites, demonstrating that molecular design affects SAP-driven internal curing behavior [85].

Differences in curing regimes significantly affect the interpretation of SAP performance. Sealed curing conditions tend to amplify the perceived benefits of internal curing, whereas external water curing can often mask SAP functionality by providing an alternative moisture source. Furthermore, the effectiveness of SAPs is highly sensitive to thermal boundaries; for instance, high temperature curing (e.g., steam curing) accelerates hydration kinetics, which may force a premature release of entrained water compared to ambient conditions. Critical evaluation suggests that the timing of this moisture exchange is not merely a function of the polymer's chemical structure, such as its crosslinking or ionic charge density, but is also governed by external thermal and humidity boundaries. These boundaries dictate the complex transition from early-stage osmotic pressure-driven flow to later-stage humidity-dependent desorption, meaning that inconsistent curing protocols across studies can lead to conflicting conclusions regarding shrinkage mitigation and long-term hydration behavior.

The thermodynamics and kinetics of water release from SAPs are key determinants of internal curing performance in cementitious systems. SAP desorption behavior reflects distinct kinetic regimes, with retentive and non-retentive polymers showing different temporal water release profiles that govern when entrained water becomes available to the hydrating matrix, as observed by differences in the saturated cavity behavior monitored by X-ray tomography and NMR [83]. The addition of SAPs also alters the overall hydration reaction thermodynamics in low-water systems, as evidenced in alkali-activated slag pastes, where internal curing delays the peak heat release rate and increases the total degree of reaction, indicating that SAP-mediated water release influences the energetics of hydration and effective heat evolution [86]. Recent studies further demonstrate that the thermodynamics and kinetics of SAP water release are governed by polymer physicochemical characteristics such

as crosslinking density, ionic charge density, and pH-responsive functional groups, which control absorption–desorption behavior through osmotic pressure gradients and swelling pressure within the polymer network. Water migration from SAPs into the cementitious matrix occurs via a combination of osmotic pressure-driven flow, ionic crosslinking effects, and capillary suction, with desorption evolving from an early osmotic-controlled stage to a later humidity-dependent process as pore solution chemistry and relative humidity change during hydration [7].

5.3. Impact on Hydration Processes

The nucleation of calcium silicate hydrate (CSH) plays a pivotal role in controlling early hydration kinetics and microstructural development in cementitious systems, as effective nucleation sites lower the activation barrier for CSH precipitation and accelerate hydration. CSH–polycondensate nanocomposites synthesized via co-precipitation from $Ca(NO_3)_2$ and Na_2SiO_3 form nanofoil morphologies with characteristic dimensions below 100 nm, where the small particle size provides a high density of interfacial area, making them highly effective seeding agents that promote early CSH formation and early strength development [87]. Similarly, calcium silicate hydrate nanoparticles modified with polycarboxylate ether (CSH-PCE) provide a high density of nucleation sites that reduce the energy barrier for CSH formation and promote early age hydration, although compatibility effects may modulate fluidity and microstructural outcomes [88].

The chemical characteristics of polymer shells, including functional group composition and surface charge, play a decisive role in governing whether polymer admixtures delay or accelerate cement hydration. When highly carboxylated, colloidal polymers are introduced into cement systems and their negatively charged surfaces rapidly adsorb onto cement grain surfaces via electrostatic interactions, forming a polymer covering layer that creates unfavorable conditions for calcium silicate hydrate (CSH) nucleation and leads to pronounced hydration retardation. The extent of this retardation is governed by the type and density of surface charge, particularly carboxylate groups, rather than the bulk polymer composition. Notably, chemical modification of the same polymer shells through enrichment with Ca^{2+} and silicate species reverses this behavior, as CaSi-decorated colloidal surfaces become effective nucleation sites for CSH formation, acting as seeding agents that accelerate hydration and enhance early strength development [80]. In parallel, superabsorbent polymers with different chemical structures exhibit distinct interactions with cement pore solutions, where variations in functional group chemistry and anionic charge density control water absorption and release behavior, leading to either delayed or sustained hydration kinetics depending on ion complexation and desorption profiles [85].

5.4. Microstructure Evolution

The incorporation of SAPs into cementitious systems exerts significant effects on microstructural evolution, particularly on porosity, microcrack development, and pore connectivity. Experimental analyses reveal that SAP addition alters the pore structure by increasing the volume of pores in specific size ranges while simultaneously promoting a refinement of smaller pore populations as hydration progresses, leading to a more heterogeneous and intricate pore network [89]. SAP incorporation in cementitious systems is, also, associated with pronounced changes in porosity, including smaller average pore diameters and modified connectivity metrics, which relate directly to the redistribution of water and hydration products within the matrix. These microstructural changes can influence the initiation and propagation of microcracks by modifying stress concentrations and transport pathways. These effects are particularly evident in specialized mixtures, such as those enhanced with metakaolin [90].

SAPs play a significant role in reducing autogenous shrinkage in cementitious materials by maintaining internal relative humidity as hydration progresses. Adding them to cement paste, mortar, and concrete mixtures significantly reduce autogenous shrinkage, although the degree of mitigation varies with mixture composition and SAP type, and complete elimination is not always achieved [91]. This shrinkage reduction is linked to the internal curing mechanism, whereby SAPs release stored water in response to internal relative humidity drop and self-desiccation, with water release timing strongly influencing shrinkage evolution and strain development. However, the relationship between dosage and shrinkage reduction is non-linear, and excessive contents may introduce voids that negatively affect compressive strength [92].

6. Performance in Construction Materials

6.1. Mechanical Properties

In core–shell-based composite carriers designed for cementitious systems, the elastic modulus of the shell phase is a key design parameter affecting mechanical responsiveness and stress distribution between the inclusion and the matrix, suggesting that tuning shell stiffness directly influences how stress is carried and how cracks propagate within the composite material [93]. Additionally, core–shell superabsorbent composites have been shown to exhibit enhanced mechanical stability and overall strength when inorganic nanofillers are incorporated into the shell structure, implying that tailored core–shell architecture can improve load transfer and resilience against mechanical stresses. In the work of Pastrafidou et al., the multifunctional role of superabsorbent core–shell composites by linking their structural design to a broad spectrum of applications is illustrated in Figure 4. It highlights how the combination of a swellable core and a functional shell enables controlled fluid uptake, mechanical reinforcement and environmental responsiveness. The figure further emphasizes their versatility across fields such as construction, agriculture and packaging, underlining the importance of structure–property relationships. Overall, it provides a conceptual overview of how engineered core–shell systems can deliver both performance enhancement and application-specific functionality [94] (Figure 4). Studies on polymer composites with core–shell fillers further indicate that well-bonded core–shell particles can increase tensile strength and fracture toughness compared to systems without such fillers, highlighting the importance of interphase adhesion and morphology in determining composite mechanical performance [95]. While investigations in conductive hydrogels emphasize different applications, they similarly demonstrate that nano-reinforcement within a structured polymer network enhances tensile strength and toughness, illustrating the potential for core–shell conductive polymers to combine electrical functionality with mechanically robust behavior [96].

6.2. Durability Enhancements

In cementitious composites incorporating advanced polymeric additives, freeze–thaw resistance and chloride penetration mitigation are markedly improved through tailored material design. Concrete modified with superabsorbent polymers (SAPs) and nano-silica exhibited enhanced frost durability, retaining a higher relative dynamic elastic modulus and showing reduced scaling after hundreds of freeze–thaw cycles, while also improving resistance to ion erosion compared with unmodified SAP concrete [97]. Similarly, polymer impregnation treatments, particularly those involving silicone and other polymers, were shown to significantly lessen strength loss after freeze–thaw cycling and decrease chloride ingress depths, demonstrating superior durability performance under combined environmental stressors [98]. The incorporation of cellulose/polyvinyl alcohol hydrogels within concrete was also reported to optimize the pore structure and mitigate crack propa-

gation caused by freeze–thaw attack, by restricting water activity and releasing absorbed water in a controlled manner to alleviate expansion stress [99]. Furthermore, studies on internal curing with these polymers in reinforced concrete structures indicate that SAP incorporation not only limits early crack formation (which in turn reduces pathways for chloride ingress) but also contributes to prolonged crack-free performance under field conditions, hence enhancing overall durability and potential for self-healing in service. In the work associated with Figure 5, the monitoring approach is schematically illustrated using embedded electrodes that enable continuous assessment of corrosion potential within the concrete matrix. The figure highlights how electrochemical signals can be collected in situ, providing real-time insight into the initiation and progression of reinforcement corrosion. It also demonstrates the integration of sensing functionality directly into the material system, supporting predictive maintenance strategies. Overall, the schematic emphasizes the role of smart monitoring techniques in extending service life and improving durability assessment of reinforced concrete structures [100].

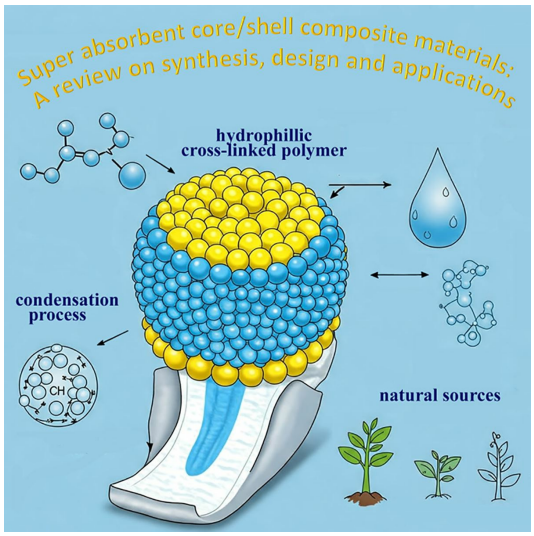


Figure 4. Schematic representation of superabsorbent core–shell composites, depicting the hydrophilic crosslinked polymer network, synthesis via condensation processes, and incorporation of components derived from natural sources. The diagram highlights the core–shell architecture and its role in water absorption, transport pathways, and structure–function relationships relevant to advanced materials applications [94].

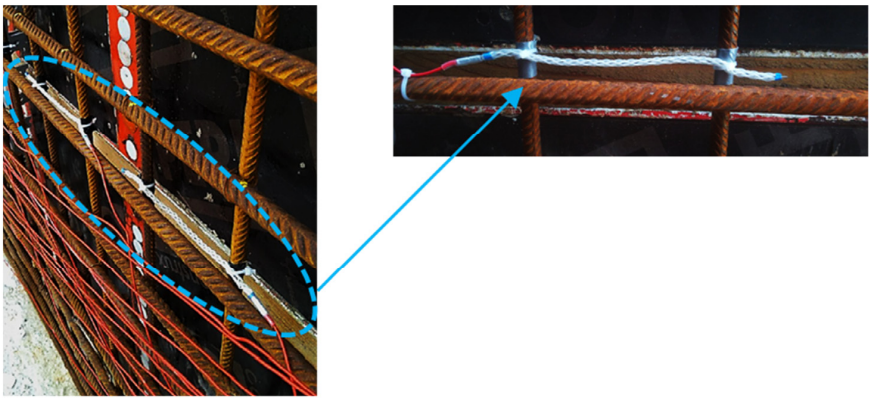


Figure 5. Monitoring setup for measuring corrosion potential using embedded electrodes in reinforced concrete. The highlighted areas and inset detail illustrate the placement of electrodes along the steel reinforcement and the conductive paths that allow electrochemical detection. This configuration demonstrates the practical application of in situ corrosion monitoring in structural materials [100].

6.3. Functional Performance

In cement-based construction materials, superabsorbent polymers (SAPs) serve as internal curing agents that absorb and release water to promote hydration and enhance self-healing of microcracks, although their influence on electrical properties such as piezo resistivity has not yet been fully explored. SAP-modified concrete with optimized SAP content has been shown to improve later-age strength and crack self-healing through promoted hydration product formation, indicating potential for multifunctional performance when combined with other functional fillers. In the work of Cheng et al., the macro- and microstructural characteristics of SAP particles are presented, illustrating their morphology and internal porous network. Figure 6 highlights the capacity of these materials to absorb and retain significant amounts of fluid, which is directly linked to their role in internal curing. It also emphasizes how particle size, shape and surface features influence their interaction with the cement matrix. Overall, visualization provides insight into the structure–function relationship governing their performance in cementitious systems [101]. In piezoresistive construction composites, ultra-high-performance concrete (UHPC) reinforced with conductive recycled carbon fibers exhibits enhanced electrical conductivity and a measurable piezoresistive response under mechanical loading, enabling self-sensing capabilities for detecting stress and strain in structural components [102]. Similarly, self-sensing cement composites formulated with conductive particulate additions demonstrate the ability to self-monitor changes in stress–strain conditions without external sensors, highlighting the critical role of well-connected conductive pathways within the matrix for piezoresistive behavior [103]. Further, the integration of conductive fillers such as multi-walled carbon nanotubes into cementitious materials can lead to distinct changes in electrical resistance in response to cyclic loading and unloading, reflecting sensitive piezoresistive performance relevant to structural health monitoring [104]. Although direct piezoresistive self-sensing behavior of SAP-modified cementitious materials has yet to be demonstrated, studies collectively suggest that coupling them with conductive networks (e.g., carbonaceous or nanofiller additions) could be a promising route to achieving multifunctional construction materials capable of both internal curing and real-time structural health assessment.

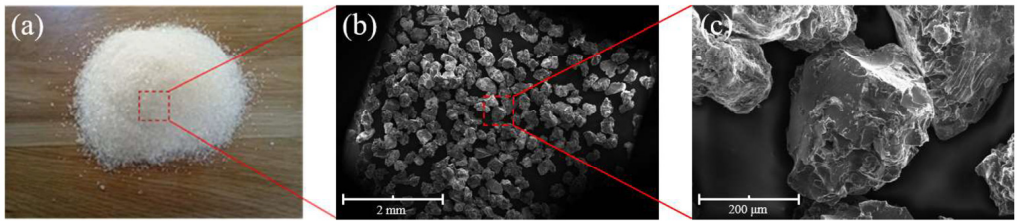


Figure 6. (**a**) Optical image and (**b**,**c**) SEM micrographs of SAPs, showing the macroscopic appearance and detailed microstructural morphology. The images highlight the particle size, surface texture, and internal porous structure relevant to their swelling and transport properties [101].

Recent research has demonstrated that SAPs incorporated into cementitious composites can significantly influence the electromagnetic wave interaction characteristics of these materials, transforming ordinary concrete into electromagnetic wave-absorbing composites with enhanced performance. Concrete mixtures containing them have been shown to achieve high levels of electromagnetic absorption across microwave frequency ranges, for example, SAP-modified 3D-printed concrete samples with up to 40 vol.% SAP exhibited a peak reflection loss of −19.12 dB at 7.53 GHz and an absorption rate of 98.77%, significantly outperforming untreated counterparts, largely due to the SAP-induced porous microstructure that improves impedance matching and multiple internal reflections of electromagnetic waves within the material matrix. In the work of Zhang et al., the morphology of the in-

dividual constituents used in cementitious composites is presented, including ordinary Portland cement (OPC), silica fume (SF), superabsorbent polymers (SAPs), copper slag (CS), quartz sand and polypropylene (PP) fibers. Figure 7 highlights the distinct surface textures and particle geometries that influence dispersion and interfacial interactions within the matrix. It also provides insight into how these microstructural features contribute to the overall performance of the composite system [105]. Additionally, studies of electromagnetic shielding in cement-based materials reinforced with conductive fillers confirm that augmenting the electrical properties of the composite, typically via conductive phases, can enhance shielding effectiveness, indicating that multifunctional construction materials can be designed with both structural performance and electromagnetic attenuation in mind [106].

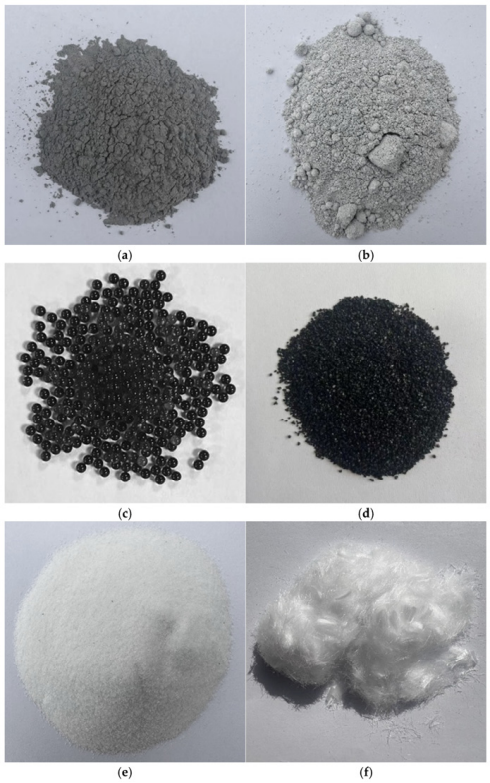


Figure 7. Optical images of the morphologies of (**a**) OPC, (**b**) SF, (**c**) SAP, (**d**) CS, (**e**) quartz sand, and (**f**) PP fiber, illustrating differences in particle shape, size distribution, and surface characteristics. These morphological variations influence the dispersion, interfacial interactions, and overall performance of the composite system [105].

In cement-based construction materials, these polymers can significantly enhance the interaction of mortars and plasters with moisture and environmental humidity due to their ability to absorb and desorb large amounts of water, which in turn substantially improves moisture transport properties such as the water absorption coefficient and water vapor diffusivity. The increased open porosity and enhanced sorption/desorption isotherms observed in SAP-modified cement–lime plasters lead to a notably higher moisture buffering potential, with the mass increase in SAP-enriched composites being several times greater than that of reference materials, demonstrating their passive humidity-responsive behavior. In the work of Fort et al., as illustrated in Figure 8, this process involves the utilization of SAPs to modify interior plasters, resulting in a significant passive reduction in indoor humidity fluctuations by dampening the peak relative humidity levels [107] (Figure 8). Although research on construction materials specifically engineered for electrical humidity sensing remains limited, analogous work in advanced materials shows that conductive

polymer-based hydrogels with core–shell structures (e.g., PEDOT:PSS) exhibit humidity-dependent changes in electrical resistance, where water adsorption into the hydrophilic shell swells the polymer network and increases resistivity under high humidity conditions, pointing to a mechanism by which conductive SAP systems could transduce humidity into measurable electrical signals. Such findings from conductive hydrogel sensors suggest a promising future direction for integrating conductive core–shell SAPs into construction materials to achieve real-time moisture and humidity sensing in structural matrices, combining traditional SAP moisture management with active humidity monitoring [108].

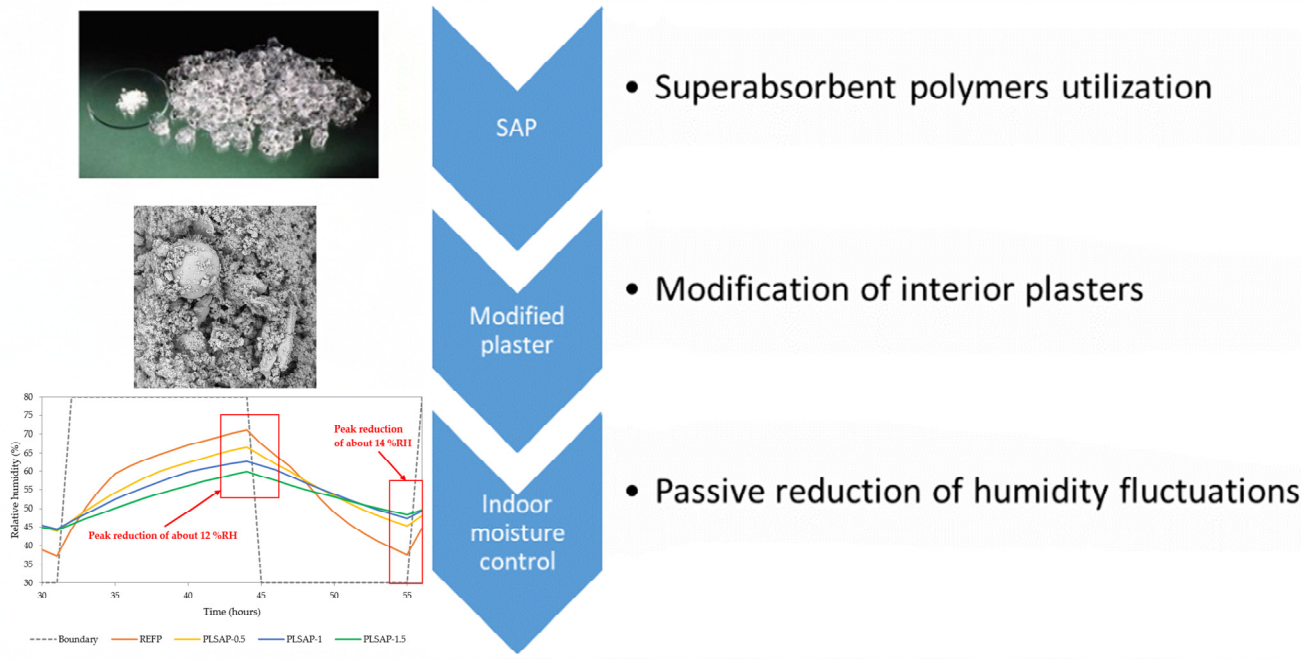


Figure 8. Demonstration of the influence of superabsorbent polymers on moisture regulation in building interiors. The figure illustrates the absorption and release behavior of SAPs under varying humidity conditions, highlighting their role in stabilizing indoor moisture levels and improving durability and comfort in construction materials [107].

6.4. Comparative Assessment of Other Additives

Compared to superabsorbent polymers, which are primarily incorporated in cementitious materials to control internal curing and shrinkage cracking through water absorption and release, carbon-based and metallic nano-/micro-additives enhance electrical, mechanical, and multifunctional properties in different ways. While they improve moisture distribution and reduce autogenous shrinkage without significantly increasing conductivity, carbon-based nano-additives, such as graphene oxide or carbon nanotubes (CNTs) can form percolating conductive networks in cement matrices, enabling enhanced both electrical conductivity and mechanical strength, as well as piezoresistive self-sensing behavior when adequately dispersed [109].

Although graphene nanoplatelets (GNPs) are theoretically expected to improve composite performance, experimental studies have revealed notable limitations. In multifunctional cementitious mortars, GNP incorporation resulted in reduced mechanical strength (up to 16%) and higher electrical resistivity compared to recycled carbon fillers, primarily due to agglomeration and macropore formation associated with GNP hydrophobicity. In contrast, recycled fillers such as GCH and UFS outperformed GNPs, improving compressive strength by up to 25% while providing superior electrical conductivity. These results highlight the critical role of dispersion quality and interfacial interactions, indicating that

without appropriate carriers or functionalization, conventional carbon nanomaterials may adversely affect cement matrix integrity [110].

Comparative studies further report that graphene oxide (GO), pristine or functionalized CNTs, and metallic nanoparticles such as silver influence durability, strength, and resistance to aggressive environments, with filler functionalization and impregnation methods, playing a key role in optimizing their performance [111]. Additionally, conductive carbon fibers and recycled carbon fillers can decrease resistivity and refine microstructure while also improving compressive strength and reducing water absorption, offering a balance between cost, workability, and multifunctionality that differs from the hygroscopic role of SAPs [110]. Based on the comparative assessment above, conductive core–shell SAPs could represent a promising hybrid approach. In this design, the SAP particle might serve as a localized carrier to mitigate the dispersion challenges commonly observed with CNTs and graphene, potentially combining the moisture-management advantages of SAPs with the enhanced electrical performance of carbonaceous or metallic fillers.

7. Challenges and Future Perspectives

7.1. Physicochemical Design Constraints

In cementitious materials, the high pH (often >13) of the pore solution presents a significant physicochemical constraint for the design of functional shell materials, as many polymeric shells and encapsulation systems may undergo chemical reactions with alkali species that compromise durability and structural integrity over time. Studies focusing on polymer-based microcapsules in alkaline cementitious environments indicate that alkali solutions can interact with the capsule shell, potentially leading to changes in the capsule or reduced long-term stability, making the evaluation of microcapsule–matrix interactions essential for durable performance in mortars and concretes. In the work of Kamaraj et al., the thermal cycle of phase-changing materials (PCMs) is illustrated in Figure 9, showing the transition between energy absorption and release during heating and cooling processes. It highlights how temperature variations drive phase transitions, which are associated with latent heat storage and release. The schematic emphasizes the reversible nature of this process and its relevance for thermal regulation within material systems [105].

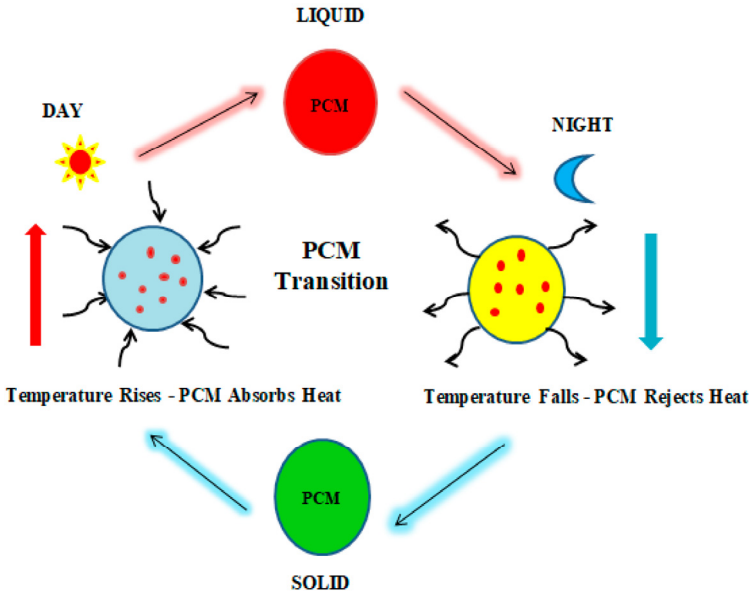


Figure 9. Schematic illustration of the phase-changing materials that undergo a transitional process. The red arrow indicates the rise in the temperature while the blue arrow shows the drop in the temperature [112].

Research on superabsorbent polymer hydrogels exposed to alkaline media similar to cement pore solutions shows that while gels maintain significant water uptake and retention capacity, their behavior is strongly influenced by their chemical environment and ion concentration, indicating that polymer network stability and water retention under alkaline conditions must be carefully considered in design for internal curing and functional performance [113].

Moreover, comprehensive reviews of self-healing microcapsule systems highlight shell instability and premature leaching as key challenges in alkaline cement-based materials, with pH, emulsifier type, and shell composition all affecting the survivability and performance of polymer shells embedded in concrete matrices [114].

For conductive core–shell SAPs envisaged as multifunctional additives in construction materials, shell stability under highly alkaline conditions is critical, as shell degradation may compromise the encapsulation of conductive phases, disrupt electrical pathways, and reduce long-term functional performance (e.g., sensing or self-healing). Future design strategies should therefore prioritize alkali-resistant shell architectures, such as inorganic–organic hybrids or crosslinked polymers with pH-stable covalent bonds, to ensure that conductive networks remain functional throughout the service life of the construction material. In the work of Al-Shawafi et al., macroscopic and microscopic observations of various dry SAP particles highlight significant variability in their size distribution and morphological characteristics. The irregular, angular surface features and the range of particle dimensions directly dictate the swelling kinetics and the surface area available for moisture interaction within the material matrix. Consequently, this morphological analysis explains how the initial polymer geometry influences the overall water absorption and performance of the modified composite [112–114] (Figure 10).

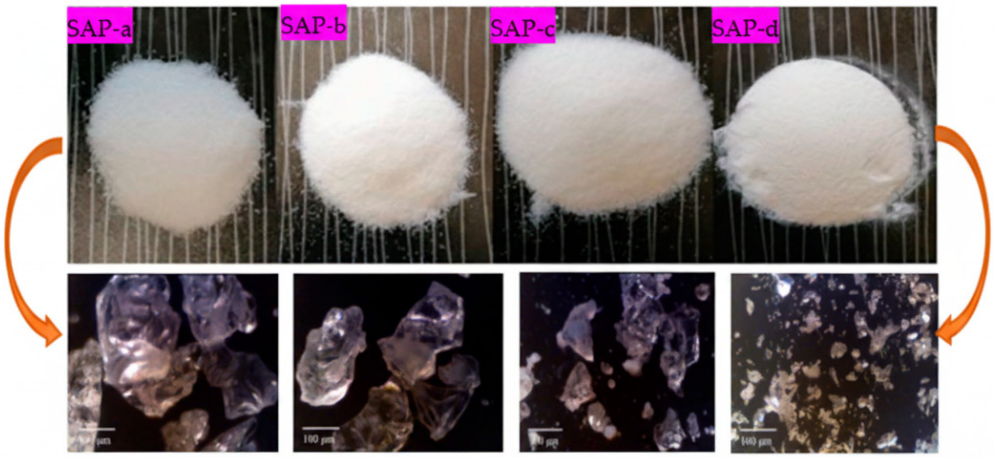


Figure 10. Microscopic observation of the particle size distribution of dry superabsorbent polymers. The image highlights particle morphology, size variability, and surface features, which are critical for understanding swelling behavior and absorption performance [113].

In cementitious systems these polymers fulfill their primary function through extensive swelling, enabling internal curing and mitigation of autogenous shrinkage. Experimental studies demonstrate that SAP absorptivity and swelling behavior are governed by the polymer network structure, ionic environment, and hydration conditions of the cement matrix, underscoring the sensitivity of swelling performance to physicochemical parameters [115]. In contrast, studies on conductive hydrogel composites and polymer systems incorporating conductive phases show that electrical conductivity strongly depends on the continuity and percolation of conductive domains within the polymer network, which can be affected by network expansion and crosslinking density [116]. Excessive swelling has been reported to relax polymer networks and disturb conductive pathways, leading to reduced electrical

performance and mechanical stability in conductive hydrogel systems [117]. For conductive core–shell SAP architectures in construction materials, this trade-off is particularly relevant: designs that maximize water uptake to enhance internal curing or humidity response may inadvertently disrupt or dilute conductive networks if the shell or embedded conductive phase is not carefully engineered. Balancing swelling capacity with stable, percolating conductivity therefore represents a key physicochemical design constraint for achieving multifunctional SAPs that remain electrically and mechanically compatible with cementitious matrices. As illustrated in the work of Elyashevich et al., the swelling behavior of polyacrylic acid (PAA)-based hydrogel composites (PAA/PANI and PAA/PPy) are highly pH-dependent, with the pure PAA matrix exhibiting significantly higher expansion compared to the conductive composites. This differentiation highlights how the incorporation of conductive polymers (PANI, PPy) restricts the swelling capacity of the hydrogel, a trade-off that is critical for maintaining the structural integrity and connectivity of the conductive network under varying environmental conditions [115–117] (Figure 11).

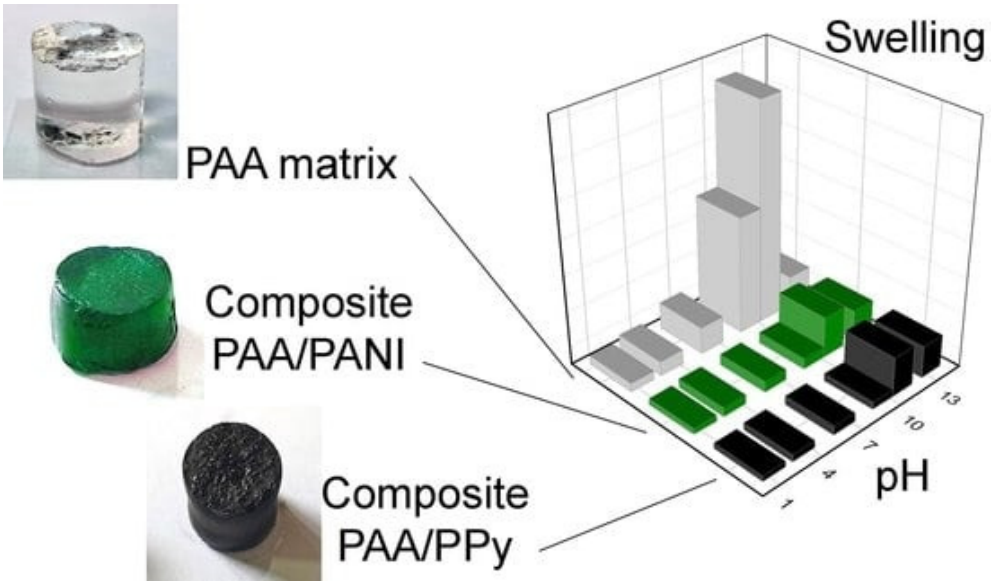


Figure 11. Schematic representation of composite properties based on polyacrylic acid hydrogels combined with conducting polymers. The illustration highlights the reversible swelling–deswelling behavior and pH-responsive characteristics, emphasizing their impact on transport, conductivity, and adaptive performance in functional materials [116].

7.2. Modeling and Simulation Opportunities

In soft polymer systems like hydrogels and polyelectrolytes, molecular dynamics (MD) simulations help visualize and predict molecular changes during network formation, crosslinking, and swelling. They reveal how intermolecular interactions and microstructure evolve, affecting properties. MD shows detailed structural and dynamic features of crosslinked hydrogel networks, linking interchain interactions to swelling and mechanics, while highlighting challenges in creating accurate force fields for realistic environments [25].

First principles methods such as density functional theory (DFT) and quantum mechanical/molecular mechanical (QM/MM) hybrid models complement MD by enabling atomistic insights into charge distribution and electronic structure, thereby facilitating the theoretical analysis of electronic properties, charge transport kinetics, and conductive behavior in conjugated polymers or ion containing systems. DFT calculations have been widely applied to elucidate ground state and excited state electronic characteristics, band structure, and charge carrier dynamics in polymeric materials, providing fundamental understanding relevant to conductivity and transport phenomena [118].

At larger length and time scales, mesoscale models can bridge atomistic detail to mesoscopic morphology and transport behaviors, providing predictive insight into how pore structure, network organization, and ionic confinement influence macroscopic transport and swelling properties in polymeric systems [25]. For example, multiscale simulation frameworks have been used to derive effective diffusivities of water and ions in gel like materials by coupling MD data at the nanoscale with mesoscale random walk models, illustrating how confinement and structure impact transport phenomena critical for swelling and ionic conductivity [119].

Although these mesoscale modeling approaches have primarily been applied to polymeric and hydrogel systems, similar strategies could be extended to conductive SAPs embedded in cementitious matrices, enabling prediction of water diffusion, ionic transport, and swelling behavior relevant to internal curing and multifunctional performance. The computational modeling approaches discussed here not only provide insight into fundamental mechanisms but also offer predictive tools to guide the design of conductive core–shell SAPs with optimized swelling, stability, and charge transport properties, bridging atomistic understanding with mesoscale material performance [25,118,119].

In a recent construction-oriented study, a deep learning-based digital twin was developed to predict concrete compressive strength with high fidelity compared to baseline models, demonstrating that digital twins can capture complex non-linear relationships inherent to concrete material behavior and guide material design decisions [120]. Additionally, reviews of digital twin applications in emerging construction technologies such as 3D concrete printing highlight how digital twin models can be used throughout the life cycle of concrete elements to optimize process parameters, quality, and sustainability outcomes [121]. For SAP-enhanced concrete, predictive digital twin systems could incorporate sensor feedback, hydration models, and machine learning predictions to forecast the effects of SAP characteristics on long-term mechanical, hygrothermal, and self-sensing behaviors, with conductive core–shell SAPs acting as intrinsic sensors that provide real-time piezoresistive data to 'feed' the digital twin, effectively creating a closed-loop system where the material itself reports its structural health status to its virtual counterpart, thereby enhancing the accuracy and responsiveness oflife cyclee performance predictions. As shown in Figure 12, such smart sensors embedded within a structure can monitor electrical resistance changes ($R < R_0$) triggered by structural defects or crack propagation. This real-time piezoresistive feedback allows for the immediate detection of damage, enabling an automated alarm system that informs the digital twin and ensures the continuous monitoring of structural integrity [116,120–122].

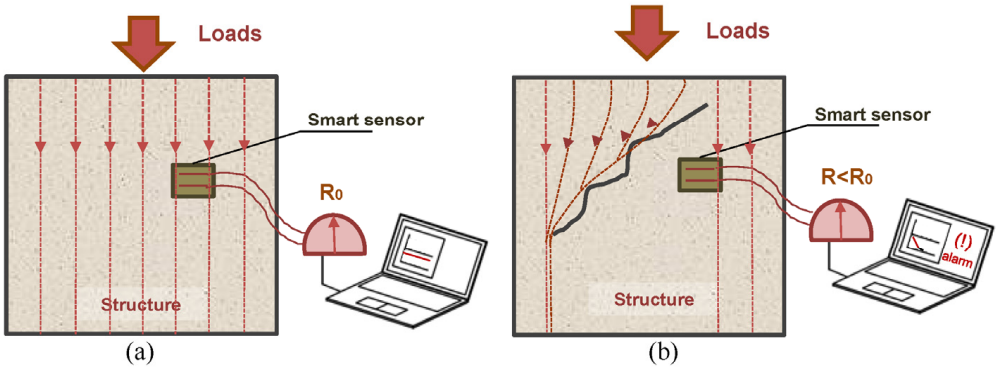


Figure 12. Schematic concept of a single embedded smart material sensors, with (**a**) undamaged structural element monitored with an embedded smart sensor and (**b**) the behavior of the structural element and of the smart sensor after a variation in loads [123].

7.3. Scaling Up: Synthesis, Cost, and Environmental Footprint

Life cycle considerations play a pivotal role in evaluating the sustainability of SAP-modified cementitious materials as they are scaled up for real-world applications. Environmental and economic assessments of reinforced concrete with incorporated SAPs demonstrate that, while the incorporation can enhance autogenous crack mitigation and internal curing, a holistic life cycle assessment (LCA) combined with life cycle costing (LCC) is necessary to balance initial production impacts against long-term durability benefits and maintenance reduction, revealing potential reductions in environmental burdens in extended service scenarios [124].

Comprehensive LCA studies of self-healing concrete systems that integrate microcapsules and impregnated lightweight aggregates show that although such multifunctional additives can lead to higher embodied carbon footprints during production (≈30–50% more than conventional concrete), they may yield substantial whole-life environmental advantages through reduced structural repair and material usage over the service life of the structure [125].

Cradle-to-gate life cycle assessments of SAP-modified cementitious composites further reveal that the environmental impact of the incorporation is highly dependent on the specific polymer chemistry and production route. The use of generic LCA datasets for SAPs can lead to significant under- or over-estimations of environmental burdens, as semi-synthetic and fully synthetic SAPs exhibit markedly different impact levels relative to cement. These findings underline the necessity of polymer-specific inventory data and tailored impact modeling when assessing the sustainability of SAP-based construction materials [126].

A more rigorous perspective on LCA is illustrated by recent carbon footprint analyses of modified cementitious systems, where the environmental impact is evaluated in relation to specific formulation changes. In such studies, the incorporation of chemical modifiers alters both material performance and total carbon emissions, highlighting that the environmental outcome depends on the balance between improved functionality and the impact of added components. These results demonstrate that LCA must be system-specific and cannot be generalized across different material modifications. By extension, the introduction of conductive polymers or nanoparticles in SAP systems should be assessed on a case-by-case basis, considering both performance gains and embodied carbon [127].

In systems incorporating conductive carbon-based fillers, life cycle assessment studies indicate that the inclusion of electrically functional phases can increase the overall environmental footprint of cementitious materials, primarily due to energy-intensive production of carbon nanomaterials, with analyses showing higher embodied impacts compared to reference materials [128]. These findings highlight the critical trade-off between enhanced functional performance and sustainability, underscoring the need for whole-life evaluations where benefits in durability, sensing capability, and extended service life are weighed against initial environmental burdens in the design of conductive SAP-based concretes.

7.4. Emerging Applications

Emerging applications of conductive and multifunctional cementitious materials increasingly target smart infrastructure systems capable of intrinsic monitoring and damage diagnosis. Nanoengineered cement composites incorporating conductive fillers such as carbon nanotubes have been demonstrated as embedded sensing materials for corrosion monitoring, where changes in electrical impedance or resistivity enable detection of reinforcement corrosion initiation and progression under aggressive exposure conditions [129]. Broader state-of-the-art reviews of smart conductive cement-based composites further indicate that dispersed conductive networks can impart self-sensing functionality to concrete,

enabling the monitoring of strain, cracking, and damage while maintaining compatibility with structural materials and avoiding the durability limitations of conventional external sensors [130]. Experimental studies on self-sensing concrete sensors with varying carbon-based fillers show that electrical signal variations can be reliably correlated with mechanical loading and deformation, supporting the feasibility of large-scale structural health monitoring applications in real infrastructure systems [123].

Importantly, comprehensive analyses show that self-sensing cement composites can automatically detect structural deformations and damage through intrinsic electrical responses without external instrumentation, which aligns with the concept of autonomous monitoring materials embedded in concrete matrices, while also highlighting challenges related to filler selection, dispersion control, and the need for standardized testing protocols prior to widespread implementation [131]. In this context, conductive core–shell SAPs represent a promising emerging platform, as they could combine internal curing and moisture responsiveness with intrinsic electrical sensing, enabling multifunctional concrete materials that autonomously monitor structural condition while simultaneously contributing to durability and long-term infrastructure resilience.

8. Conclusions

Summarize key physical chemistry insights, highlight the impact of core–shell conductive SAPs on cementitious systems and emphasize future research directions.

This review has examined conductive core–shell superabsorbent polymers (SAPs) through the lens of physical chemistry with particular emphasis on swelling thermodynamics, charge transport mechanisms and interfacial phenomena relevant to cementitious systems. At the molecular and mesoscale levels, swelling in SAP-based networks is governed by a delicate balance between osmotic driving forces, elastic constraints imposed by crosslinking and ionic effects arising from fixed charges and counterion interactions. In core–shell architectures, this balance becomes spatially heterogeneous, with the shell playing a decisive role in controlling water uptake kinetics, diffusion pathways and the extent of constrained swelling, while simultaneously hosting conductive phases that enable electronic or ionic transport.

A key insight emerging from the reviewed literature is the strong cooperation between hydration-driven swelling and conductivity. Swelling-induced changes in pore size, polymer chain mobility and water content directly modulate ionic mobility and in conductive shells influence the continuity and stability of electronic percolation networks. This dynamic coupling is further complicated in cementitious environments, where high alkalinity and multivalent ions (notably Ca^{2+} and Al^{3+}) alter polymer ionization, suppress swelling through charge screening and counterion condensation and affect both water release behavior and conductive performance. These physicochemical interactions highlight why the cement pore solution cannot be treated as a passive environment, but rather as an active chemical field that reshapes SAP functionality.

Within cementitious systems, conductive core–shell SAPs represent a significant evolution beyond conventional, non-conductive ones. Their ability to simultaneously provide internal curing, mitigate autogenous shrinkage and influence microstructure development is complemented by the introduction of electrical functionality, enabling pathways toward self-sensing, humidity-responsive and electromagnetically active concretes. The core–shell design offers clear advantages by decoupling swelling capacity from conductive performance: the hydrogel core can be optimized for water management, while the conductive shell maintains charge transport even under volumetric changes, wet–dry cycling, and mechanical deformation. This multifunctional behavior positions conductive core–shell SAPs as promising building blocks for next-generation smart cementitious materials.

Despite this progress, several critical knowledge gaps remain. Long-term stability of conductive networks under repeated hydration-dehydration cycles, chemical aging in highly alkaline pore solutions and mechanical loading is still poorly understood. The interplay between SAP-induced porosity, mechanical strength and percolation-based conductivity requires more systematic, multiscale investigation. Moreover, most current studies treat swelling, conductivity and mechanical performance separately; future research must adopt integrated experimental and modeling approaches that capture their coupled evolution in realistic cementitious environments. Advanced characterization techniques, such as in situ electrical measurements during hydration and deformation, combined with molecular simulations of polymer–ion–cement interfaces, will be essential to bridge this gap.

In summary, conductive core–shell SAPs offer a physically grounded and chemically versatile platform for unifying moisture regulation and electrical functionality in cement-based materials. By leveraging insights from polymer physical chemistry and interfacial science, future research can move from proof-of-concept demonstrations toward robust design principles that enable durable, multifunctional and truly intelligent construction materials.

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Abbreviations

AA	Acrylic Acid
AAm	Acrylamide
CNT(s)	Carbon Nanotube(s)
CS	Copper Slag
C–S–H	Calcium Silicate Hydrate
DFT	Density Functional Theory
DSC	Differential Scanning Calorimetry
EIS	Electrochemical Impedance Spectroscopy
EMI	Electromagnetic Interference
FTIR	Fourier Transform Infrared Spectroscopy
GNP(s)	Graphene Nanoplatelets
GO	Graphene Oxide
hc	Critical hydration value
IL	Ionic Liquid
LbL	Layer-by-Layer
LCA	Life Cycle Assessment

LCC	Life Cycle Costing
MD	Molecular Dynamics
OPC	Ordinary Portland Cement
PAA	Polyacrylic Acid
PANI	Polyaniline
PCMs	Phase-Changing Materials
PEDOT	Poly(3,4-ethylenedioxythiophene)
PP	Polypropylene
PPy	Polypyrrole
QM/MM	Quantum Mechanics/Molecular Mechanics
SAP/SAPs	Superabsorbent Polymer(s)
SEM	Scanning Electron Microscopy
SF	Silica Fume
TEM	Transmission Electron Microscopy
UHPC	Ultra-High-Performance Concrete
XPS	X-ray Photoelectron Spectroscopy

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