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# Unlocking toughness in 3D-printed cement through bio-inspired designs: current status and future perspectives

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To address the intrinsic brittleness of conventional cementitious materials, researchers have drawn inspiration from natural biological structures to develop advanced bio-inspired cementitious composites through three-dimensional (3D) printing. This approach exploits additive manufacturing to replicate natural architectures—such as the “brick-and-mortar” structure of nacre—by precisely organizing hard cementitious and soft polymeric phases across multiple scales. As a result, novel composites exhibiting a synergistic combination of high strength and toughness have been achieved. This review systematically summarizes recent progress in this interdisciplinary field, emphasizing four key aspects: the composition and structure of bio-inspired materials; the design principles and fabrication methods for bio-inspired cementitious materials; bio-inspired toughening strategies and their associated mechanical models; and the integration of bio-inspired concepts with 3D printing technology. The interrelationships among these topics, key scientific challenges, and state-of-the-art developments are discussed in depth. Finally, this work highlights the remaining obstacles for engineering applications and provides a forward-looking perspective on future research directions.

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## 1 Introduction

Driven by the global pursuit of sustainable development, the “dual-carbon” strategy has imposed disruptive demands for material innovation within traditional high-carbon emission industries. As the most widely used building material in civil engineering, conventional cementitious materials play a pivotal role in global infrastructure construction. However, their intrinsic brittleness, weak interlayer bonding, and lack of multifunctionality have become fundamental bottlenecks hindering the advancement of modern engineering toward high performance and multi-functionality.<sup>1</sup> Natural biological materials (such as bamboo, nacre, and bone) achieve exceptional mechanical efficiency, particularly high toughness through complex hierarchical architectures, despite relatively simple chemical compositions.<sup>2</sup>

To address these fabrication constraints, 3D printing serves as a critical technological bridge. Unlike conventional casting

methods, this technique enables the direct construction of complex topologies through precise layer-by-layer deposition. Such unprecedented design freedom allows for the faithful replication of intricate natural hierarchies that are otherwise impossible to manufacture. Thus, the convergence of bio-inspired principles and advanced manufacturing presents a robust strategy to “unlock” the latent toughness of cement-based materials.<sup>3</sup>

However, how to accurately reproduce the microscopic and delicate multi-level structure of this natural material in the macroscopic cement-based material is a key challenge to achieve biomimetic toughening. Traditional pouring and forming processes are difficult to meet the demanding requirements for customized design and precise control of the internal components of the material. For this purpose, 3D printing technology has been adopted. 3D printing technology, also known as additive manufacturing, is based on the basic principle of forming physical parts by stacking materials layer by layer based on digital models. In the construction of traditional cement-based materials, complex geometric configurations such as curved walls, hollow structures, and biomimetic forms usually require customized templates to assist in molding. This not only increases construction costs and prolongs the construction period, but also makes some special structures difficult to achieve due to traditional processes. In contrast, 3D printing technology adopts a forming method without template layer by layer deposition, which can directly and accurately construct

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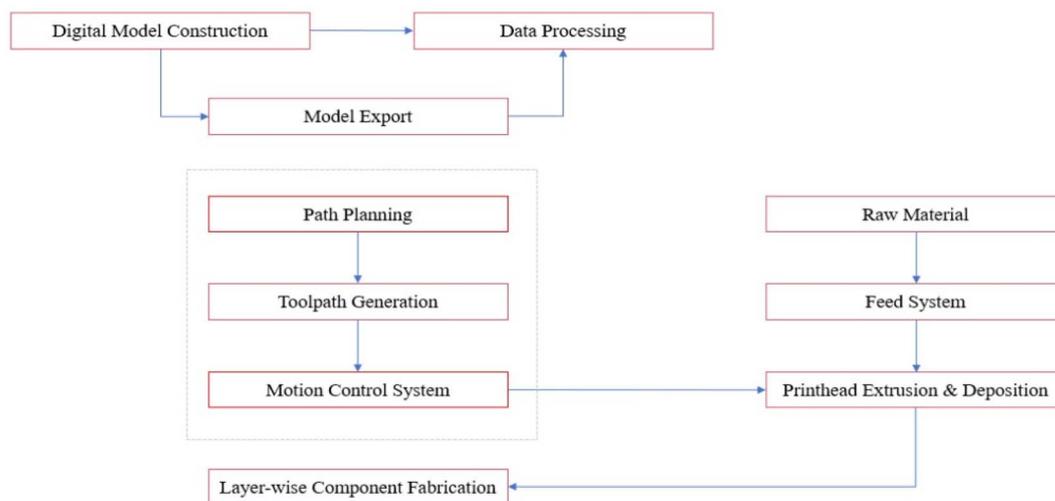


Fig. 1 3D printing process.

parts with complex topologies based on digital models without the assistance of templates. This feature broadens the possibilities of structural design and optimizes the construction process.<sup>4</sup> The schematic diagram of the 3D printing process is shown in Fig. 1.

## 2 Composition and structure of biomimetic materials

### 2.1 Multi-level structure of shell nacre “brick-and-mortar” and its mechanical mechanism

Shell nacre (as shown in Fig. 2) is a typical natural organic-inorganic layered composite with a characteristic “brick-and-mortar” structure composed of about 95% aragonite flakes (brick “phase”) and 5% organic matter (the “mud” phase) alternately layered.<sup>5</sup> Among them, aragonite sheets present a sheet-like form with a diameter of 5–8  $\mu\text{m}$  and a thickness of 0.4  $\mu\text{m}$ , and a single layer is further formed by the aggregation of pebble-like polygonal nanograins.<sup>6,7</sup>

These structural characteristics determine its unique mechanical properties: at the nanoscale, the nanoparticles inside aragonite sheets relieve local stress through lattice deformation and relative slippage, thereby inhibiting stress concentration; At the microscopic level, the mechanical interlock formed by the microscopic protrusions on the surface of the inorganic phase and the connection of mineral bridges between the layers jointly enhance the interfacial bonding strength. Organic matter undergoes plastic deformation under stress and the effective combination of hydrogen bonds and aragonite flakes is effectively combined with aragonite flakes through hydrogen bonding and other intermolecular forces.<sup>8</sup> At the macroscopic level, crack deflection and aragonite chip extraction mechanism prolong the crack propagation path during fracture and constitute the main energy dissipation mechanism. The corrugated morphology of the lamellae can inhibit the lateral propagation of cracks between layers, which helps to maintain the overall stability of the structure.<sup>9</sup>

It is the synergistic effect of this multi-scale structural feature that improves the fracture toughness of nacre by three orders of magnitude compared with monoaragonite.<sup>10</sup> The internal correlation between structure and performance provides an important theoretical basis and practical enlightenment for the design and construction of biomimetic materials. However, there are still key challenges in reproducing nacre structures in cementitious materials. On the one hand, the mechanical properties of the inherent interfacial transition zone (ITZ) in cement matrix are much weaker than those of natural organic matrix due to their heterogeneity and high brittleness. On the other hand, the volume shrinkage and pore formation associated with the hardening process of cementitious materials can impair the overall stability of the layered structure.<sup>11</sup> Therefore, how to regulate the orderly assembly of layers at the macroscopic scale and achieve “rigidity-toughness” coupling at the interface scale has become the core scientific problem of current research in this field. Existing studies have introduced functional components such as cellulose nanocrystals, graphene oxide, and polymer emulsions to construct artificial organic matter, and these attempts have made preliminary progress in enhancing crack propagation resistance and interlayer bonding, but there is still a significant gap in the hierarchical regulation of structural stability compared with natural nacre.<sup>12</sup>

### 2.2 Characterization methods and multi-scale performance analysis

To successfully replicate the sophisticated strategies of nature described above, mere observation is insufficient. We must first rigorously quantify the micro-to-macro structural features and interface behaviors. The specific mechanical properties of biomimetic structures mainly stem from their hierarchical geometry across multiple scales. Therefore, to establish the internal correlation between its structure and performance, it is necessary to comprehensively use a variety of advanced characterization methods for systematic quantitative



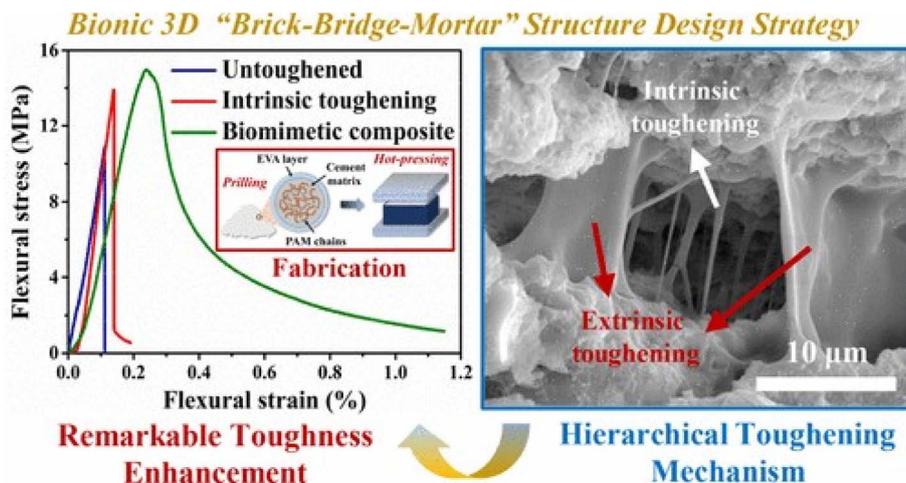


Fig. 2 Bionic 3D "brick-bridge-mortar" structure design strategy.<sup>5</sup>

characterization.<sup>13</sup> Scanning electron microscopy (SEM) and focused ion beam scanning electron microscopy (FIB-SEM) can display the thickness, interface morphology and pore structure characteristics of the sheet layer with high resolution. Micron X-ray computed tomography ( $\mu$ -CT) is used for three-dimensional structural reconstruction (as shown in Fig. 3)<sup>14</sup> for the analysis of pore network topology, the calculation of sheet orientation sequence parameters (S), and the quantitative evaluation of interlayer defects.<sup>15,16</sup>

The characterization of interface properties is usually carried out from two levels: physical topography and chemical bonding. At the physical level, atomic force microscopy (AFM) and its force spectroscopy mode can quantitatively characterize the interface roughness (Rq), adhesion (Fadh) and local modulus distribution, which provide a data basis for analyzing the slip and mechanical interlocking behavior of the interface. At the chemical level, combined with Fourier transform infrared spectroscopy (FTIR), Raman spectroscopy and X-ray photoelectron spectroscopy (XPS) and other technologies, the chemical interactions such as hydrogen bonds, carboxyl groups and Ca-O coordination bonds formed between cement hydration products and organic phases can be identified. The literature suggests that the combination of specific interfacial roughness

and weak chemical bonding can induce "controlled slip" behavior, which is a key factor affecting the combination of macroscopic strength and toughness of materials.<sup>17</sup>

The characterization of local mechanical properties is mainly done by nanoindentation technology. The distribution of elastic modulus ( $E$ ), hardness ( $H$ ) and creep characteristics of brick phase, mud phase and interface transition zone in the material can be drawn by grid indentation test. It has been shown that the elastic modulus of the cement phase is usually only 20–40%,<sup>18</sup> and the interfacial bonding strength is a key determinant of the crack propagation path. A weak interface causes cracks to deflect along it, while a strong interface causes cracks to pass through the layer. Shi *et al.*<sup>19</sup> used *in situ* scanning electron microscopy (SEM) for digital image correlation (DIC/DVC) technology to observe the crack propagation behavior in real time. This method can characterize the key toughening mechanisms such as strain field evolution, interfacial debonding, and crack bridging at the crack tip, so as to establish the correlation between microscopic damage events and macroscopic fracture properties of materials. These physical processes directly observed and quantified through experiments provide a key basis for constructing and validating multi-scale mechanical models.

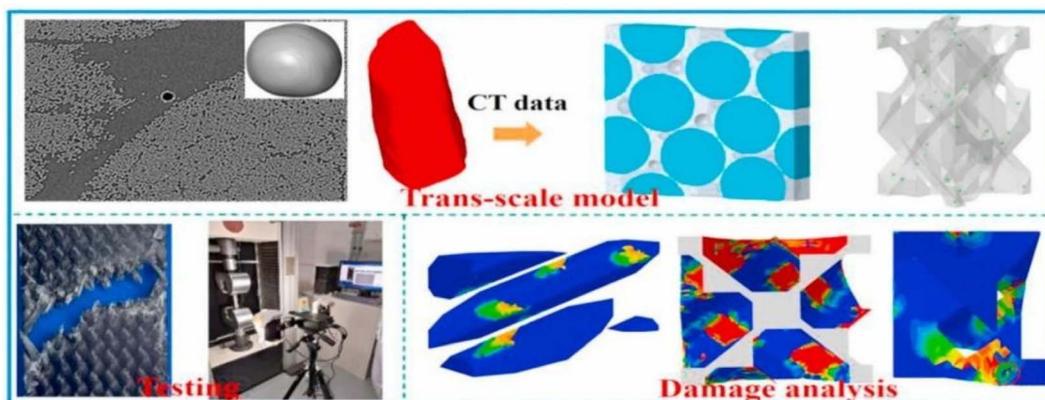


Fig. 3 Cross-scale modeling and damage analysis of materials based on CT scans.<sup>14</sup>



### 2.3 Other bionic prototypes and their applications

In addition to nacre, a variety of other biological structures can also be used as a reference for the design of cementitious materials. In 2019, Professor Zadpoor<sup>20</sup> pointed out that the porous and ordered structure of bones can be used to construct materials that are both lightweight and mechanically anisotropic. Similarly, Sun *et al.*<sup>21</sup> systematically reviewed the multi-scale structural properties of bamboo and its biomimetic applications, revealing the advantages of imitation bamboo design in generally improving material bearing capacity and energy absorption. In the exploration of lightweight and high-strength structures, Mao *et al.*<sup>22</sup> designed and fabricated porous material structures with efficient mechanical properties through 3D printing, inspired by the lightweight porous properties of cuttlefish bones. In terms of protection, Dura *et al.*<sup>23</sup> bending stiffness characteristics of bionic protective beams inspired by arc-shaped fish scales in 2023 by combining numerical simulations and experimental methods, and found that their bending stiffness can be increased by 9% and about 6% higher than that of the reference design using straight scales. In addition to the imitation of mechanical structures, biomimicry has also opened up new ideas for the design of functional materials. For example, Li *et al.*<sup>24</sup> combined magnetic nanoparticles, carbon nanotubes, and biomass porous carbon to rationally construct a coral-like graded composite, which provided a new idea for the design of biomimetic porous biomaterials. In addition, Gu *et al.*<sup>25</sup> reported for the first time a simple spider silk imitation strategy to prepare ultra-tough fiber spider silk that can be produced on a large scale through chemical synthesis routes, providing a reference model for studying the toughening mechanism of nanofibers. In summary, the study and imitation of these biological structures aims to improve the mechanical properties of cement-based materials on the one hand, and on the other hand, they give them new functional application possibilities, as summarized in Table 1.

A critical comparison reveals that the applicability of biomimetic prototypes is dictated by the trade-off between resolution and scale. While nacre-like laminates offer superior toughness for thin-walled structures, their micro-scale fabrication is often inefficient for large-scale printing. Conversely, bone- and bamboo-inspired gradient structures are more

compatible with coarse-aggregate extrusion, making them suitable for load-bearing infrastructure. Thus, successful biomimetic design requires matching the biological prototype to both the structural performance goals and the manufacturing constraints of the 3D printing system.

## 3 Design theory and preparation method of biomimetic cementitious materials

Having analyzed the structural blueprints provided by nature in the previous section, the next challenge lies in translating these biological principles into engineering reality. The design and preparation methods of biomimetic cementitious materials directly determine whether they can reproduce the functions of biological structures. This process involves the combination of modern cement technology with the principles of natural biomaterials, with the goal of not only meeting specific mechanical properties but also the multifunctional integration of materials.<sup>26</sup> This chapter mainly covers the design principles, optional material systems, and main preparation methods of biomimetic cementitious materials, and analyzes the current technical challenges in this field.<sup>27</sup>

### 3.1 Bio-inspired structural design principles

For cement-based materials, the design guidelines of biomimetic structures are the key to realizing the functionalization of materials. These guidelines emphasize how to draw on the structure–function relationship of biomaterials to optimize the performance of cementitious materials. The core is to construct a multi-level ordered internal configuration to regulate the mechanical properties of materials. Gao *et al.*<sup>28</sup> pre-constructed a mesoscale organic matrix with a layered topology to guide the orderly assembly and mineralization of nanoscale mineral crystals, resulting in the formation of bulky composites with hierarchical structures (from nanometers to millimeters). Achieve synergistic improvement of strength and toughness. In 2023, Mao *et al.*<sup>29</sup> used interfacial chemical bonding to regulate the toughening mechanism, selected the appropriate bonding strategy according to the load type, and adopted a gradient bonding design to suppress stress concentration. Biomimetic

Table 1 Bio-inspired design applications of various natural biomaterials

Biological prototype	Key structural features	Key mechanical/functional benefits	Biomimetic applications
Bones <sup>20</sup>	Directional hole structure	Lightweight and high specific strength	Design porous lightweight materials
Bamboo <sup>21</sup>	Gradient fiber distribution	Crack deflection and blocking	Fiber directional enhancement
Cuttlefish bone <sup>22</sup>	Honeycomb-like porous structure	High strength and high toughness work together	Development of insulation composite materials
Fish scales/Carapace <sup>23</sup>	Layered lamination structure	It has excellent protection functions	Design new protective materials
Coral <sup>24</sup>	Branched structure	High porosity, good biocompatibility	Biomimetic porous biomaterials
Spider silk <sup>25</sup>	Nanofiber hierarchy	High strength and high toughness work together	Research on nano toughening mechanism



design should focus on the integrated integration of function and structure, and learn from the principle of multifunctional coupling of organisms to achieve the specific functions of biomimetic materials. Ma *et al.*<sup>30</sup> studied the wettability analysis and design of micro-nano superhydrophobic surfaces and found that the micro-nano hierarchical configuration of lotus leaves reproduces superhydrophobic properties with a contact angle of  $>150^\circ$ . Dai *et al.*<sup>31</sup> “ice template method” to fabricate high-performance biomimetic porous materials by drawing on bioporous materials such as polar bear hair, wood, bamboo, and cuttlefish bones. At the same time, we should fully consider the adaptability constraints of the preparation process, be compatible with engineering production lines, strictly control costs,<sup>32</sup> and ensure environmental tolerance.<sup>33</sup> Together, these design principles form the basic framework for the functionalization of biomimetic cementitious materials. The bionic design and manufacturing process diagram are shown in Fig. 4.

### 3.2 The main material system and its functional characteristics of biomimetic cementitious materials

In biomimetic design applications, different cement systems exhibit their own unique performance characteristics.<sup>35</sup> The

process of ordinary silicate cement (OPC) is mature and low-cost, and the masonry–mortar composite interface is constructed by incorporating graphene and other reinforcing components. As the most widely used cementitious material, although its toughness and strength are low, its rheology and mechanical properties can be effectively regulated by incorporating nanomaterials such as graphene, nanosilicon, and carbon fiber and functional admixtures.<sup>36</sup> In contrast, geopolymer cement has low carbon emissions and high temperature resistance.<sup>37</sup> Studies have shown that it can be used to simulate the inorganic binding mechanism during natural biomineralization, and has potential for application in the design of biomimetic materials in high-temperature or corrosive environments.<sup>38</sup> At present, how to effectively improve the toughness of geopolymer-based biomimetic materials and optimize their interlayer interface properties is a key problem to be solved in this field. In addition, magnesium-based cement has carbonation self-healing ability and low carbon emissions, making it suitable for wet environments. Combining it with biomimetic structures is one way to improve the toughness and durability of materials. However, the regulation of the carbonization reaction rate of this system and its impact on long-term

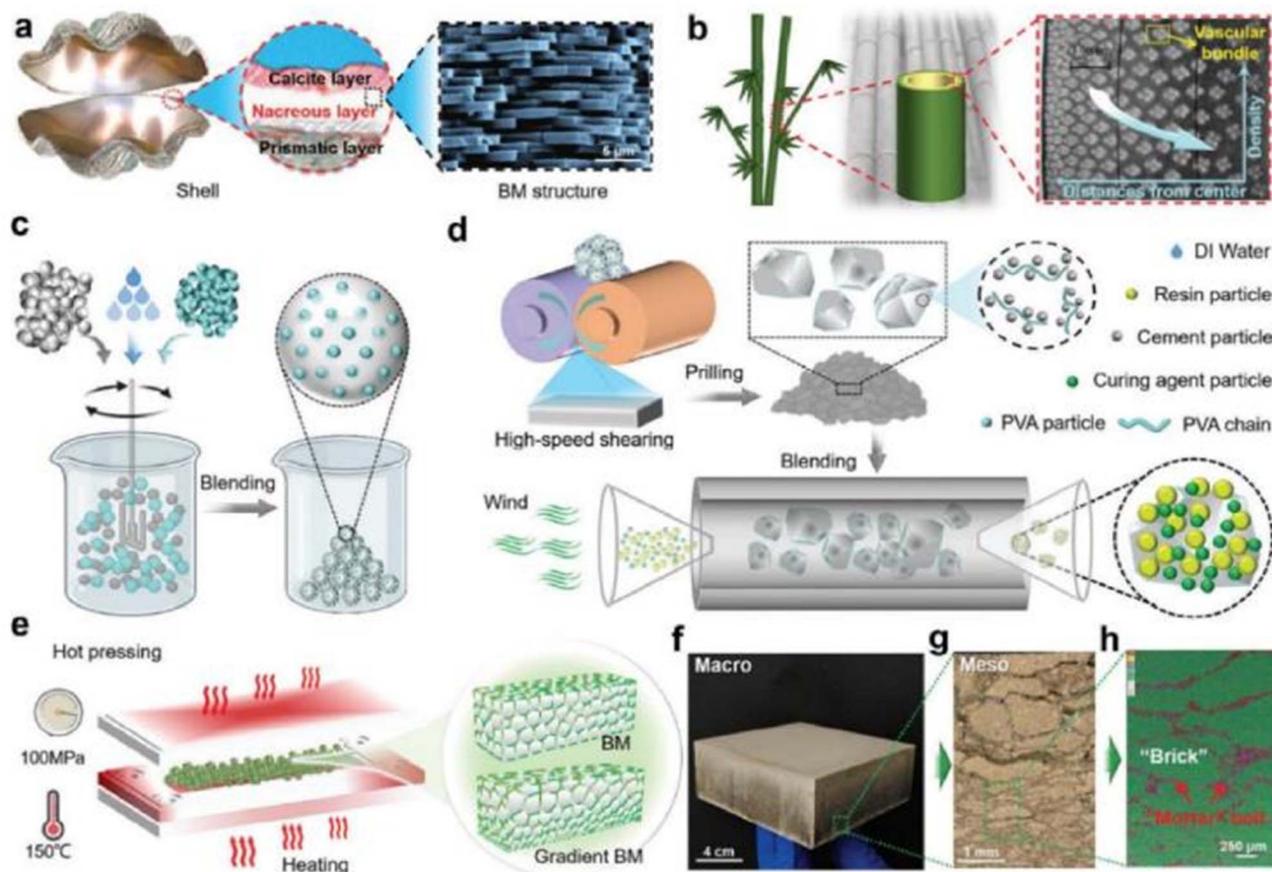


Fig. 4 Biomimetic design and manufacturing process diagram of cementitious composites with gradient “brick-mortar” (BM) structure.<sup>34</sup> (a) Hierarchical BM structure of mollusk shell nacre; (b) gradient structure in bamboo, schematic diagram of cement–resin composite preparation technology, including (c) pre-mixing, (d) high-speed shearing and granulation, and (e) hot pressing; (f–h) multiscale characterization of large-scale gradient biomimetic composites.



Table 2 Biomimetic compatibility of different cement-based systems<sup>44</sup>

Materials systems	Key advantages	Key challenges/research focus	Application of bio-inspired design
Ordinary portland cement (OPC) <sup>36</sup>	Low cost, high adaptability	Low fracture toughness	Foundational “brick-and-mortar” structural mimicry
Geopolymer cement <sup>37</sup>	Low-carbon, high-temperature and corrosion resistance	Toughness enhancement and interfacial optimization	Biomimetic structures for harsh environments
Magnesium-based cement <sup>38</sup>	Low CO <sub>2</sub> emission, self-healing capability	Carbonation rate control	Self-healing and durable structures
Ultra-high performance concrete (UHPC) <sup>39</sup>	Ultra-high strength and toughness	High cost, workability issues	Scenarios requiring synergistic high performance
Fiber-reinforced composites <sup>41</sup>	Enhanced toughness and crack resistance	Homogeneity of fiber dispersion	Impact-resistant biomimetic structures
Calcium sulfoaluminate (CSA) cement <sup>42</sup>	Rapid hardening, high early strength	Long-term durability studies	Biomimetic structures for rapid construction

structural stability need to be further clarified.<sup>39</sup> Ultra-high-performance concrete (UHPC) matrix is dense and reinforced with steel fibers, making it suitable for high-precision biomimetic structures in complex stress environments. Combined with fiber reinforcement effects and interface regulation methods, the application of UHPC in biomimetic structure replication is being studied.<sup>40</sup> Meanwhile, fiber-reinforced composite concrete (FRCC) is a class of composite materials that incorporate fibers (such as steel fibers, polymer fibers, or basalt fibers) into the cement matrix to improve toughness and crack resistance. The toughening mechanism is mainly due to the crack bridging of fibers, combined with 3D printing technology to enhance the directionality of the material.<sup>41</sup> Sulfur aluminate cement has fast hardening, early strength, low alkalinity and good durability. Compared with ordinary silicate cement, aluminate sulfate cement hydrates quickly, can achieve high strength within a few hours, and is suitable for rapid printing and molding.<sup>42</sup> Based on the above characteristics, sulfoaluminate cement has application prospects in emergency repair and rapid prefabrication of biomimetic components.<sup>43</sup> A comparison of the biomimetic adaptability for these different cementitious material systems is presented in Table 2.

A comparative analysis indicates that no single material system constitutes a universally ideal solution for biomimetic 3D printing at present. While Ordinary Portland Cement (OPC) is economically viable, its intrinsic brittleness severely limits its ability to mimic the flexible “organic-like” phases found in nature. Conversely, while polymers and fiber-reinforced composites offer superior toughness and ductility, they introduce significant challenges regarding interfacial compatibility and entail higher material costs. Geopolymers present a promising sustainable alternative with enhanced fire resistance; however, their rheological consistency during printing remains less standardized compared to OPC. Consequently, the field is witnessing a paradigm shift toward “multi-material printing”. Rather than relying on a monolithic material, future research is increasingly pivoting toward dual-nozzle systems that simultaneously deposit high-strength cement (simulating the “brick”) and compliant polymers (simulating the “mortar”). Nevertheless, it must be noted that this approach significantly escalates the complexity of interfacial bonding control between the heterogeneous phases.

### 3.3 Preparation method of biomimetic cementitious materials

The preparation of biomimetic cementitious materials aims to learn from natural biological configurations (such as nacre “brick-and-mortar” structure), and to compound flexible phases such as polymers and cement-based hard phases in a multi-scale orderly manner through advanced technology to construct high-strength and tough composites. At present, the formwork-assisted method is one of the mainstream technical paths, and its core is to first construct an orderly porous skeleton and then carry out secondary filling. For example, Chen *et al.*<sup>45</sup> combined the ice template method and vacuum pressure impregnation to induce cement particles to self-assemble into a directional layered skeleton (thickness 10–100 μm) through a bidirectional freezing process, which was filled with PVA hydrogel impregnation to form a highly ordered “brick-and-mortar” structure. In order to obtain more precise structural control, Gui *et al.*<sup>46</sup> introduced an internal cold index (ICF) in the freeze-casting process to achieve programmable control of the ordered microstructure of the aerogel layer. Similarly, Guo *et al.*<sup>47</sup> used 3D printing technology to prefabricate 3D graphene aerogel templates, combined with vacuum infusion of polymer (PCL) and low-temperature hot pressing, to prepare multi-scale composites with shell-like “brick-and-mortar” structures, which achieved synergistic improvement in strength and toughness.

Unlike the formwork method, another broad category of methods focuses on integrated molding, which is the direct construction of composite materials through means such as mechanochemistry or additive manufacturing. In order to solve the brittleness problem of traditional concrete, Wu *et al.*<sup>48</sup> combined high-speed shearing, hot roll calendaring, and hot press forming to directly construct a multi-level structure imitating nacre inside the cement matrix, which significantly enhanced its mechanical properties. Song *et al.*<sup>49</sup> pioneered an innovative coaxial 3D printing method can directly prepare composites with imitation enamel microstructure by extruding rigid silicate ceramics and flexible epoxy resin simultaneously, and their flexural strength and fracture toughness reach 2.1 times and 47.5 times that of pure silicate ceramics, respectively. Fig. 5 illustrates the primary fabrication process of the bio-inspired, high-toughness cementitious composite.<sup>50</sup>



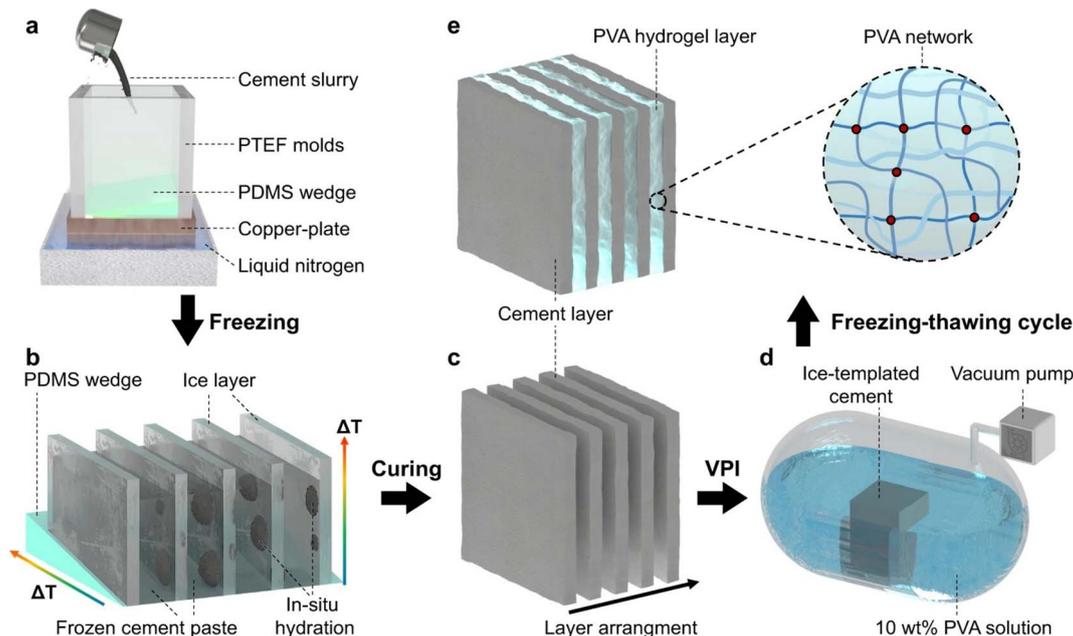


Fig. 5 The primary fabrication process of the bio-inspired, high-toughness cementitious composite. (a) The mixing of cement slurries in the mold with internal dimensions of  $50 \times 50 \times 50$  mm. (b) Cement slurries solidify into ice layers by a bidirectional freezing gradient both vertically and horizontally. Cement particles are squeezed between ice layers and slowly *in situ* hydrate. (c) Cement particles hydrate into order layers during a thawing and curing period with a thickness of  $10\text{--}100$   $\mu\text{m}$ . (d) The PVA solution was filled into the pores between neighboring cement sheets by VPI (vacuum pressure impregnating). (e) PVA hydrogels were formed between the cement lamellae after 2–3 freezing–thawing cycles.<sup>45</sup>

## 4 Bionic toughening strategy and mechanical model

### 4.1 The realization and role of bionic toughening mechanism

In order to overcome the inherent brittleness of cement-based materials, researchers have developed a series of biomimetic toughening strategies based on the principle of structure–function synergy of biomaterials. Fig. 6 shows the microstructure of natural mother-of-pearl and biomimetic high-strength cement-based materials. The core of these strategies is the introduction of multi-scale energy dissipation mechanisms such as crack deflection, crack bridging, and interfacial slip within the material through engineering design.<sup>51</sup> The quantitative analysis and performance prediction of these complex mechanical behaviors rely on advanced mechanical models. This includes classical models of hysteresis theory, as well as finite element methods (FEM) and discrete element methods (DEM) that can accurately capture micromechanical responses. In recent years, data-driven methods represented by machine learning have provided a new paradigm for such modeling, providing a powerful tool for connecting experimental data with theoretical analysis.<sup>52</sup>

**4.1.1 Crack deflection and bridge connection.** The crack deflection mechanism is the basis of biomimetic toughening. In natural materials, cracks deflect when they encounter mineral sheets or fibers, dispersing energy and prolonging the crack propagation path. The crack deflection effect of biomimetic

cementitious materials is widely used in natural biomaterials, especially the layered structure of nacre and the fiber-reinforced design of bamboo.<sup>54</sup> Crack deflection effectively extends the crack propagation path, thereby consuming more energy. For example, the nacre is structurally designed to allow cracks to deflect between layers and be inhibited by bridging effects. In actual biomimetic cementitious materials, the propagation of cracks can be effectively controlled at the microscopic scale through fiber reinforcement and mineral bridges, thereby improving the toughness and crack resistance of cementitious materials.<sup>55</sup>

**4.1.2 Interface slippage and energy dissipation.** Interfacial slip is another toughening mechanism in biomimetic cementitious materials. In natural materials, the interface layer is usually composed of soft or organic materials that provide controlled slippage when cracks encounter, greatly reducing the rate of crack propagation. In the design of biomimetic cementitious materials, the interfacial slip mechanism is considered to be one of the important means to improve the toughness of materials.<sup>56</sup> The interfacial layer in natural materials can effectively slow down the propagation of cracks through slip energy consumption. Biomimetic design simulates the natural interface structure by introducing soft phase materials (*e.g.*, polymer emulsions, nanoparticles) to optimize the interlayer bond strength and crack control capabilities of cementitious materials.<sup>57</sup> Interfacial slip not only contributes to energy dissipation, but also avoids crack propagation along a straight line through local plastic deformation, enhancing the crack resistance and toughness of the material.



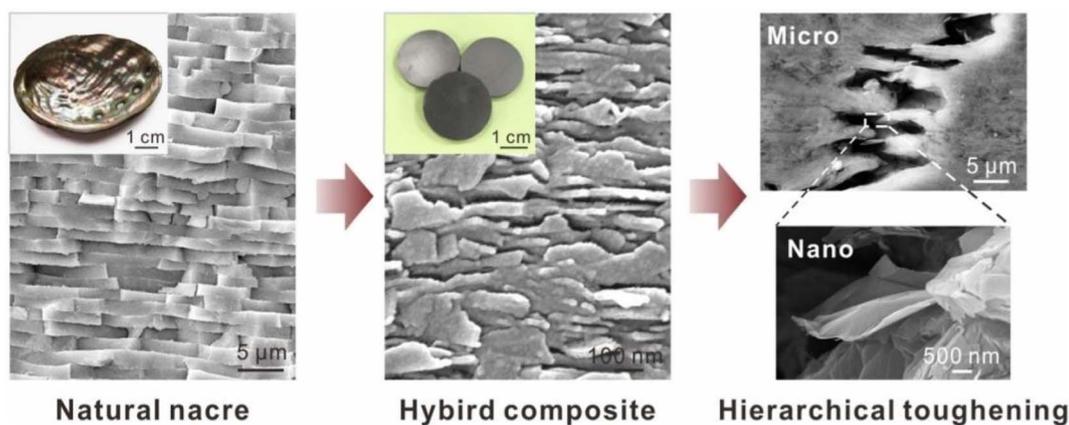


Fig. 6 Microstructure diagram of natural mother-of-pearl and biomimetic high-strength cementitious materials.<sup>53</sup>

#### 4.2 Quantitative analysis method of mechanical modeling and toughening mechanism

While experimental observations reveal the phenomenological toughening mechanisms, theoretical verification is equally crucial for predicting material behavior under complex stress states. The mechanical model provides a key tool for the research and optimization of biomimetic toughening mechanism. With the help of multi-scale modeling and numerical simulation technology, researchers can quantify the contribution weights of different toughening mechanisms to the macroscopic properties of materials, and provide scientific support for the rational design of biomimetic cementitious materials. Commonly used models include shear hysteresis models, finite element models (FEMs), discrete element methods (DEMs), and data-driven machine learning models.

**4.2.1 Stress transfer between shear hysteresis model and layered structure.** As shown in Fig. 7,<sup>58,59</sup> the shear hysteresis model is a classical model for analyzing stress transfer and crack propagation in layered structures. In the design of biomimetic cementitious materials, shear hysteresis models are used to describe the propagation path of cracks in interlayer structures, especially in designs with mineral bridges and fiber reinforcement. By establishing the mechanical properties of each layer of material (such as elastic modulus, strength,

fracture toughness, etc.), the shear hysteresis model can predict the deflection angle and propagation path of cracks, and help design parameters such as thickness and arrangement of layered structures (Fig. 7). The shear hysteresis model assumes that the shear stress of the matrix is uniformly distributed along the thickness direction, while the shear stress is not uniformly distributed along the length of the “brick”. Based on the equilibrium conditions of the force, the equilibrium equation of the representative unit can be derived:<sup>60,61</sup>

$$\tau(x) = h \frac{d\sigma_{\text{top}}}{dx} = -h \frac{d\sigma_{\text{bottom}}}{dx} \quad (1)$$

where  $\tau(x)$  is the shear stress in the matrix of the bio-inspired structure,  $\sigma_{\text{top}}$  denotes the stress in the upper brick of the Representative Volume Element (RVE),  $\sigma_{\text{bottom}}$  is the stress in the lower brick of the RVE, and  $h$  is the thickness of the brick.

Assuming that the “bricks” are arranged regularly and the overlapping length is  $L/2$ , the equivalent elastic modulus  $E$  of the stress distribution and material in the representative unit under the shear hysteresis model is further derived:

$$\sigma_{\text{top}}(x) = \frac{\sigma_0 \sin h \left( \frac{\lambda x}{2} \right) \cos h \left( \lambda \left( x - \frac{L}{4} \right) \right)}{\sin h \left( \frac{\lambda L}{4} \right)} \quad (2)$$

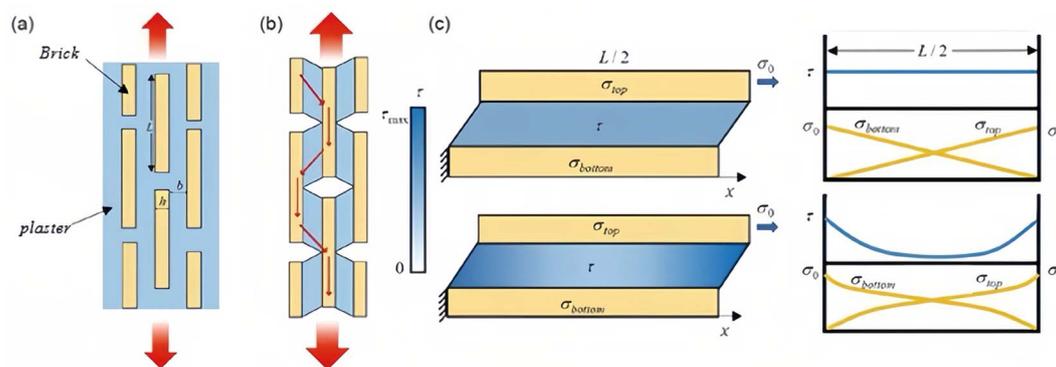


Fig. 7 Basic mechanical model of representative units of biomimetic “brick-and-mortar” staggered structures.<sup>58,59</sup> (a) Geometric model under tensile loading, (b) shear stress transfer mechanism, (c) analytical models of interfacial stress distributions.



$$\sigma_{\text{bottom}}(x) = \sigma_{\text{top}} \left( \frac{L}{2} - x \right) \quad (3)$$

$$\tau'(x) = \frac{\sigma_0 h \lambda \cos h \left( \lambda \left( \frac{L}{2} - x \right) / 2 \right)}{2 \sin h \left( \frac{\lambda L}{4} \right)} \quad (4)$$

$$E = \frac{E_p \lambda h L}{(2h + b) \left[ \frac{\lambda L}{2} + 2 \cos h \left( \frac{\lambda L}{4} \right) \right]} \quad (5)$$

where  $b$  is the thickness of the matrix layer,  $L$  is the length of the brick,  $h$  is the thickness of the brick.  $\sigma_0$  is the applied external load;  $\mu_m$  is the shear modulus  $\lambda = \sqrt{\frac{2\mu_m}{E_b h b_0}}$ .

Relevance to 3D printing: in the context of 3D-printed biomimetic structures, the parameters  $h$  and  $b$  are strictly governed by the nozzle diameter and layer height settings. The shear lag model is particularly useful here to calculate the critical overlap length required between adjacent printed layers to ensure that the stress is effectively transferred through the weaker cementitious interface, preventing delamination failure typical in printed components.

#### 4.2.2 Finite element analysis and multi-scale modeling.

Finite element analysis (FEM) is a numerical simulation method widely used in biomimetic cementitious materials. FEM calculates the mechanical response of each element by dividing the material into small elements, resulting in the stress, strain, and displacement distribution of the entire material. In biomimetic design, FEM models can be used to simulate the propagation of cracks in materials and calculate the contribution of different toughening mechanisms to the overall properties of the material.<sup>62</sup>

Multi-scale modeling, which combines the relationship between macroscopic mechanical behavior and microstructure, can reveal the synergy of different toughening mechanisms.<sup>63</sup> The application of FEM in biomimetic cementitious materials, especially in combination with nanostructures and porous materials, enables the microscopic behavior of materials to be closely linked to macroscopic properties, thereby enabling accurate design and prediction of material properties.<sup>64</sup>

Unlike traditional cast concrete, 3D-printed cementitious composites inherently contain structural anisotropies, including inter-filament voids and Interface Transition Zones (ITZs) derived from the raster deposition path. Advanced FEM techniques, such as Cohesive Zone Modeling, are essential for capturing crack propagation behaviors along these specific printing interfaces. By integrating the exact printing toolpath into the finite element mesh, it becomes possible to predict how various deposition patterns influence the energy dissipation and overall toughness of the biomimetic structure.

**4.2.3 Discrete element methods and particle behavior.** The discrete element method (DEM) is used to simulate the interaction between particles and the effect of particle buildup on material properties. In biomimetic cementitious materials, particle size distribution, particle accumulation mode and pore

structure inside the material directly affect the chemical properties. Through DEM simulations, researchers can quantify the effects of different particle configurations on crack propagation, interfacial binding, and energy dissipation, and then predict the optimization effect of different designs on the mechanical properties of materials.<sup>65</sup>

DEM has unique advantages in simulating the fracture process of materials, especially the complex fracture behavior of biomimetic materials. In the DEM model, the initiation and propagation of cracks are naturally represented as the breakage of the bonding bonds between the particles. Researchers can visually observe how cracks bypass hard aggregate particles (crack deflection) and how fibrous or unbroken aggregate particles act as bridging between crack surfaces (crack bridging). In addition, DEM can explicitly simulate the fiber/aggregate extraction process and calculate the energy dissipated due to friction during the process, thereby quantitatively verifying and explaining the toughening effect of biomimetic designs (such as weak interface strategies) at the particle scale.<sup>66</sup>

Furthermore, DEM is particularly valuable for simulating the granular flow dynamics within the extrusion nozzle. By modeling the interaction between the nozzle walls and the stiff particles, DEM allows researchers to predict flow-induced fiber orientation and local particle packing. This ensures that the printing process successfully achieves the specific anisotropic alignment required by bio-inspired designs, which is critical for replicating the directional strengthening mechanisms of natural materials.

## 5 Research on 3D printing fusion

3D printing technology provides a unique manufacturing platform for biomimetic cementitious materials, enabling complex natural biological structures to be reproduced in artificial materials. Compared to traditional pouring methods, 3D printing not only allows for a high degree of freedom in shape and scale, but also enables the construction of functional gradients, interface designs, and multi-scale hierarchies within the material. This degree of freedom provides the possibility to simulate the multi-level toughening mechanism of natural materials such as shells, bones, and bamboo, thus promoting breakthroughs in the direction of strength, toughness, durability, and multifunctionality of cement-based materials.<sup>67</sup>

### 5.1 Pathways for fabricating bio-inspired structures via 3D printing

The design core of biomimetic cementitious materials is a multi-level hierarchical structure coupled with macrogeometry, mesoscopic arrangement and micro interface. 3D printing technology can achieve accurate simulation of natural biological structures at various scales through differentiated strategies.<sup>68</sup> At the macro level, relying on path programming and layer-by-layer deposition technology, it can construct complex curved surfaces, spirals, or curved forms, and reproduce natural structural features such as bamboo joints and shell arcs, thereby optimizing the overall force transfer mode of components.<sup>69</sup> At the mesoscopic level, by



regulating the sedimentary path and the overlapping parameters between layers, lamellary, corrugated, or honeycomb mesoscopic arrangements can be constructed to simulate the core toughening mechanism of nacre “brick-and-mortar” composite structures or honeycomb structures.<sup>70</sup> At the micro level, relying on multi-printhead collaborative printing and functional material component regulation, flexible phases or customized interface transition layers can be accurately introduced between layers, giving the interface the characteristics of “rigid-flex synergy” and simulating the efficient energy dissipation mechanism of the organic–inorganic interface of shells.<sup>71</sup> This cross-scale structure construction strategy can enable biomimetic cementitious materials to meet the engineering bearing requirements at the macro level, and at the same time realize crack initiation inhibition and energy dissipation regulation at the micro level, and finally achieve the synergistic optimization of mechanical properties.

On this basis, the introduction of functional nanomaterials is an effective means to achieve precise microscopic control and performance enhancement. As shown in the technical route in Fig. 8, carbon-based nanomaterials such as graphene and graphene oxide are evenly dispersed in the slurry composed of cement, water, admixtures and aggregates through sonic treatment to ensure that they are uniformly dispersed, which is a key prelude to the preparation of high-performance 3D printed cementitious composites.<sup>72</sup> These dispersed nanomaterials play multiple roles during the printing process. They are both efficient nucleus growth points and nanofillers, which can accelerate cement hydration and densify the matrix microstructure. More importantly, when microcracks occur, high-strength nanosheets can effectively bridge the two ends of the crack and dissipate energy by inhibiting its propagation, thereby improving the toughness of the material at the source.

## 5.2 Self-healing strategies and durability assessments

In traditional self-healing concrete, the distribution of repair agents, whether incorporated into microcapsules or bacterial

spores, is random and limited, and once the repair agent is exhausted, the self-healing function is permanently lost.<sup>73</sup> 3D printing technology provides a new paradigm for building more efficient and reproducible self-healing systems, with the design of biomimetic vascular networks being one of the core directions.<sup>74</sup> This technology draws on the characteristics of the vascular system that circulates in living organisms, and can directly print a fine and connected hollow pipe network inside cement components by sacrificing ink or multi-channel nozzle technology.<sup>75</sup> It works by the fact that sacrificial ink (wax or gel-like material) is removed by heating or solvent rinsing after the cement has hardened, forming preset vascular channels that can be connected to external “reservoirs”; When a crack occurs somewhere in the structure and cuts off the vascular channel, capillary action or a micro-pressure gradient drives the repair agent (*e.g.*, epoxy resin prepolymer and curing agent, sodium silicate solution) to flow to the damaged area, achieving crack healing through a chemical reaction.<sup>76</sup>

Compared with traditional methods such as embedded glass tubes, the vascular network constructed by 3D printing (as shown in Fig. 9) shows higher controllability and adaptability in terms of structural designability and functional integration.<sup>77</sup> Its geometric topology can be optimized according to the structural stress concentration area to achieve precise configuration of repair functions at key parts.<sup>78</sup> At the same time, the 3D printed vascular network has good reproducible repair capabilities, and the repair agent can be perfused multiple times from the outside and circulated in the internal channel, so that the structure maintains continuous self-healing potential during the service cycle.<sup>79</sup> In addition, the network system can further realize multi-functional integrated design, and the same vascular network can undertake multi-scenario tasks in different service environments.<sup>80</sup> For example, the delivery of antifreeze liquid under low temperature conditions to enhance freeze-thaw resistance, the circulation of cooling media under high temperature conditions to relieve thermal stress, or the deployment of channels as sensors to achieve structural health monitoring reflect the integrated characteristics of “one network for multiple functions”.<sup>81</sup>

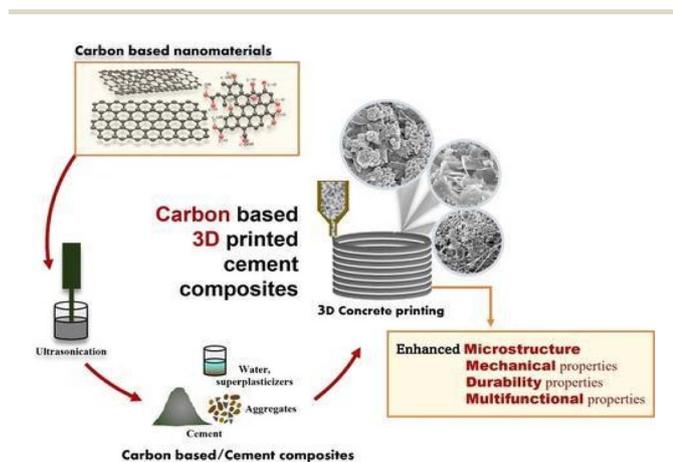


Fig. 8 Schematic diagram of the preparation process and performance improvement of carbon nanomaterial-enhanced 3D printed cementitious composites.<sup>72</sup>

## 5.3 Engineering application prospects and future research directions

The research goal of biomimetic cementitious materials is not limited to the local improvement of material properties, but is committed to transforming the principles of structural structure efficiency in nature into components and systems with engineering application value.<sup>82</sup> The hierarchical arrangement, orientation, and functional gradient characteristics of natural materials such as nacre, bone, bamboo, and honeycomb structure have direct implications for the optimization of bearing capacity, crack resistance, energy absorption efficiency, and thermal performance of building components.<sup>83</sup> The synergistic integration of such bionic design concepts with 3D printing processes can realize the synergistic coupling of form, material and function at the component scale, making the building structure more stable.



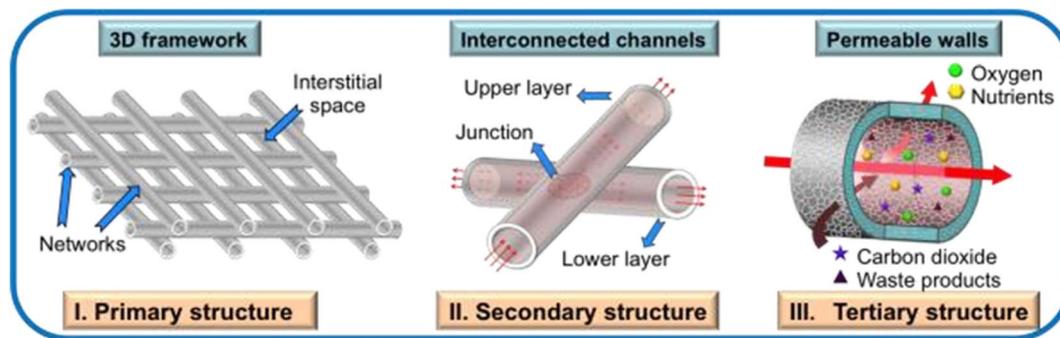


Fig. 9 Biomimetic vascular network diagram constructed by 3D printing.<sup>77</sup>

There is a clear mapping relationship between biomimetic structures and building functions.<sup>84</sup> For example, the “brick-and-mortar” staggered composite structure of nacre can be used to improve the crack resistance and toughness of walls and cladding components; the dense-porous gradient structure of bone can be applied to load-bearing components such as beams and columns to achieve a synergistic balance between lightweight and energy absorption performance. Honeycomb and bamboo structure are suitable for the design of cushioning components, thermal insulation layers and acoustic noise reduction components. In engineering design, based on the functional orientation of the above mapping relationship, the design process clarifies the structural stress and service conditions in turn, determines the microscopic mechanism of the target, maps the biomimetic structure to the component scale through parametric topology design, *etc.*, and finally completes the preparation by relying on the printing path and material allocation.<sup>85</sup>

Recent experimental validations have successfully bridged the gap between theoretical biomimicry and structural engineering, revealing that the optimal printing strategy is strictly governed by the specific service conditions. A critical comparison of recent findings highlights a distinct performance trade-off: while unidirectional printing paths ( $0^\circ$  pitch) are identified as the optimal configuration for maximizing static bearing capacity in primary gravity-load elements like beams and columns,<sup>86</sup> bio-inspired Bouligand architectures (particularly at a  $15^\circ$  pitch) prove superior for protective infrastructure subjected to impact loads, offering significantly enhanced energy dissipation through complex crack propagation mechanisms.<sup>87</sup> These results establish a concrete framework for ‘performance-based’ manufacturing, where the internal printing architecture is tailored to match the specific static or dynamic demands of the infrastructure.

3D printing technology provides support for the engineering implementation of bionic structures. By covering print path programming, local fill rate regulation, and precise control of pore distribution, the complex spatial structure of natural materials can be reproduced. With the help of multi-component collaborative printing, rigid-flexible phase coupling or functional phase directional embedding can be realized within a single component to meet the multi-scale performance requirements of biomimetic structures.<sup>88</sup> Based on this, high-strength dense slurry is selected in the main structure bearing

area, and flexible phase or slippery interface phase is used in the key area of energy dissipation or crack deflection. The porosity distribution is controlled by gradient printing technology to achieve spatial adaptation of stress performance and thermal performance. Self-repairing agents or sensing functional materials are embedded in weak areas such as component stress concentration to improve long-term service reliability. Prototype studies have confirmed that such strategies can significantly improve specific strength, fracture energy, and thermal insulation performance in small-scale samples, but they still face outstanding problems when generalized to actual engineering components.<sup>89</sup>

At present, the core challenge in the engineering process of biomimetic cementitious components is that the performance gain at the laboratory scale is difficult to be stably transmitted to large components,<sup>90</sup> and the material-process coupling relationship is complex.<sup>91</sup> However, there is currently no unified design specification and long-term service performance evaluation system for bionic printing components, which is difficult to support their engineering certification.<sup>92</sup> In order to promote technology transformation, subsequent research should carry out cross-scale multiphysics numerical simulation and experimental coupling research, formulate printing process parameter standards and quality control processes for bionic components, ensure the consistency of component preparation, and establish standardized durability test methods and life prediction models to provide a performance guarantee basis for engineering applications.<sup>93–95</sup>

## 6 Conclusions and prospects

Based on the accurate reproduction ability of natural biological structures under 3D printing technology, this paper systematically reviews the research progress of biomimetic cementitious materials in terms of composition and structure, design theory and preparation, toughening strategy and mechanical model, and focuses on the integration path of bionics and 3D printing technology and its future development direction. The following problems and future prospects are derived.

### 6.1 Existing problems

(1) Although biomimetic structural design provides new ideas for improving the brittleness of cement-based materials, the



material itself is still a performance bottleneck. Existing material systems, whether they are traditional silicate cement or new cementitious materials such as geopolymers, still face challenges in achieving the synergistic optimization of “good printability, low-carbon environmental protection characteristics, and long-term structural stability”, especially in the combination of multi-material interfaces and long-term service reliability of large-size components.

(2) The excellent mechanical properties of small-sized samples prepared at the laboratory scale are difficult to reproduce stably when enlarged to the actual size of engineering components. There is a complex coupling relationship between the rheological properties of materials, printing process parameters and the macroscopic properties of the final components, which makes it difficult to accurately predict and regulate the properties at multiple scales.

(3) At present, unified specifications and standards have not been formed for the design, manufacture and performance evaluation of bionic 3D printing components. In the process of transformation from laboratory research to engineering application, there is a lack of standardized design theory, quality control process and long-term performance evaluation method, which limits the engineering certification and popularization and application of this technology.

## 6.2 Future prospects

(1) Future research needs to be committed to building a research system from “material innovation” to “engineering transformation”. The focus is on developing multi-scale mechanical models and service life prediction methods that can accurately correlate printing processes, material microstructures and macroscopic performance of components, so as to realize the accurate design and active regulation of bionic component performance.

(2) In view of the limitations of current materials, future research focuses on the development of new cementitious material systems that can meet the three requirements of excellent printability, low-carbon attributes and long-term stability. By breaking through key technical problems such as the stable combination of interfaces and the regulation of service reliability of large-size components, it provides a new generation of high-performance material support for intelligent construction and sustainable infrastructure.

(3) In order to promote the practical application of technology, it is urgent to improve the correlation accuracy of cross-scale models, and establish a standardized system and engineering certification process from laboratory to pilot verification. This includes the formulation of printing process parameter standards and quality control methods for bionic components, as well as the establishment of standardized durability testing and life prediction models to provide reliable performance guarantee for engineering applications.

## Conflicts of interest

No potential conflict of interest was reported by the authors.

## Data availability

No data was used for the research described in the article.

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