

Recent Advances in Hybrid Polymer–Cement Composites: Interfacial Engineering, Self-Curing Mechanisms, and Microstructural Performance

Rinu Isah R.J.^{1,*}, K. Vijay Bhaskar Raju², R. Venkata Krishnaiah³

Abstract

The increasing demand for high-performance polymer-modified cementitious composites has stimulated extensive research into hybrid self-curing composite systems with engineered aggregate phases. Self-curing hybrid polymer–cement composite systems, using recycled aggregates, constitute one of the solutions proposed by researchers to overcome problems related to moisture loss during the curing process, shrinkage, cracking, and the limited amount of natural aggregate available. This critical review provides an overview of recent developments concerning the production, processing, characterization, and properties of Advanced self-curing hybrid polymer–cement composite systems made from recycled aggregates. It focuses on the mutual interactions between the polymers used as additives to the system, cement hydration products, internal curing agents, and the nature of the recycled aggregate that can affect the properties of the composite. In this study, several aspects concerning the production of the self-cured hybrid cementitious systems with regard to processing technologies, mixing and curing methods, surface treatments of recycled aggregates, and the use of other additives are critically evaluated. Some of the most important results point to the significance of proper hybridization between polymers as binders and self-cured materials that result in improvements of the interface transition zone quality, reduction of water requirements, reduction of autogenous shrinkage, and improved longevity. There is still much work to be done on the variability in recycled aggregate characteristics, interactions with polymers, practicality of use in the field, and the lack of design standards. Some future areas of investigation have been identified for this technology.

Keywords: Hybrid polymer–cement composites; self-curing mechanisms; polymer film formation; interfacial transition zone; microstructural engineering; composite performance; recycled aggregate reinforcement

INTRODUCTION

Recent advances in polymer-modified cementitious composites have enabled the development of

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multifunctional hybrid systems exhibiting enhanced interfacial bonding, microstructural refinement, and superior mechanical performance. Self-curing hybrid polymer–cement composites have emerged as an important class of advanced composite materials due to their ability to integrate internal moisture regulation with polymer-mediated structural enhancement [1].

Recycled aggregates derived from construction and demolition waste have received great recognition as Advanced substitutes for natural aggregates. Their application ensures Aggregate functionalization and enables Composite

engineering processes. Unfortunately, recycled aggregates possess high porosity, mortar residues, and increased water absorption properties.

Hybrid polymers–cement composites are highly useful materials to increase the efficiency of cement-based construction materials. Polymer addition makes the material denser, lowers its permeability, increases adhesion capacity, and makes it resistant to cracking. All these properties become even more important when using recycled aggregates in order to strengthen the interfacial transition zone and counteract some weaknesses caused by recycled aggregates.

This review critically examines recent advances in hybrid polymer–cement composites containing recycled aggregate phases, with emphasis on interfacial interactions, microstructural evolution, phase compatibility, mechanical response, and emerging composite encountered, and future perspectives [2]. The connection between goals of Advanced construction, recycled aggregates use, polymer modification, and self-curing of the material is shown in Figure 1 below.

FUNDAMENTALS OF HYBRID POLYMER–CEMENT COMPOSITE SYSTEMS

Hybrid polymer–cement composites represent an advanced class of multifunctional composite systems formed through coupled interactions between hydration products and polymer phases that govern microstructural development and performance. In such materials, the stiffness and compressive strength of cement-based materials are combined with the flexibility and adhesion ability of polymers, resulting in efficient composites [3].

The structure–property relationship of hybrid polymer–cement composites is governed by physicochemical interactions between hydration products and dispersed polymer phases. Specifically, during cement hydration, C–S–H gel and Ca (OH)₂ compounds are generated, providing enhanced strength of the materials. Additionally, the polymer particles start coalescing and creating continuous films in the pores [16]. This interaction improves the microstructure and increases the efficiency of the bond. The process of microstructural evolution, including cement hydration, coalescence of polymer particles, and the refinement of the interfacial transition zone, is illustrated in Figure 2.

The polymers that are often utilized in cementitious composite materials comprise styrene-butadiene rubber (SBR), polyvinyl acetate (PVA), acrylic, epoxy resin, and dispersible polymer powder. The polymers make the mixture more malleable, decrease permeability, increase tensile and flexural strength, as well as prevent shrinkage cracks. The impact of polymers varies based on their type, dose, dispersion properties, and compatibility with the cement paste matrix.

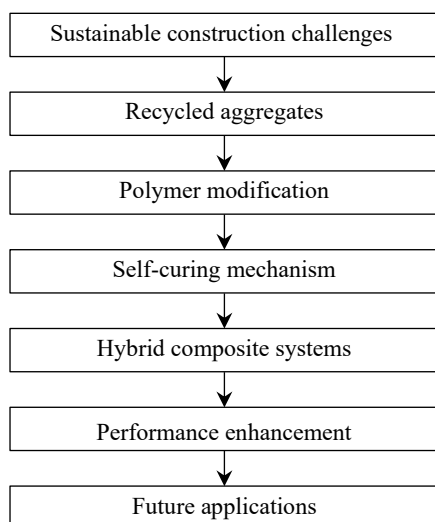


Figure 1. Conceptual framework of Advanced self-curing hybrid polymer–cement composite systems with recycled aggregates.

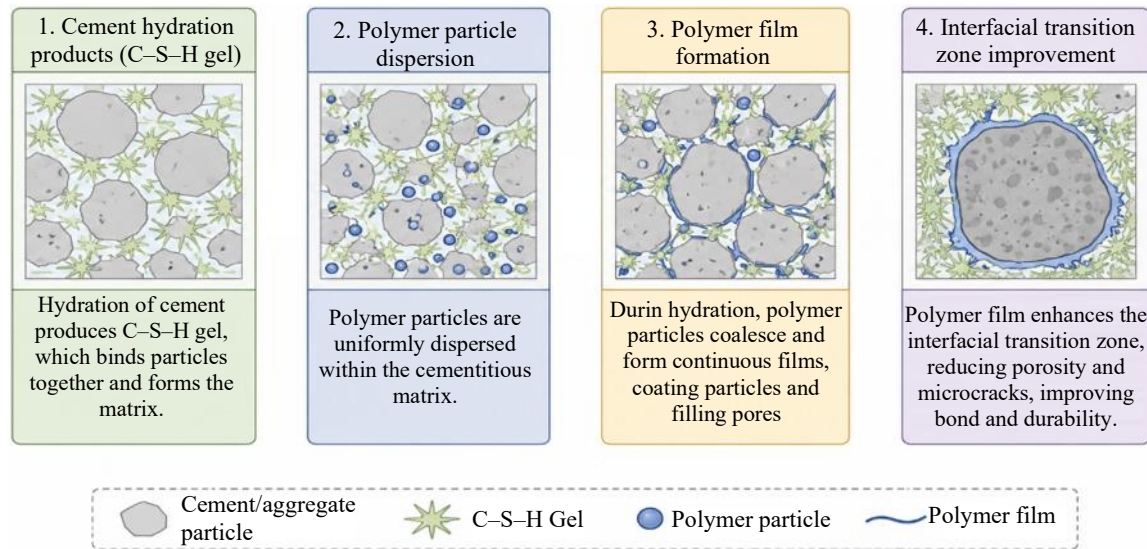


Figure 2. Schematic representation of polymer–cement matrix formation and interfacial interactions.

Table 1. Common polymers used in hybrid cement composites.

Polymer type	Main function	Advantages	Limitation
SBR	Flexibility improvement	Crack resistance	Cost
PVA	Bond enhancement	Durability	Water sensitivity
Acrylic	Surface densification	Chemical resistance	Expensive
Epoxy	High strength	Excellent adhesion	Brittleness

One of the essential characteristics of hybrid systems is the enhancement of the interfacial transition zone (ITZ), which is generally the weakest part of conventional concrete. In addition, the use of polymers leads to a reduction in micro voids, stronger bond between aggregates and cement paste, and a denser microstructure. This is especially important when dealing with the recycled aggregates system [4].

The durability is also increased because of the microstructure improvements offered by the polymers. With the reduction in pore interconnection, there is less moisture intrusion and less ability of aggressive ions, like chlorides and sulfates, to be absorbed. As a result, hybrid polymer-cement composites show greater resilience to freeze-thaw cycles, chemical exposure, and degradation [5].

Another advantage of using self-healing systems lies in the assurance that there is sufficient internal moisture for hydration purposes [6]. This synergy between polymer uses and self-curing technology results in composites having greater hydration efficiency, decreased autogenous shrinkage, and better dimensional stability.

In summary, hybrid polymer-cement composite materials offer an excellent basis for Advanced construction applications if used in combination with recycled aggregate and self-healing technology. The commonly utilized polymers in hybrid cementitious materials and their functions, advantages, and disadvantages are listed in Table 1.

SELF-CURING MECHANISMS IN HYBRID COMPOSITE SYSTEMS

Concept of Self-curing

Self-curing represents an intrinsic moisture-regulation mechanism that controls hydration kinetics and microstructural evolution within polymer–cement composite matrices without external curing intervention. This method works best in concrete mixes with low water to cement ratios since dehydration is common in such materials, resulting in self-desiccation and autogenous shrinkage.

Mechanism of Internal Moisture Supply

Self-curing mechanism takes place via gradual release of stored water due to hydration reaction in the cement. As the hydration reaction continues and the relative humidity level reduces, water from within the self-curing material releases itself and forms C-S-H gel. The formation of calcium silicate hydrate helps in densifying the matrix while reducing the pores. The effect of the mechanism is that of stress reduction and dimensional stability in the material [7]. The internal moisture transport and sustained hydration mechanism in self-curing material is shown in Figure 3 below. Table 2 shows a comparative analysis of various self-curing materials [17].

Self-curing Materials and Techniques

Several materials are commonly used to achieve self-curing in hybrid composite systems. Superabsorbent polymers (SAPs) can absorb large amounts of water and release it gradually during hydration. Lightweight porous aggregates serve as internal reservoirs due to their high absorption capacity. Chemical self-curing admixtures such as polyethylene glycol reduce evaporation and improve moisture retention. The selection of self-curing material depends on mix design requirements and targeted performance characteristics [8].

Synergistic Interaction with Polymer–Cement Systems

In polymer-cement composite materials, the self-compaction process works synergistically with polymer modification. In-plate curing ensures that there is adequate moisture for hydration, and the film-forming property of polymers leads to enhanced pore refinement and prevents water loss.

Contribution of Recycled Aggregates

Recycled aggregates can make a major contribution towards self-compaction because of their porous nature and their ability to absorb water. Pre-saturated, they will behave as internal sources of water that will slowly release the moisture during the hydration process. This not only increases the efficiency of hydration but also helps counter the poor interfacial characteristics of recycled aggregates [18].

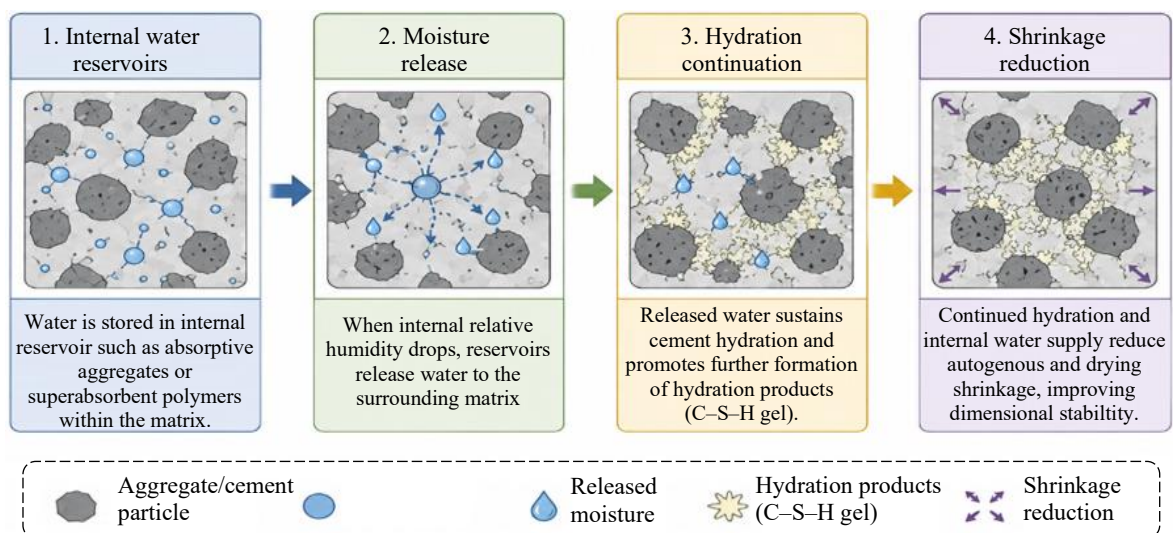


Figure 3. Internal moisture release and hydration continuation mechanism in self-curing composites.

Table 2. Self-curing agents and their functions.

Self-curing agent	Mechanism	Benefits
SAP	Water absorption/release	Shrinkage reduction
Lightweight aggregate	Internal curing reservoir	Hydration enhancement
Recycled aggregate	Water retention	Performance optimization
PEG	Moisture retention	Improved hydration

Performance Enhancement

Incorporating self-curing mechanisms leads to the refinement of microstructure, minimizes shrinkage, provides improved quality of interfacial transition zone, and contributes towards long-term mechanical strength and durability. Self-curing is therefore vital in creating Advanced high-performance hybrid composite materials [9].

FUNCTIONAL CONTRIBUTION OF RECYCLED AGGREGATE PHASES IN HYBRID POLYMER COMPOSITES.

Importance of Recycled Aggregates

Recycled aggregate phases have emerged as engineered particulate reinforcements that significantly influence the morphology and interfacial characteristics of hybrid polymer–cement composites. The rising quantity of Recycled particulate phase alongside the dwindling supply of natural aggregates has led to a dire necessity for the adoption of effective recycling methods within the construction industry. Recycling helps achieve Composite engineering objectives such as reduced landfill space, reduced reliance on natural resource extraction, reduced transportation impacts, and Performance optimization. The application of recycled aggregates in hybrid composites is thus a significant stride towards High-performance composite applications materials.

Physical and Microstructural Characteristics

Recycled aggregates usually have old mortar bound to their surface, which greatly affects the physical properties of the aggregates. These old mortar remnants increase the pore size, roughness, water absorption properties, and reduce the particle density. These factors lead to higher demands for moisture and variations in the material properties. Porous microstructure of the recycled aggregates also impacts the interfacial transition zone, which is considered the weakest part of any composite [19]. These factors are mostly dependent on the quality of recycling, the process of crushing, and contamination in the recycled materials. Comparisons of the physical and microstructural properties of recycled and natural aggregates can be found in Figure 4. The effects of different levels of recycled aggregate substitution on composite behavior are listed in Table 3.

Property	Recycled aggregates (RA)	Natural aggregates (NA)	Comparison
Water absorption 	 Higher (4.0–8.0%) 	 Lower (0.5–2.0%) 	RA>>NA
Porosity 	 Higher (12–25%) 	 Lower (1–5%) 	RA>>NA
Density 	 Lower (2.1–2.4g/cm ³) 	 Higher (2.5–2.7g/cm ³) 	RA<NA
Surface 	 Higher (Rougher) 	 Lower (Smoother surface) 	RA>NA
Attached mortar 	 Higher (10–30%) 	 Negligible (≈ 0%) 	RA>>NA

Figure 4. Properties of recycled versus natural aggregates.

Table 3. Influence of recycled aggregate replacement level.

Replacement (%)	Workability	Strength	Durability
25%	Slight decrease	Stable	Improved
50%	Moderate reduction	Slight drop	Acceptable
75%	Significant reduction	Noticeable loss	Reduced
100%	High reduction	Lower strength	Needs modification

Influence on Composite Performance

The incorporation of recycled aggregate phases directly modifies rheological behavior, interfacial microstructure, and load-transfer efficiency within hybrid composite architectures. In the case of fresh concrete, the presence of increased water absorption capacity can have adverse impacts on concrete's workability as well as change its rheological properties if proper management of the water is not done. In hardened concretes, a poor interfacial transition zone causes decreased compressive strength, high shrinkage levels, and low durability in comparison to aggregates of natural origin [10].

Interaction with Polymer Modification and Self-curing

Modification of polymers is vital for increasing the performance of hybrid composites made using recycled aggregates. Polymer film formation in the matrix increases the bond strength between the aggregate surface and the cement paste, as well as filling micro voids and increasing interfacial bonding. Recycled aggregates may be useful in helping to promote self-compaction of the material because of its great ability to absorb water. Pre-saturated recycled aggregates function as an internal water source that releases stored water during hydration processes.

Benefits and Current Challenges

The use of recycled aggregates in conjunction with polymer and self-curing concrete technologies presents a number of strengths, such as dimensional stability, increased durability, low environmental footprint, and resource efficiency. However, there are still some problems that need to be considered, such as quality variation in recycled aggregates, standardization, and uncertainty in terms of the results achieved in practice.

On the whole, recycled aggregates serve as a key constituent in hybrid composites due to their ability to ensure Performance optimization, alongside with providing some operational benefits in combination with other techniques.

POLYMER MODIFICATION IN CEMENTITIOUS HYBRID COMPOSITES

Concept of Polymer Modification

Polymer modification governs phase evolution and interfacial network formation within cementitious composite matrices through controlled polymer dispersion and film development. In hybrid composite materials, the role of polymers is critical since they provide interlinking in order to form an interpenetrating network that provides better bonding between materials. Hybrid composite materials with the use of recycled aggregate possess various shortcomings; hence, polymer modification in such materials acts as an effective approach for overcoming these weaknesses [20].

Types of Polymers Used

Different kinds of polymers can be incorporated into cement-based composites depending upon the application and properties required. The commonly used polymers in such composites include styrene butadiene rubber (SBR), polyvinyl acetate (PVA), acrylic polymers, epoxy resins, and latex. Different types of polymers provide different types of advantages for the composites. For instance, the styrene butadiene rubber enhances adhesion and flexibility, while acrylics add waterproofing capacity and improve durability. Similarly, epoxy resins improve bonding strength as well as chemical resistance in the system.

Mechanism of Performance Enhancement

The primary contribution of polymer modification arises from polymer phase coalescence and continuous network formation within the hydrated matrix. This occurs through the combination of polymer particles during hydration and hardening of the cement. The result is the formation of films that cover micro-cracks, fill up inner porosities, and enhance connectivity among the cement hydration compounds. This increases the strength of the interfacial transition zone between the aggregate particles and the cement paste. In a system where the interface is compromised by mortars present on the surface, film development helps improve bonding performance.

Influence on Mechanical Properties

Polymer modification results in significant improvement of many mechanical properties of the hybrid composites. Tensile and flexural strength increases because of enhanced ductility and ability to resist cracks. Better bonding within the matrix leads to more efficient transfer of stresses, thus preventing cracks from forming under applied loads. Compressive strength is not necessarily increased proportionately to other mechanical properties; however, the toughness and ability to absorb energy of the composite are greatly increased [11].

Influence on Durability Performance

Enhancement of durability is one of the biggest advantages resulting from the use of polymer-modified concretes. As already mentioned before, polymer chains help limit water, chloride, and other aggressive substances from penetrating into concrete by plugging up capillary pores. Therefore, the modified concrete becomes more resistant to the freeze-thaw phenomenon and sulfate aggression, as well as to steel corrosion. When dealing with hybrid mixtures that involve recycled aggregates, this advantage comes out even clearer [12].

Challenges and Limitations

Despite its many benefits, however, polymer modification does pose some difficulties. First, it is costly relative to the other components in traditional cement mixes. In addition, inappropriate quantities of polymer and/or incompatibility with particular cements may cause undesirable effects on hydration and setting properties. Finally, more research needs to be done regarding long-term performance under various conditions.

In general, polymer modification is one of the primary methods used to improve the structural performance of hybrid composites that utilize recycled aggregate and self-curing technology [13]. This is illustrated in the comparative Figure 5, which show how different types of hybrid polymer-modification and self-curing strategies can result in multiple benefits. Experimental results from the most recent scientific literature are summarized in Table 4.

SYNERGISTIC INTERACTIONS IN HYBRID POLYMER–CEMENT COMPOSITE ARCHITECTURES

The performance of hybrid polymer–cement composites is governed by coupled interactions among polymer film formation, internal moisture regulation, and particulate phase morphology. Recycled aggregate phases introduce controlled heterogeneity and internal moisture reservoirs, while polymer modification promotes interfacial adhesion and pore refinement [14]. The synergy between these mechanisms improves matrix continuity, enhances stress-transfer pathways, minimizes microcrack propagation, and produces superior composite mechanical performance [20].

Table 4. Summary of reported experimental findings.

Study	Polymer	Self-curing Method	Recycled Aggregate	Key Outcome
Zhang et al.	Acrylic	SAP	50% RCA	Durability improved
Al-Mansour et al.	Polymer-nano silica	Internal curing	40% RCA	Waterproofing improved
Mohammed et al.	SBR	Moisture retention	Rubberized RCA	Crack resistance improved

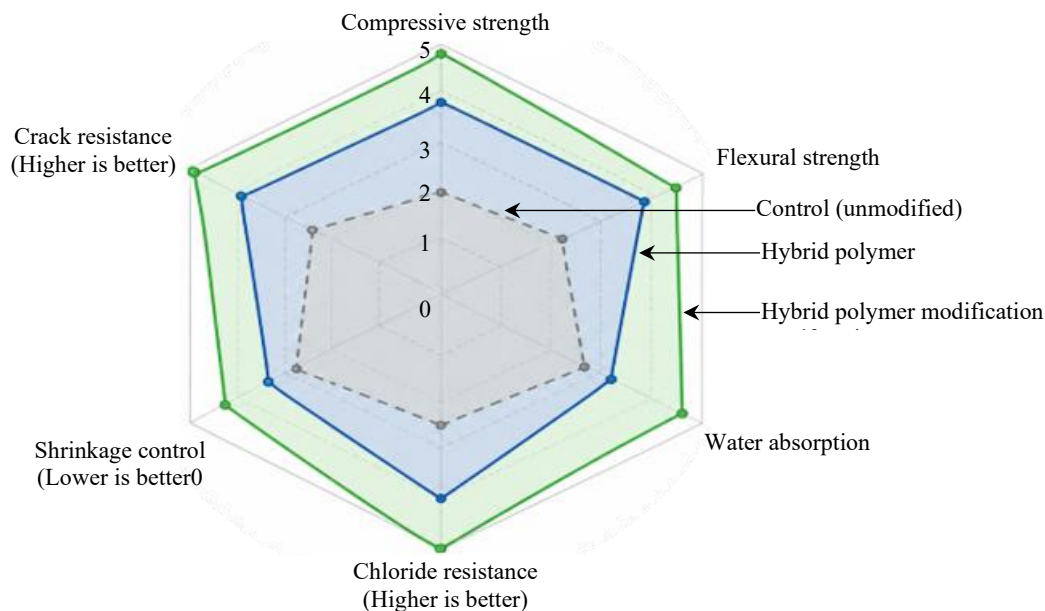


Figure 5. Performance improvements achieved through hybrid polymer modification and self-curing strategies.

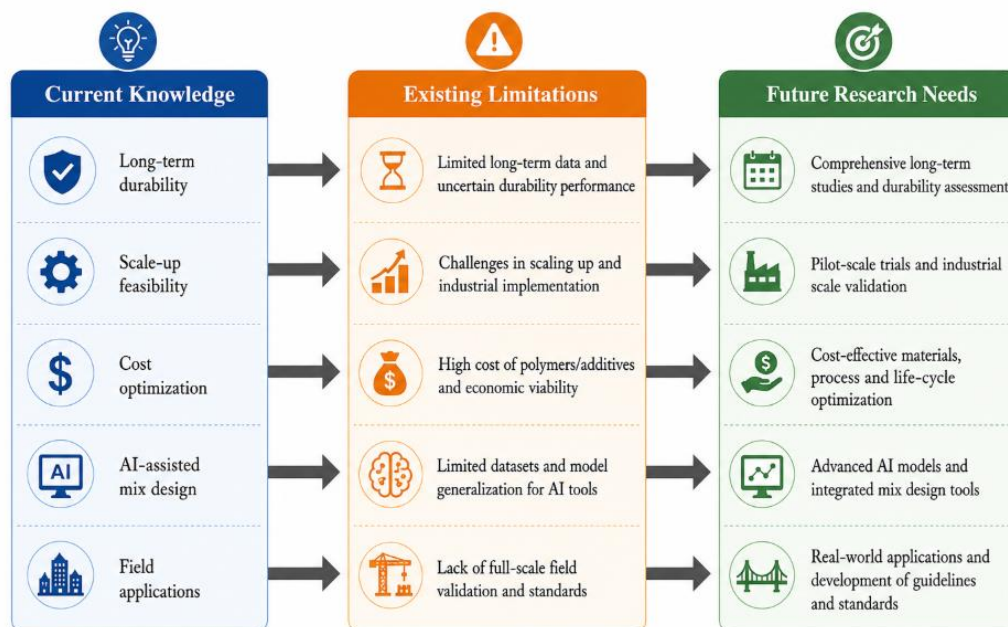


Figure 6. Research gaps and future directions in self-curing hybrid polymer–cement composite engineering

POLYMER COMPOSITE DESIGN CHALLENGES AND EMERGING RESEARCH OPPORTUNITIES.

Despite their multifunctional advantages, hybrid polymer–cement composites present complex challenges associated with phase compatibility, interfacial instability, and microstructural heterogeneity. Some of the problems associated with hybrid composite systems include the variability of the quality and physical characteristics of the recycled aggregates. Adhering old mortar on recycled aggregates is known to increase the levels of water absorption and porosity, thus adversely affecting the strength of the material. Polymer modification increases material cost because of additional polymeric constituents and may create compatibility challenges with specific cementitious systems. Effective self-

curing performance depends on precise moisture regulation and uniform distribution of internal curing agents within the composite matrix. Failure to provide the right amount of internal water release will hinder proper hydration and strengthening [15]. Also, there is limited data on the durability of such systems for long periods. Future research needs to explore ways to develop sophisticated processing methods of recycled aggregates, effective polymer compositions that are cost-effective, and better internal curing systems to regulate moisture. Future investigations should prioritize micromechanical characterization, interface evolution analysis, and predictive modeling of structure–property relationships. Such investigations will support the optimized design and broader implementation of advanced hybrid polymer–cement composites. The key areas requiring further research are illustrated in Figure 6. into Advanced hybrid polymer-cement composites have been illustrated in Figure 6 .

CONCLUSION

Hybrid polymer–cement composites incorporating recycled aggregates and internal curing systems represent an effective strategy for enhancing the mechanical performance, durability, and sustainability of cement-based materials. The integration of polymer modification, internal curing mechanisms, and recycled aggregate utilization promotes microstructural densification, improved interfacial bonding, and reduced pore connectivity, resulting in superior composite performance.

Despite these advantages, several challenges remain, including variability in recycled aggregate properties, compatibility issues between polymer modifiers and cementitious phases, and the need for precise control of internal curing processes. Addressing these limitations requires systematic optimization of material selection, interface engineering, and moisture regulation strategies.

Future research should emphasize long-term durability assessment, advanced microstructural characterization, and predictive modeling of structure–property relationships. These efforts will support the development of high-performance, sustainable hybrid polymer–cement composites for advanced construction applications.

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