

Toward alleviating anisotropy of 3D printed cementitious composites through rheological regulation and interfacial modification

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ABSTRACT

The inherent anisotropy generated by interlayer defects and weak interfaces between printed filaments remains a critical challenge for the structural application of 3D printed concrete (3DPC). This study presents a physico-chemical strategy to alleviate anisotropy in 3D printed white cementitious materials (3DP-WCMs) by leveraging the combined effects of needle-shaped wollastonite (WL) and spherical silica fume (SF). Oscillatory shear tests indicated that the addition of WL and SF significantly broadened the linear viscoelastic region (LVR) and delayed the onset of structural flow, suggesting the formation of a rigid physical network in the fresh state. Considering both fresh viscoelastic behavior and hardened mechanical properties, C80W12S8 was identified as the optimal mixture. At 28 days (28 d), the compressive strength of C80W12S8 reached 43.10 MPa in the A direction, while its flexural strength increased by 46.4% relative to C100 in the weakest C direction. Importantly, the proposed strategy substantially reduced mechanical anisotropy while simultaneously lowering the embodied carbon through partial replacement of WPC by WL and SF. The embodied carbon was reduced by 15.6% for C80W12S8 and 45.9% for C50W30S20 compared with C100. Meanwhile, the flexural anisotropy coefficient (I_p) of C80W12S8 at 28 d decreased from 0.29 to 0.13, while its compressive anisotropy coefficient (I_c) dropped from 0.52 to 0.40 relative to C100. In addition, separate-hydration analyses supported a possible mechanism in which Ca^{2+} -mediated electrostatic interactions may promote the association of SF with WL surfaces, while SF provided nucleation sites for the formation of a denser and more continuous C-S-H network.

1. Introduction

The emergence of 3D printed concrete (3DPC) is advancing the construction industry, offering significant potential for innovative design and construction automation (Yang et al., 2026). This technology facilitates the fabrication of complex geometries and customized structural components, while significantly offering the potential to reduce material waste and formwork demand (Nguyen-Van et al., 2026). Despite these advantages, Portland cement-based materials (PCMs) remain the predominant matrix in 3D printing technology, and high cement demand can offset part of the environmental benefit enabled by digital fabrication. Thus, developing lower-carbon printable cementitious materials while maintaining adequate printing and mechanical performance remains important (Robayo Salazar et al., 2024).

However, 3DPC shares inherent vulnerabilities with traditional

PCMs, notably a brittle failure response under tension and low fracture toughness, which severely restricts its large-scale structural applications (Gan et al., 2024; Khan and McNally, 2024). Furthermore, the layer-by-layer deposition of 3DPC complicates the simultaneous integration of conventional reinforcement elements, such as rebar (Li et al., 2024). Consequently, numerous studies have incorporated various fiber-reinforcement strategies to enhance the toughness of 3DPC, including glass fiber (Jin et al., 2022), carbon fiber (Ateeq et al., 2023), polypropylene fiber (Moelich et al., 2022) and fiber textiles (Marchment and Sanjayan, 2020). Nevertheless, fibers tend to align along the printing direction during nozzle extrusion, particularly when their length exceeds the nozzle diameter (Dong et al., 2024). This alignment unintentionally exacerbates the mechanical anisotropy of 3DPC, which stems from the layered fabrication process (Xu et al., 2025; Zhou et al., 2023). For instance, Zhang et al. (Zhang et al., 2023) reported that the

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anisotropy ratio significantly rose from 1.11 to 1.37 in the flexural strength of 3D printed specimens as the steel fiber content varied from 0% to 2%.

Such mechanical anisotropy is characterized by significant variations in bearing capacity under different loading orientations (Yu et al., 2023). In practical applications, structural stresses tend to concentrate at weak interlayer boundaries, increasing the risk of cracking under long-term service loads or extreme environments (Flatt and Wangler, 2022; Le et al., 2012). This ultimately threatens the structural integrity and reliability of printed components. Computational modelling has been widely used to capture and predict the anisotropic response of 3D printed cementitious structures by considering factors such as printing direction, interlayer interfaces, and material orientation (Nguyen-Van et al., 2023a; Nguyen-Van et al., 2023b). Such approaches are valuable for understanding directional structural behavior, while they mainly focus on predicting anisotropy rather than alleviating its material origin. Therefore, strategies capable of mitigating the material origins of anisotropy remain highly desirable. One possible route to alleviate anisotropy is to reduce the preferential orientation of reinforcing fibers during extrusion. Although enlarging the nozzle diameter can theoretically increase the probability of disordered fiber orientation, its effectiveness diminishes significantly beyond a certain ratio of nozzle diameter to fiber length (Arunothayan et al., 2021). Alternatively, incorporating intrinsically disordered microfibers may offer a more suitable solution to overcome anisotropy in flexural strength of 3DPC without requiring an increase in nozzle diameter.

Wollastonite (WL, CaSiO_3), a natural mineral with a distinctive needle-shaped morphology, is widely available as a by-product of mining and industrial processes (He et al., 2020). Due to its lower cost than some common fibers (He et al., 2020; Soliman and Nehdi, 2012), high whiteness and distinct micro-fibrous characteristics, WL represents a promising candidate for reinforcing cementitious materials through physical bridging and reducing the global warming potential of the construction sector (Nair and Sairam, 2021). More recently, WL has also been introduced into 3D printed cementitious composites. Bong et al. (Bong et al., 2022) incorporated wollastonite into a 3D printed geopolymer and reported improvements in rheological properties and flexural performance, while Zhao et al. (Zhao et al., 2025) investigated the influence of wollastonite content on the rheological and mechanical properties of 3D printed magnesium potassium phosphate cement. These studies demonstrated the feasibility of WL as a mineral modifier for 3D printing, but primarily considered WL as an individual component and focused on printability, rheology, and overall mechanical performance rather than the mitigation of directional mechanical anisotropy.

In PCMs, the highly alkaline environment of cement paste renders WL chemically stable, hindering the release of calcium and silicate ions (Xu et al., 2020). Although acid treatments can decompose WL to release active ions (Xue et al., 2015), they may damage the needle-shaped morphology, while unavoidably generating wastewater that challenges environmental regulations. To resolve this limitation, green and non-destructive surface modification methods are urgently required to optimize the interfacial bonding of WL. Beyond traditional chemical modifications, physical adsorption of ultrafine particles onto the WL surface represents a more sustainable approach to enhance the interfacial contribution of WL. Supplementary cementitious materials, including ground granulated blast-furnace slag (GGBFS), fly ash (FA) and silica fume (SF), have been widely used in 3D printed cementitious materials to regulate rheology and improve mechanical performance (Zhao et al., 2024a). Among them, SF is particularly attractive because of its ultrafine particle size and high pozzolanic reactivity. Previous colloid chemistry studies verified that Ca^{2+} can adsorb onto the surface of SF particles, altering their surface charge from negative to positive (C. F. Zukoski and Saville, 1986; Zelic et al., 2000). Meanwhile, SF acts not only as a filler component, but also consumes calcium hydroxide to form C-S-H gel, thereby accelerating cement hydration and the hardening

process (Hou et al., 2015; Xu et al., 2023). These characteristics suggest that the combined incorporation of WL and SF may simultaneously improve physical bridging and interfacial microstructural development. Although the effect of WL on rheological regulation and printability has been widely investigated, its combined incorporation with SF in relation to interfacial microstructure and mechanical anisotropy has not been systematically clarified.

Therefore, this study systematically investigated the combined effects of WL and SF on the rheological properties, separate-hydration kinetics and mechanical anisotropy of 3DP-WCMs. Specifically, oscillatory shearing tests, including linear viscoelastic region (LVR) and time-dependent evaluations, were conducted to characterize the initial stiffness and structural build-up of the fresh mortar. Secondly, the mechanical strength and anisotropic coefficients of the printed components were comprehensively evaluated up to 28 d. To explore possible physicochemical interactions, a separate-hydration test, together with zeta potential measurements and SEM, was conducted to examine possible electrostatic adsorption and the subsequent nucleation process. In addition, the partial replacement of WPC by WL and SF was also evaluated in terms of carbon emissions per unit mechanical strength. The study aims to provide a strategy for regulating printability, alleviating mechanical anisotropy while reducing embodied carbon through partial replacement of WPC.

2. Materials and methods

2.1. Raw materials and preparation procedure

In this study, the white Portland cement (WPC, 42.5 grade, Albo Co., Ltd), needle-shaped WL (Haiyang Co., Ltd) and spherical SF (Shandong Pengcheng Co., Ltd) constituted the cementitious components of the compositions. The particle size distribution, morphologies and chemical compositions of these materials are presented in Fig. 1, Fig. 2 and Table 1, respectively. The median particle sizes (D_{50}) of WPC, SF and WL were 10.39 μm , 1.78 μm and 8.61 μm , respectively. Quartz sand with a fineness of 0.15 mm, provided by Hongrun Co. Ltd, was used as the aggregate. Water reducing admixture (WRA, Shandong Building Science Academy Co., Ltd) and hydroxypropyl methyl cellulose (100,000 mPa·s, HPMC, Shandong Heda Macklin Co., Ltd) were added to improve the extrudability and buildability (Chen et al., 2020). In addition, calcium formate (CF, Macklin Co., Ltd) served as an accelerating admixture.

Table 2 shows the mix compositions of 3DP-WCMs. Fig. 3 illustrates the specific mixing protocol of 3DP-WCMs. Initially, the dry WPC, WL and SF were blended homogeneously for 60 min. Subsequently, WRA, HPMC and CF were dissolved in water, introduced into the dry materials and mixed at low speed (140 rpm) for 1 min followed by high speed (285 rpm) for 2 min. Finally, the quartz sand was added and mixed at high speed for an additional 1 min.

2.2. Analytical methods

2.2.1. Rheology

The rheological characterization was performed on a rotational rheometer (HAAKE MARS 40, Thermo Fisher Scientific). The fresh mortar was transferred to the cylinder of rheometer after 4 min of initial water contact. The following test commenced at exactly 5 min of hydration age, allowing 1 min for mortar transfer. During the rheology measurements, the fresh mortar was initially pre-sheared at 50 s^{-1} for 1 min and subsequently allowed to rest for another 1 min. Oscillatory stress-sweep tests were then carried out at a constant frequency of 1 Hz by increasing the applied shear stress from 0.1 to 2000 Pa, as shown in Fig. 4(a). The limit of the LVR was defined as the critical stress at which the elastic modulus (G') decreased by 10% from its plateau value (Jiao and De Schutter, 2022; Zheng et al., 2026). This critical stress was recorded as the onset yield stress, representing the initial structural breakdown. The flow point was determined as the stress where G' and

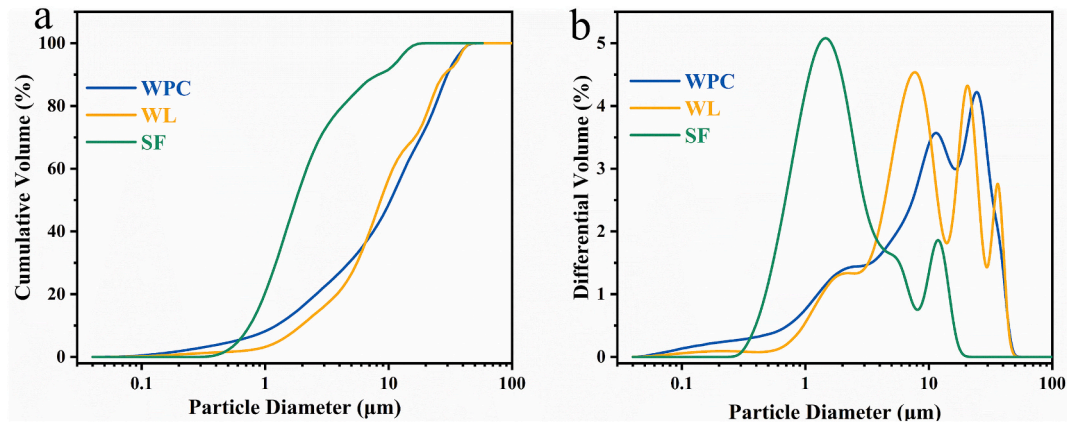


Fig. 1. The particle size distribution of WPC, WL and SF.

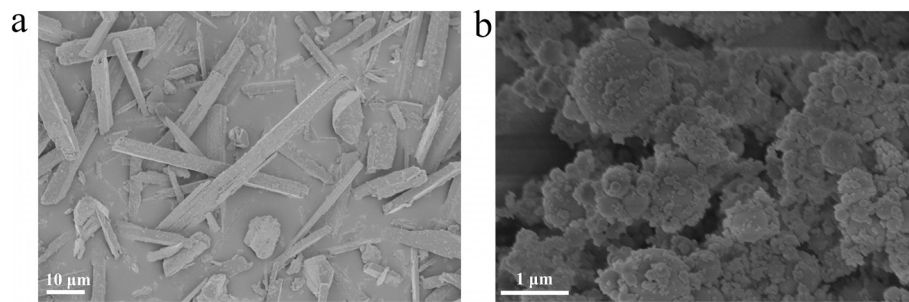


Fig. 2. The SEM images of (a) WL and (b) SF.

Table 1

Chemical compositions of WPC, WL and SF (% by mass).

Components	CaO	SiO ₂	SO ₃	MgO	Al ₂ O ₃	K ₂ O	Na ₂ O	Fe ₂ O ₃	Others	Loss
WPC	63.91	20.57	4.24	3.64	3.36	0.52	0.35	0.18	1.47	1.76
WL	45.79	47.62	0.02	0.61	0.07	0.03	–	0.16	5.7	–
SF	0.03	98.19	–	–	–	–	–	0.17	1.61	–

Table 2

The mix composition of 3DP-WCMs.

Mix design	WPC (wt%)	WL (wt%)	SF (wt%)	Water (wt%) ^a	WRA (wt%) ^a	HPMC (wt%) ^a	CF (wt%) ^a	Quartz sand (wt%) ^a
C100	100	0	0					
C90W6S4	90	6	4					
C80W12S8	80	12	8	40	0.3	0.1	1	30
C70W18S12	70	18	12					
C60W24S16	60	24	16					
C50W30S20	50	30	20					

(C: White Portland cement (WPC); W: wollastonite (WL); S: silica fume (SF); a: added as % by total mass of WPC, WL and SF; the water/cementitious solids (WPC, WL, SF) were controlled at 0.40).

loss modulus (G'') intersected, revealing the transition from solid-like to liquid-like state, as detailed in Fig. 4(b).

Since 5 Pa remained within the LVR of all mixtures, this stress was adopted for the time-sweep measurements. Fig. 4(c) represents the model of elastic modulus over 3600 s using oscillatory time-sweep at a constant frequency of 1 Hz. The resulting time-dependent G' curves plotted in Fig. 4(d) were fitted using a two-term structural build-up equation, where the first term presented the recovery of flocculation or reflocculation after pre-shear, and the second term described the structural build-up of cement paste due to hydration (Ma et al., 2018; Mostafa and Yahia, 2016; Zhao et al., 2023). The equation can be written as follows:

$$G' = c \left(1 + (\lambda - 1) e^{-\frac{t}{\theta}} \right) + G_{\text{rigid}} t \quad (1)$$

where G' and t represent the elastic modulus (Pa) and testing time (s), c is the amplitude of structural flocculation (Pa); G_{rigid} is the structural build-up rate driven by hydration (Pa/s), λ and θ represent the residual structural factor after pre-shear and the characteristic time (s), respectively.

2.2.2. Printability

The 3D printer (HC-3DPRT, Jianyanhuace company, CHN) with a

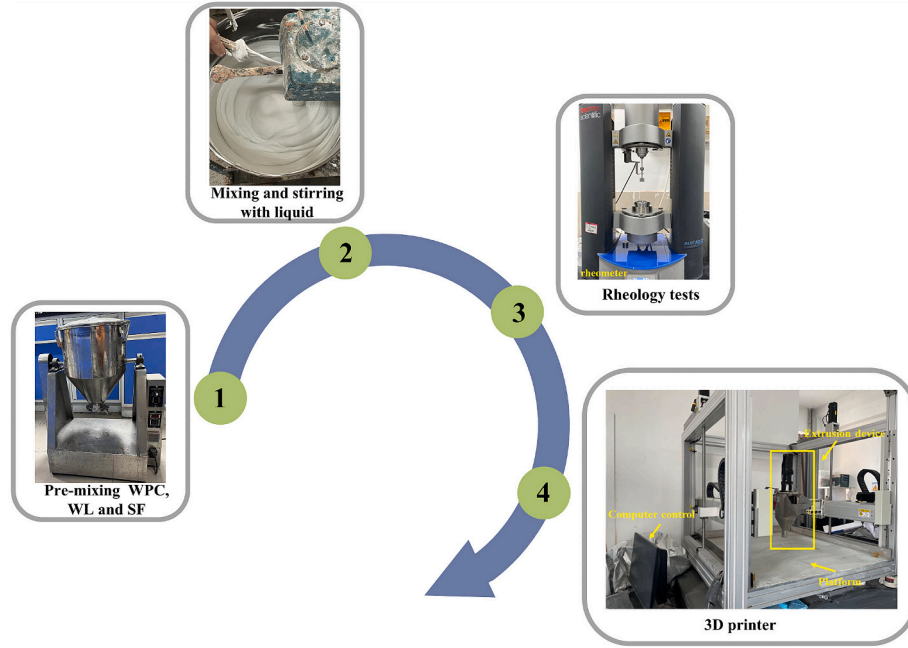


Fig. 3. Schematic diagram of the mixing process for rheological measurements and printability tests.

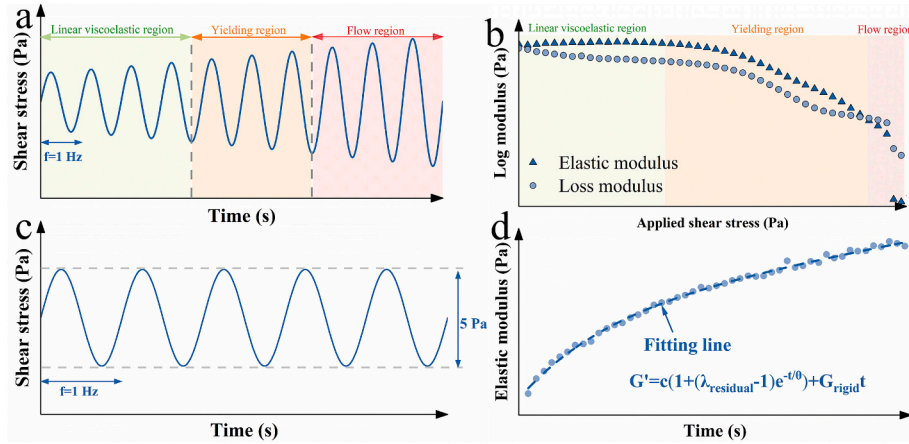


Fig. 4. Schematic illustration of the oscillatory stress-sweep test and determination of the LVR, yield stress and flow point.

build volume of $1.2 \text{ m} \times 1.3 \text{ m} \times 1.4 \text{ m}$ was employed in this study. As modeled in Fig. 5(a), the specific dimensions of the printed components were $380 \text{ mm} \times 220 \text{ mm} \times 52 \text{ mm}$ and $450 \text{ mm} \times 40 \text{ mm} \times 26 \text{ mm}$, respectively. The nozzle diameter and single layer height were set to 20 mm and 13 mm, respectively.

The structural deformation was used to estimate the printability of 3DP-WCMs, which was calculated from the dimensional deviations between the designed model and actual printed component, according to Eq. (2) (Chen et al., 2018):

$$D_s = \left(\frac{l - l_0}{3l_0} + \frac{w - w_0}{3w_0} + \frac{h_0 - h}{3h_0} \right) \times 100\% \quad (2)$$

where D_s represents the structural deformation rate; l_0 , w_0 and h_0 are the length, width and height of designed model; l , w and h are the length, width and height of actual printed component. It was worth noting that the three-dimensional size was recorded at 10 min after the liquid components contacted with solid components.

2.2.3. Mechanical strength

The mechanical strength of 3DP-WCMs was evaluated using printed cut specimens. Concurrently, the printed specimens after curing for 3 d and 28 d were cut into $40 \times 40 \times 160 \text{ mm}^3$ and $40 \times 40 \times 40 \text{ mm}^3$ for testing flexural and compressive strength, respectively. As plotted in Fig. 5(a), it is important to point out that the loading directions are divided into perpendicular (A direction) and parallel (B and C directions) to the direction of nozzle movement due to the anisotropy of 3D printed specimens.

The interface bonding strength of 3DP-WCMs was measured by direct tensile test, following the procedures reported by (He et al., 2022a; Sanjayan et al., 2018). First, 3D printed components were cured for 3 d and 28 d, then cut into samples with 40 mm width. As shown in Fig. 5(b), the upper and lower parallel surfaces of the samples were glued to the metallic brackets using high-strength epoxy resin. The glued assemblies were allowed to cure for exactly 24 h to achieve full hardening of the adhesive layer prior to mechanical testing. The interface bonding strength in A and B directions was measured at a constant loading rate of 0.5 mm/min using an MTS machine. The bonding strength can be calculated by Eq. (3):

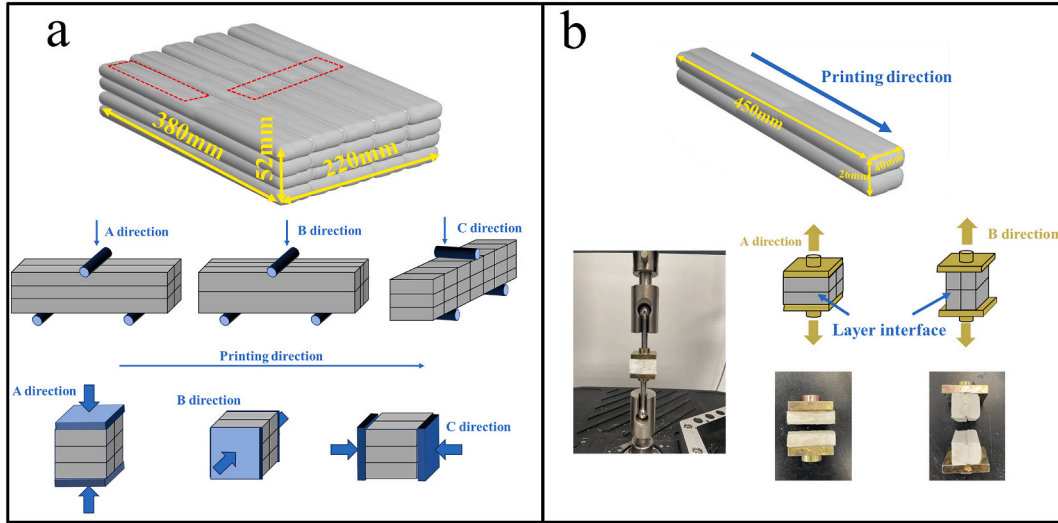


Fig. 5. The printed models with specific designed size and loading directions for (a) compressive and flexural strength test and (b) tensile test.

$$\sigma = \frac{F}{S} \quad (3)$$

where σ is bonding strength (MPa), F is the maximum tensile load (N), S is the cross-sectional area (mm^2).

According to the viewpoint of Ye et al. (Ye et al., 2021), the anisotropic parameter was calculated by Eq. (4) to quantitatively analyze the mechanical anisotropy of 3DP-WCMs. Furthermore, a mechanical anisotropic coefficient $I_{\text{Mechanical}}$ can be calculated using Eq. (5) (Chen et al., 2022):

$$I_{\text{Direction}} = \sqrt{(f_A - f_{\text{Direction}})^2 + (f_B - f_{\text{Direction}})^2 + (f_C - f_{\text{Direction}})^2} \div f_{\text{Direction}} \quad (4)$$

$$I_{\text{Mechanical}} = (I_A + I_B + I_C) \div 3 \quad (5)$$

where f_A , f_B and f_C refer to the mechanical strength of printed specimens under the A, B and C loading directions, respectively; $f_{\text{Direction}}$ is the mechanical strength of printed specimens under a specific loading direction. For example, $f_{\text{Direction}}$ and $I_{\text{Direction}}$ are f_A and I_A when the loading direction is A, representing mechanical anisotropic coefficient in A direction. $I_{\text{mechanical}}$ refers to the anisotropic coefficient of flexural (I_f) and compressive (I_c) strength, respectively.

2.2.4. Microstructural analysis

The interlayer microstructural characteristics of the 3DP-WCMs were evaluated using field-emission scanning electron microscopy (FE-SEM, Carl Zeiss). Specimens containing two adjacent printed layers were sectioned and prepared for observation. To obtain complementary microstructural information across different scales, secondary electron (SE2) mode at a voltage of 5 kV was utilized to characterize the physical interlocking or rupture behavior of the WL. Backscattered electron (BSE) imaging at voltage of 10 kV was used to examine the interfaces, pores and cracks, and the spatial distribution of WL and SF-rich regions based on compositional contrast. InLens imaging at a voltage of 3 kV was designated for high-magnification observation of SF particles and hydration products associated with the WL surfaces.

Mercury intrusion porosimetry (MIP, AutoPore V 9600, Micromeritics Instrument Corporation) was additionally performed to characterize the pore structure of the interlayer-containing regions at 28 d. Small fragments containing the inter-filament boundary and adjacent cementitious matrix were carefully extracted from C100, C80W12S8, and C50W30S20 at comparable locations. To terminate hydration and remove pore water, the specimens were immersed in isopropanol for 7 d,

followed by vacuum drying for 3 d before MIP testing.

2.2.5. Separate-hydration test

To isolate the interaction between WL and SF from the complexity of the cementitious matrix, a separate-hydration experiment was prepared using a PTFE membrane configuration (see Fig. 6). C_3S powder was placed inside the upper membrane to simulate the cement hydration and suspended in deionized water above the lower chamber containing individual WL, individual SF or combined WLSF suspensions. The mass ratio of water: C_3S : WL: SF was set as 50:1:0.1:0.01 (Zhao et al., 2024b; Zheng et al., 2023). During the hydration process, ions released during C_3S hydration freely diffused through the membrane, whereas direct physical mixing between C_3S and the lower suspension was completely avoided. Suspensions were collected at selected ages for pH and zeta potential measurements and dried for SEM observations. In addition, the mobility-to-zeta conversion was performed using the analysis model selected by the instrument software (ZS Xplorer, Auto mode, Malvern Zetasizer Pro). The conductivity values at each hydration age were recorded as well. Three independently prepared suspensions were tested at each hydration age, and the results were reported as mean $\pm$ standard deviation.

2.2.6. Carbon emissions assessment

A simplified embodied-carbon assessment was conducted to evaluate the carbon emissions of the 3DP-WCMs based on materials' carbon

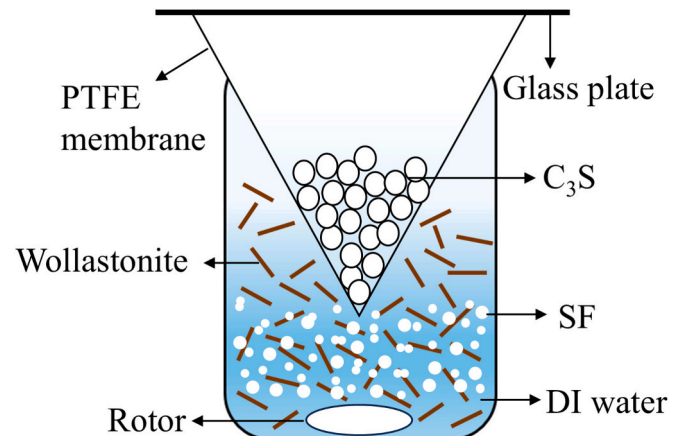


Fig. 6. Diagram of separate-hydration setup of C_3S with WL and SF.

emission factors. The adopted emission factors were derived from published studies with consideration of emissions from raw material acquisition, energy consumption during production and transportation emissions in some datasets (Wang et al., 2026). Therefore, this approach provided a reasonable estimate of the embodied carbon of the constituent materials. Additional specific transportation to the laboratory, mortar mixing, 3D printing, curing and end-of-life were not separately considered (Ye et al., 2023). Table 3 summarizes the carbon emission factors of the WPC, WL, SF and quartz sand. Chemical admixtures were excluded because of their low and identical dosages among all mixtures. The functional unit was defined as 1 m³ of fresh printable mortar and Table 4 presents the constituent contents per 1 m³ of 3DP-WCMs calculated from the measured fresh density.

The embodied carbon and the emissions considering compressive strength at 28 d of each mixture were calculated using Eq. (6) and (7) (Wang et al., 2026).

$$E_C = \sum_i m_i EF_i \quad (6)$$

$$CS_j = \frac{E_C}{S_j} \quad (7)$$

where E_C is the embodied carbon of the three mixtures (kg CO₂-eq/m³), m_i is the mass of constituent i per 1 m³ (kg/m³), EF_i is the corresponding carbon emission factor (kg CO₂-eq/kg), CS_j is the CO₂ emissions per unit strength (kg CO₂-eq/(m³•MPa)) in loading direction j (A, B and C) and S_j is the 28 d compressive strength.

3. Results and discussion

3.1. Printing performance of 3DP-WCMs

3DPC requires the fresh paste to maintain sufficient flowability during pumping and extrusion while rapidly developing buildability after extrusion (Maierdan et al., 2024). Therefore, the rheological response should be interpreted in relation to the actual printing process.

3.1.1. Rheological behavior and structural build-up

Fig. 7 plotted the oscillatory stress-sweep results of the 3DP-WCMs. In the lower stress region, G' remained nearly constant and significantly higher than G'' , indicating an intact viscoelastic particle network. As the applied stress increased, G' began to deviate from its plateau, marking the limit of the LVR and the onset of structural breakdown. At higher stress levels, the flow point occurred where G' intersected G'' , corresponding to the transition from a solid-like to a liquid-like response (Irani et al., 2019). Thus, the end of LVR and the flow point describe two different stages: the former indicates the initial loss of network integrity, whereas the latter reflects the total collapse of the load-bearing structure. Compared with C100, the incorporation of WL and SF significantly enlarged the LVR and postponed the appearance of the flow point, demonstrating the formation of a much stronger particle network (Zhao et al., 2025). The structural yielding behavior was attributed to the combined effects of WL and SF: needle-shaped WL promoted microscale interlocking, whereas ultrafine SF filled interparticle spaces and increased particle-surface interactions. At higher replacement levels, however, these physical contributions were accompanied by increasing clinker dilution (Zhao et al., 2023).

To achieve adequate buildability, the deposited filaments must rapidly develop sufficient stiffness to resist structural deformation

Table 4

Calculated constituent contents per 1 m³ of 3DP-WCMs.

Mixture	WPC	WL	SF	Quartz sand	Water
C100	1216.22	0	0	364.86	486.49
C80W12S8	1023.62	153.54	102.36	383.56	511.81
C50W30S20	651.16	390.69	260.46	390.69	520.92

induced by the self-weight of subsequently printed layers (Deng et al., 2022; Wang et al., 2021). The dynamic evolution of the elastic modulus is therefore employed to quantitatively evaluate the structural stability of the fresh paste (Mostafa and Yahia, 2016). Fig. 8 manifested the time-dependent evolution of G' over 3600 s. All mixtures exhibited a nonlinear increase in G' with time, revealing the continuous recovery of the particle network and structural build-up after structural breakdown. Notably, the group modified by WL and SF presented higher G' values than the control group, indicating enhanced rigidity of the fresh mortar.

Table 5 summarized the specific parameter values derived by fitting the G' curves with structural build-up model. As the WPC replacement level increased, both the structural flocculation amplitude (c) and the residual structural factor (λ) exhibited substantial increments. Specifically, the values of c rose dramatically from 7.32×10^5 Pa of C100 to 3.11×10^6 Pa for C50W30S20, while λ concurrently elevated from 0.22 to 0.86. This pronounced structural enhancement is primarily attributed to the intensified physical interlocking among the high-aspect-ratio needle-shaped WL particles, coupled with an increased solid volume fraction and higher matrix stiffness resulting from the pronounced water adsorption of both WL and SF. Conversely, the chemical structural build-up rate (G_{rigid}), which is dominated by early-age hydration kinetics, followed an overall declining trend upon the incorporation of WL and SF. The value of G_{rigid} decreased from 195.26 Pa/s of C100 to 103.49 Pa/s of C50W30S20. This deceleration is directly ascribed to the suppressed early hydration kinetics caused by the severe dilution of the WPC clinker under excessive replacement, simultaneously weakening the hydration-driven structural build-up.

3.1.2. Printability and shape retention

Stable and continuous filament deposition plays a fundamental role in minimizing the mechanical anisotropy of 3D printed components, as it directly determines the effective contact area and structural integrity between adjacent layers and filaments (Zhang et al., 2021). As depicted in Fig. 9, the printability of 3DP-WCMs was evaluated by quantifying the height and structural deformation rate of 3D printed components. All 3D printed samples exhibited acceptable printability and shape retention, with only marginal variations in their structural deformation rates. The binary incorporation of WL and SF generally enhanced the fresh shape stability, which is highly consistent with the elevated G' . Crucially, all mixtures achieved high geometric accuracy, with printed heights closely reaching the target design value. Among them, C80W12S8 exhibited an optimized printing behavior, showing a relatively low structural deformation of 5.8% accompanied by stable filament deposition. For C80W12S8 and C50W30S20, the extrusion parameters (screw rotation speed and nozzle movement speed) were adaptively matched to thoroughly compensate for the higher structural resistance of the mortar (Chen et al., 2024), as shown in Table 6. This minor adjustment helped maintain comparable filament geometry and uninterrupted continuous deposition across all tested groups. Consequently, C50W30S20 maintained a highly stable and comparable structural accuracy, with its structural deformation rate remaining at a low level of 10.2%.

Table 3

Carbon emission factors (kg CO₂-eq/kg) of the materials.

Components	WPC	WL	SF	Quartz sand	Water
Carbon emission factor	1.1	0.0065	0.014	0.005	0.00035
Reference	(Anderson and Moncaster, 2020)	(Wang et al., 2026)	(Shahmansouri et al., 2025)	(Ye et al., 2023)	(Shahmansouri et al., 2025)

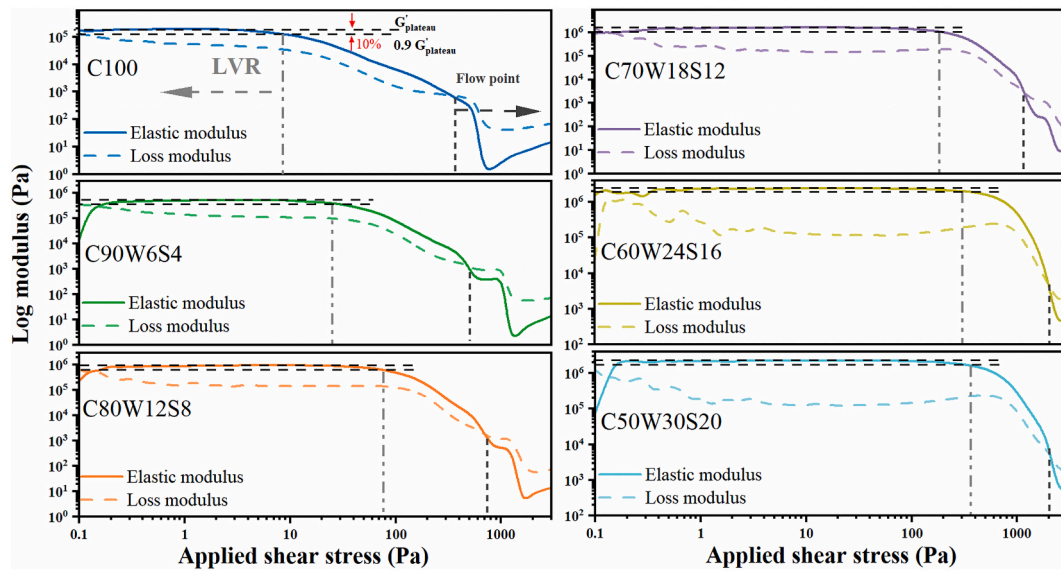


Fig. 7. The evolution of elastic modulus (G') and loss modulus (G'') of 3DP-WCMs by oscillatory stress-sweep response and the derived onset yield stress and flow point.

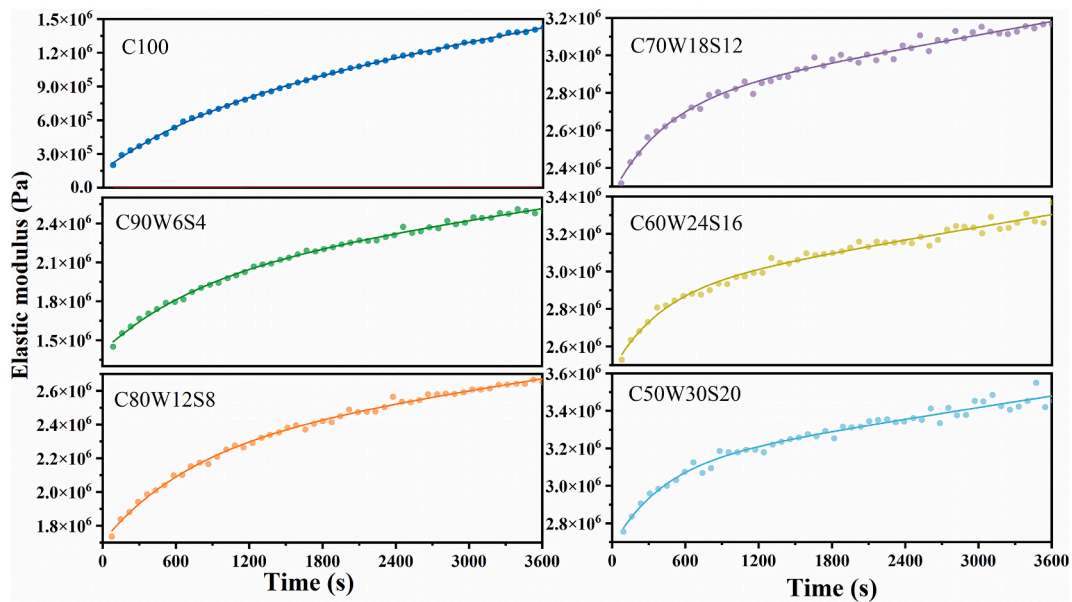


Fig. 8. The elastic modulus evaluation of 3DP-WCMs measured by oscillatory time-sweep.

Table 5

The fitting data of elastic modulus over time.

Model	$G' = c \left(1 + (\lambda - 1)e^{-\frac{t}{\theta}} \right) + G_{rigid}t$					
Plot	C100	C90W6S4	C80W12S8	C70W18S12	C60W24S16	C50W30S20
c	7.32×10^5	2.01×10^6	2.28×10^6	2.75×10^6	2.90×10^6	3.11×10^6
λ	0.22	0.70	0.75	0.82	0.85	0.86
θ	953.73	814.82	744.10	438.00	402.16	406.06
G_{rigid}	195.26	141.44	110.44	119.14	113.42	103.49
R^2	0.99	0.99	0.99	0.98	0.98	0.97

3.2. Mechanical performance of 3DP-WCMs

3.2.1. Mechanical strength and anisotropy of 3D printed samples

Fig. 10 showed the flexural and compressive strength of 3D printed

components loaded in the A, B and C directions at 3 d and 28 d. At 3 d, C80W12S8 exhibited slightly higher flexural strength in the A and C directions than C100. This suggested that the localized physical bridging and interlocking provided by needle-shaped WL partly compensated for

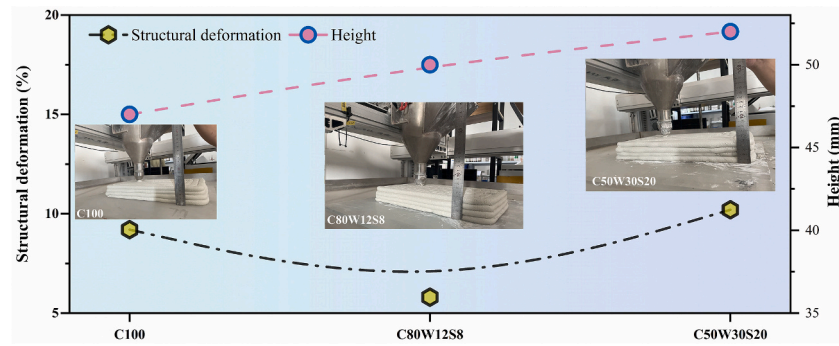


Fig. 9. The (a) 3D printed model for printability assessment and (b) height and (c) structural deformation of 3D printed components.

Table 6

Printing parameters of 3DP-WCMs.

Mixtures	Rotational speed of screw (rpm)	Movement speed of nozzle (mm/s)
C100	35	50
C80W12S8	37	50
C50W30S20	40	40

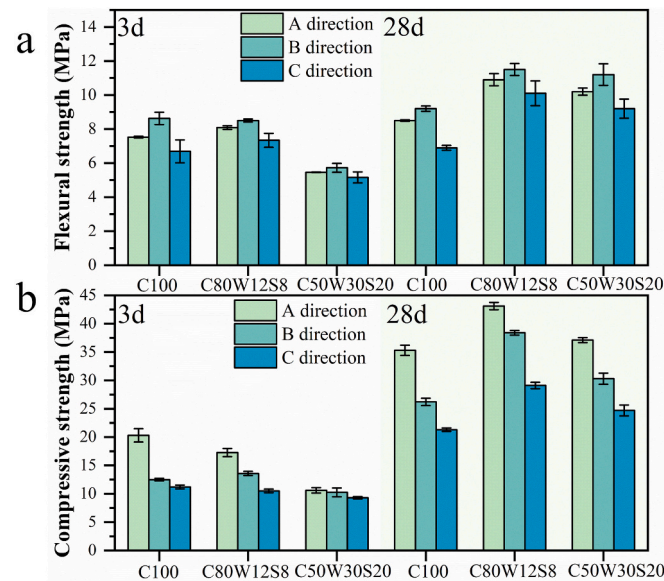


Fig. 10. (a) The flexural strength and (b) compressive strength of 3D printed components loaded in A, B and C directions at 3 d and 28 d curing ages.

the reduction in cement clinker at a moderate replacement level. However, as the replacement level escalated further, the mechanical strength at 3 d decreased markedly. For C50W30S20, its compressive strength in the A direction dropped by 47.73% compared to C100. The physical reinforcement of the WL was insufficient to offset the severe clinker dilution effect and retarded early hydration kinetics under a constant water-to-binder ratio (Abdel Wahab et al., 2017; He et al., 2022b). Moreover, Yang et al. (Yang et al., 2022) proposed that the introduction of fibers also increased the air void entrapment, resulting in a more porous matrix with weaker strength, especially at early ages. These results highlight the need to balance the beneficial effects of WL and SF incorporation against the loss of early-age strength at high replacement levels.

In contrast, the 28 d mechanical properties manifested a distinct evolutionary trend. Among them, C80W12S8 exhibited the highest 28 d mechanical strength among the printed mixtures, outperforming C100

in all loading directions. Specifically, the compressive strength of C80W12S8 reached a peak value of 43.1 MPa in the A direction, while the corresponding strengths in the B and C directions reached 38.40 and 29.10 MPa, respectively. Meanwhile, its flexural strength at 28 d was also the highest, reaching 10.10 MPa and representing a 46.4% increment over C100 in the C direction. This recovery was primarily attributed to the progressive pozzolanic reaction of SF, continued hydration-product development and improved interfacial microstructural continuity (Hamada et al., 2023; He et al., 2022b), which successfully unlocked the maximum fiber-bridging efficiency of the WL with a more developed C-S-H hydration products network. Notably, a similar late-stage surge was also observed in C50W30S20 where its flexural strength in the C direction increased by 78.3% relative to its 3 d flexural strength, while its compressive strength in the A direction also exceeded that of C100.

Detailed analysis of Fig. 10(a) revealed that the flexural strength of the 3D printed components was highest in the B direction, followed by the A direction, and minimized in the C direction. When loaded in the C direction, the longitudinal tensile stress generated at the bottom propagated perpendicular to the vertical intra-layer parallel paths. Because the vertical boundaries between adjacent paths are inherently the weakest zones within the printed architecture, failure cracks can preferentially propagate along these interfaces with minimal energy consumption, resulting in the lowest flexural capacity. Furthermore, the strength discrepancy between the A and B directions is directly related to the total number of horizontal interfaces embedded within the tensile failure zone (Mechtcherine et al., 2019; van den Heever et al., 2022). Along the cracking path, the stress field in the B direction only encounters a single horizontal interlayer boundary, whereas the specimens in the A direction contained two horizontal interlayer boundaries within the tensile region (Fig. 5). The high frequency of these horizontal interfaces increased the probability of localized interfacial shearing and multi-layer delamination from the bottom upwards, thereby compromising its flexural strength compared to the B direction.

This structural joint configuration fundamentally governed the compressive strength as well, where the compressive strength exhibited superior values in the A direction compared to the B and C directions, perfectly aligning with previous findings (Yu et al., 2023). When compressed in the A direction, the interlayer regions were subjected primarily to compressive confinement, which suppressed crack opening and promoted uniform stress transfer. Conversely, the B and C directions involved load transmission parallel to the layer interfaces, making the mechanical response highly sensitive to interfacial defects generated during the printing process. Among them, the C direction demonstrated the lowest compressive strength because the load had to be transferred across multiple neighboring filaments, where insufficient filament fusion and interfacial porosity served as preferential crack propagation paths.

Crucially, the incorporation of WL and SF reduced these interfacial defects and narrowed the strength gaps among the three loading

directions. As displayed in Table 7, the calculated mechanical anisotropic coefficients for both flexural (I_f) and compressive (I_c) strength decreased significantly with the raised dosage of WL and SF, quantifying the alleviated mechanical anisotropy of 3DP-WCMs. To be more specific, the I_c value of C50W30S20 at 3 d dropped sharply to 0.13, representing an 80.9% reduction compared to C100. As the hydration proceeded to 28 d, although the strength development slightly widened the directional discrepancies, the specimens with WL and SF retained their superior uniformity over the control group. At 28 d, the I_f and I_c values of the optimal C80W12S8 were reduced to 0.13 and 0.40, respectively. These results suggested that WL-related physical bridging, together with SF-related interfacial microstructural development, contributed to reducing directional mechanical differences.

3.2.2. Bonding strength of 3D printed samples

Fig. 11 illustrated the bonding strength of the 3DP-WCMs determined via direct tensile test at 3 d and 28 d. At both 3 d and 28 d, the bonding strength peaked for C80W12S8. Specifically, the 28 d tensile capacity of C80W12S8 reached 4.17 MPa and 3.16 MPa in the A and B directions, representing a noticeable increment of 40.4% and 79.8% relative to C100, respectively. The prominent improvement can be attributed to the physical bridging of the needle-shaped WL (He et al., 2020) and filling and hydration products development of SF particles at the interfaces. In addition, gravitational forces in the A direction also exerted additional pressure on the horizontal layers during deposition, consistently facilitating a higher bonding strength in the A direction (Wang et al., 2024). In contrast, C50W30S20 with a high replacement level displayed a distinct age-dependent performance. At 3 d, its bonding strength fell below that of C100 due to the clinker dilution that limited early hydration products formation at the interface. However, at 28 d, C50W30S20 experienced a substantial recovery in comparison with C100, increased by 20.5% and 45.5% in the A and B directions, respectively. Notably, the enhancement was more pronounced in the weaker B direction.

3.3. Microstructural mechanism of alleviated mechanical anisotropy

3.3.1. Separate-hydration evidence for WL-SF interaction

To further clarify the underlying mechanism governing the 3DP-WCMs with WL and SF, a separate-hydration experiment was established to isolate the physicochemical interactions between WL and SF from the complex cementitious matrix. A simplified C₃S hydration environment was utilized to simulate the ionic evolution of cement hydration process. To characterize the chemical environment during the electrokinetic measurements, the electrical conductivity and pH of the WL, SF and WLSF suspensions were monitored at specific ages, as plotted in Fig. 12. The conductivity increased progressively with hydration time, peaking at 3.1–3.5 mS/cm at 24 h. This increase reflected the continuous release and accumulation of mobile ionic species associated with C₃S dissolution and hydration. The differences in conductivity among the three suspensions were considered reasonable because WL and SF exhibited different dissolution, ion-adsorption and surface interaction behaviors, altering the concentration of mobile ions in the

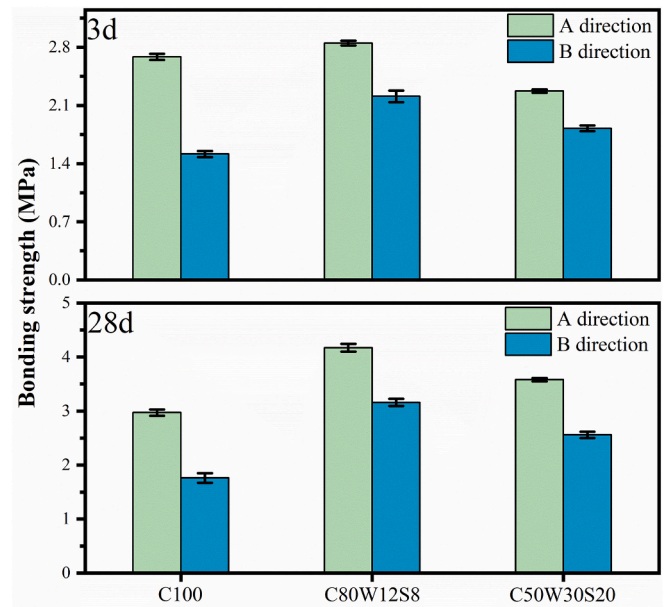


Fig. 11. The bonding strength of 3D printed components loaded in A and B directions at 3 d and 28 d curing ages.

liquid. Meanwhile, the initial pH values of the three suspensions in DI water differed noticeably, with WL, SF and WLSF exhibiting average values of 9.54, 7.06 and 8.91, respectively. The different initial pH reflected the distinct intrinsic dissolution and surface acid-base behaviors of WL and SF in DI water (Tiwari et al., 2023). After exposure to the C₃S hydration, the pH values rapidly converged to nearly 11.5 at 0.5 h and gradually increased to 12.3–12.4 at 24 h. The small difference suggested that their distinct zeta-potential evolution could not be attributed solely to differences in bulk alkalinity.

Against the evolving ionic background, Fig. 13 showed the zeta potential evolution of individual WL, individual SF and composite WLSF suspensions, at selected ages together with corresponding SEM images. Initially (at 0 h), both individual WL and SF particles exhibited negative surface charges in DI water, indicating that direct interparticle contact between the two particles was limited by electrostatic repulsion in the early stage. The SEM images of WLSF at 0 h showed only a limited number of dispersed SF particles on the WL surfaces, providing a reference for comparison. As hydration proceeded, the continuous dissolution of C₃S released Ca²⁺ and silicate ions across the PTFE membrane, triggering progressive charge screening and surface ion adsorption. Consequently, the zeta potential shifted toward less negative or positive charges. Notably, the charge reversal rate of the individual SF suspension was faster than that of individual WL due to its higher specific area and density of silanol groups. At 0.5 h, the zeta potential of SF inverted to a positive value of +4.1 mV, whereas individual WL remained negative at −11.6 mV. This charge divergence created conditions favorable for electrostatic association between positively charged SF and negatively charged WL surfaces. The corresponding SEM images at 0.5 h showed numerous spherical SF particles associated with the surfaces of needle-shaped WL, providing possible morphological evidence consistent with hetero-aggregation. Concurrently, the progressive increase in conductivity indicated an increasingly ion-rich environment, which was expected to modify the electrical double layer and interparticle electrostatic interactions. Under these conditions, the relative zeta-potential evolution, together with the SEM observations, was consistent with enhanced WL and SF association.

In the range of 0.5 h to 6 h, an accelerated charge reversal was observed in the composite WLSF suspension, suggesting intensified interparticle adsorption. Such particle association may facilitate localized ion accumulation and subsequent hydration products development

Table 7

Mechanical anisotropic coefficients for 3DP-WCMs at 3 d and 28 d curing ages.

Mixture	3 d flexural strength				3 d compressive strength			
	I_A	I_B	I_C	I_f	I_A	I_B	I_C	I_c
C100	0.18	0.26	0.32	0.25	0.59	0.63	0.82	0.68
C80W12S8	0.11	0.15	0.19	0.15	0.45	0.35	0.71	0.50
C50W30S20	0.07	0.11	0.12	0.10	0.13	0.10	0.17	0.13
Mixture	28 d flexural strength				28 d compressive strength			
	I_A	I_B	I_C	I_f	I_A	I_B	I_C	I_c
C100	0.21	0.26	0.41	0.29	0.47	0.39	0.70	0.52
C80W12S8	0.09	0.13	0.16	0.13	0.34	0.27	0.58	0.40
C50W30S20	0.14	0.20	0.24	0.19	0.38	0.29	0.55	0.41

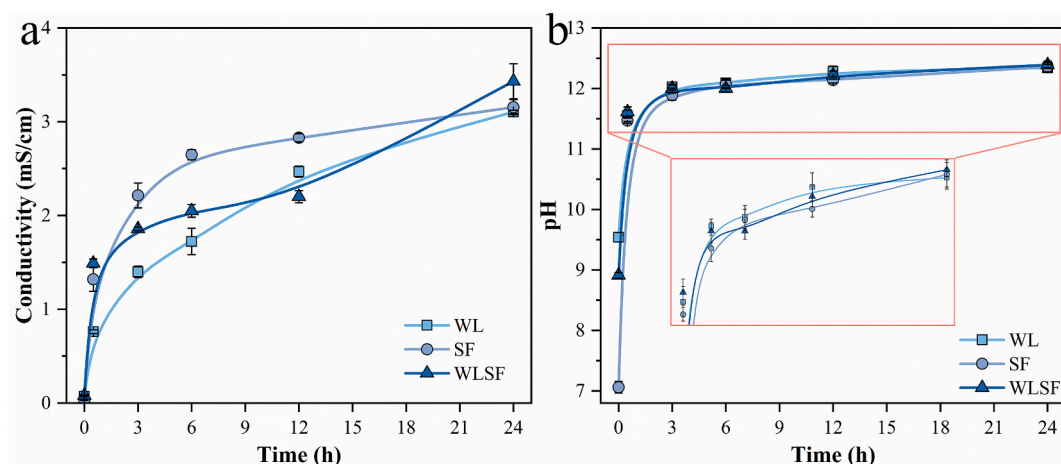


Fig. 12. Conductivity and pH of the individual WL, individual SF and WLSF suspensions during the separate-hydration process.

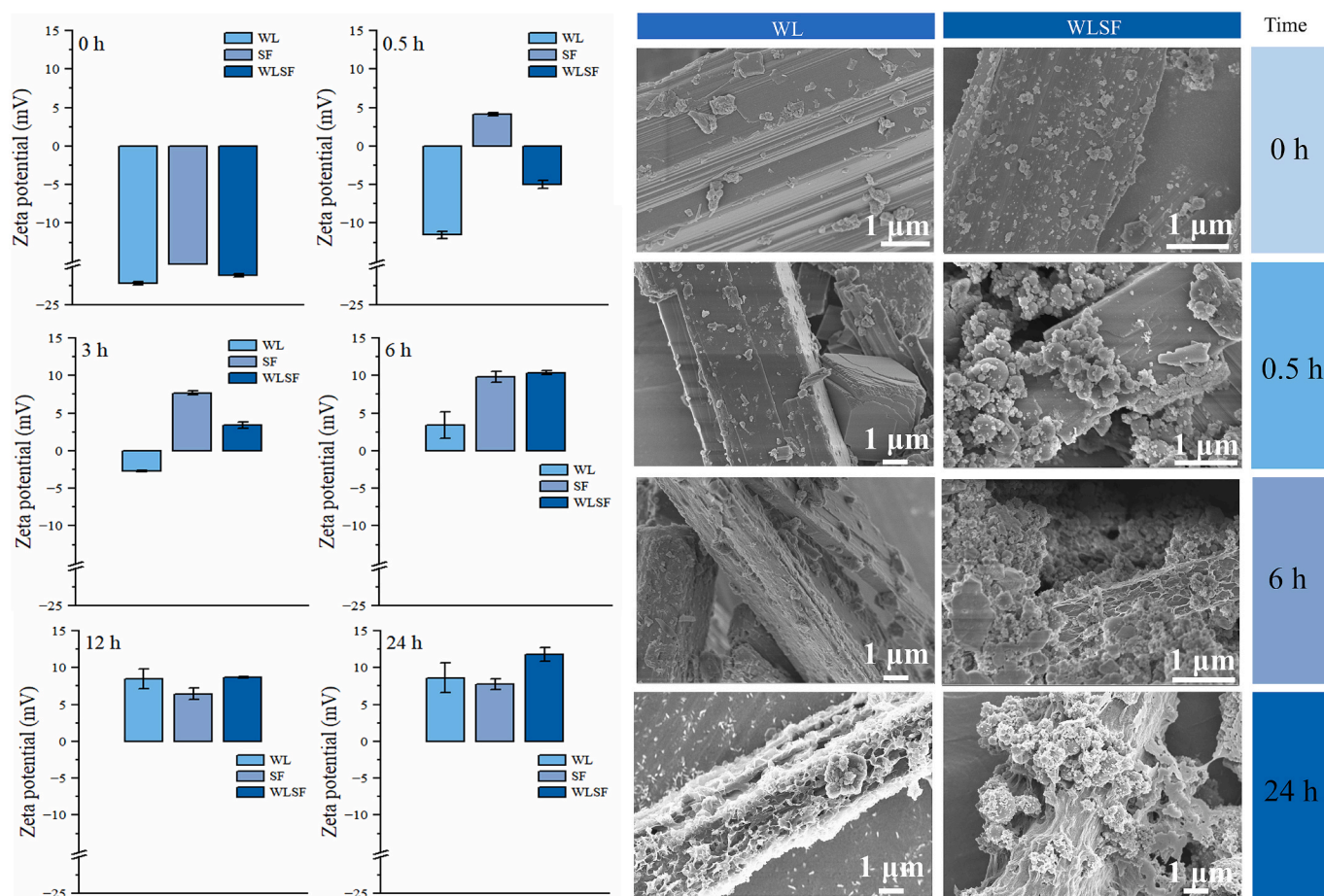


Fig. 13. Zeta potential evolution of individual WL, individual SF and WLSF suspensions during the separate-hydration process, with representative SEM images at selected ages.

(Qiao et al., 2025). As evidenced in the 6 h SEM images, more SF-rich clusters appeared on or around WL, favoring the growth of highly dense, ultrafine clusters of nanostructured C-S-H. In contrast, no significant C-S-H gel was observed in the individual WL group, maintaining a relatively smooth crystal surface. By 24 h, a relatively loose and foamy-like C-S-H layer was formed in the individual WL group. Consequently, the separate-hydration results suggest that early-stage electrostatic interactions between WL and SF may contribute to their

association and the subsequent development of the interfacial hydration network. However, in the actual printable mortar, the presence of WRA and HPMC, together with the complex pore-solution chemistry, may substantially modify particle surface charge and adsorption behavior. In particular, sulfate and aluminate species present in WPC are absent from the simplified C_3S model and may further influence the surface charge evolution and interparticle interactions. Therefore, the present C_3S model is intended to isolate the Ca^{2+} -initiated interaction between WL

and SF rather than to reproduce the complete electrochemical environment of WPC. Accordingly, electrostatic attraction should be regarded as one possible contributor to the observed association between WL and SF rather than the sole governing mechanism.

3.3.2. Formation of WL-SF interfacial network in cement paste

The separate-hydration results provided a mechanistic basis for interpreting the microstructure of the printed cementitious matrix. In the cement paste, WL can act as a micro-scale rigid skeleton, while SF can fill interparticle voids and accumulate around WL surfaces. This coupled incorporation of WL and SF produced a hierarchical particle network composed of rigid WL bridges, fine SF-rich particles and early-age hydration products. SEM-EDS results of the printed samples (C50W30S20) at 3 d were discussed in Fig. 14 to examine local WL and SF associations and hydration products development. As shown in Fig. 14(a) and (b), a large number of high-aspect-ratio WL particles were embedded within the cementitious matrix and clear evidence of WL rupture was detected across the fractured topology. This structural fracture mode suggested substantial mechanical interlocking and load transfer (Liu et al., 2025). Furthermore, nanoscale interactions were observed in the local ITZ, as identified in Fig. 14(c) and (d). Spherical SF particles were observed to associate with the WL surfaces together with surrounding hydration products, which were consistent with possible filling and nucleation role. EDS analysis further provided complementary evidence of hydration products. Specifically, Spot 1 (located on the SF particle) exhibited a relatively high Ca/Si ratio of approximately 2.02, indicating that the particle surface was enriched with Ca-rich hydration products during early hydration. In contrast, the Ca/Si ratios of Spot 2 and Spot 3, located within the dense inter-filament networks between WL and SF, decreased to 1.05 and 0.98, respectively. The close association between WL and SF observed in the actual cementitious matrix motivated the hypothesis that ion-mediated electrostatic interactions may contribute to their interparticle association. Combined with the zeta potential evolution, these results suggest that the early electrostatic attraction between positively charged SF and negatively charged WL promotes preferential interparticle association, which subsequently facilitates localized hydration-product deposition and the development of a continuous WL-SF interfacial network (Lapeyre and Kumar, 2018).

Furthermore, the interfacial regions between two layers were observed using SE2 and BSE signal, as shown in Fig. 15. Specifically, a distinct interface between layers can be clearly identified in C100,

together with visible pores and cracks along the boundary. In contrast, the interfaces in C80W12S8 and C50W30S20 became much less distinguishable, with only scattered pores remaining, while the WL particles were observed in multiple orientations near the interfacial region. Additionally, a considerable amount of unreacted SF was still observed in C50W30S20, indicating that the high SF dosage was not fully consumed during hydration. These observations provide qualitative evidence consistent with the process-induced accumulation of WL and SF particles near filament boundaries. Nevertheless, the images do not quantitatively determine the local particle fraction or orientation. Therefore, the specific migration process requires further quantitative verification in the future.

3.3.3. Pore structure characteristics of the interlayer region

According to commonly used pore size classifications for cementitious materials, the MIP-accessible pores were grouped into three size ranges: fine pores below 10 nm, small-to-medium capillary pores between 10 and 100 nm, and larger capillary pores above 100 nm (Gan et al., 2025; Li et al., 2026). As shown in Fig. 16, the pore size distribution of the specimens containing interfaces changed markedly with incorporation of WL and SF. The fraction of pores smaller than 10 nm increased from 23% in C100 to 31% in C80W12S8 and 38% in C50W30S20, whereas the proportion of pores between 10 and 100 nm decreased from 39% to 33% and 23%, respectively. These results indicated a progressive redistribution of the small-to-medium capillary pores population toward finer pores. In contrast, the overall fraction of pores larger than 100 nm varied only slightly among the three mixtures. Their fraction decreased slightly from 38% in C100 to 36% in C80W12S8, suggesting that moderate WL and SF incorporation may contribute to a more favorable pore structure through particle filling and hydration-related refinement. When the replacement level was further increased to C50W30S20, the pore fraction (>100 nm) increased again to 39%. This may be associated with the stronger dilution of WPC and the high content of needle-shaped WL and ultrafine SF, which may reduce packing efficiency and leave more unfilled or weakly hydrated regions. The residual SF observed in C50W30S20 by SEM in Fig. 15 further revealed that part of the added SF remained unreacted at the high replacement level. Therefore, excessive WL and SF addition appeared to promote the coexistence of abundant fine pores and larger structural voids rather than continuous pore refinement. Additionally, microstructural heterogeneity may have implications for long-term durability, since the persistence of larger pores could potentially

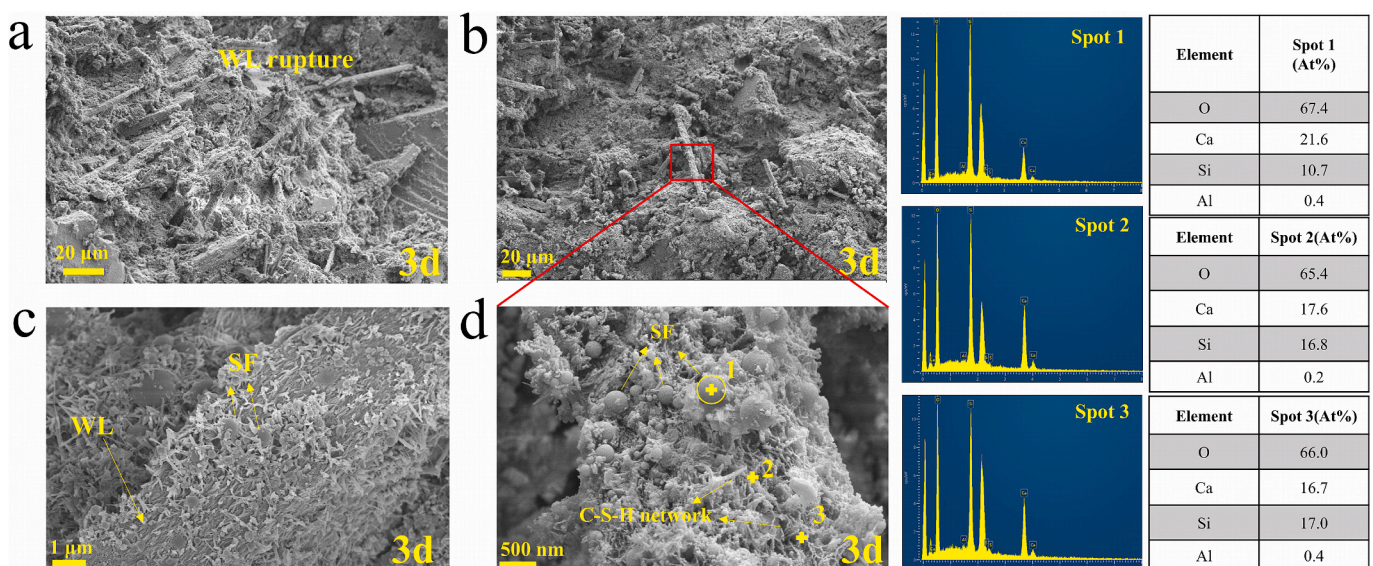


Fig. 14. WL distribution, SF-rich regions and hydration products in the interlayer region of hardened C50W30S20 at 3 d indicated by SE images and EDS.

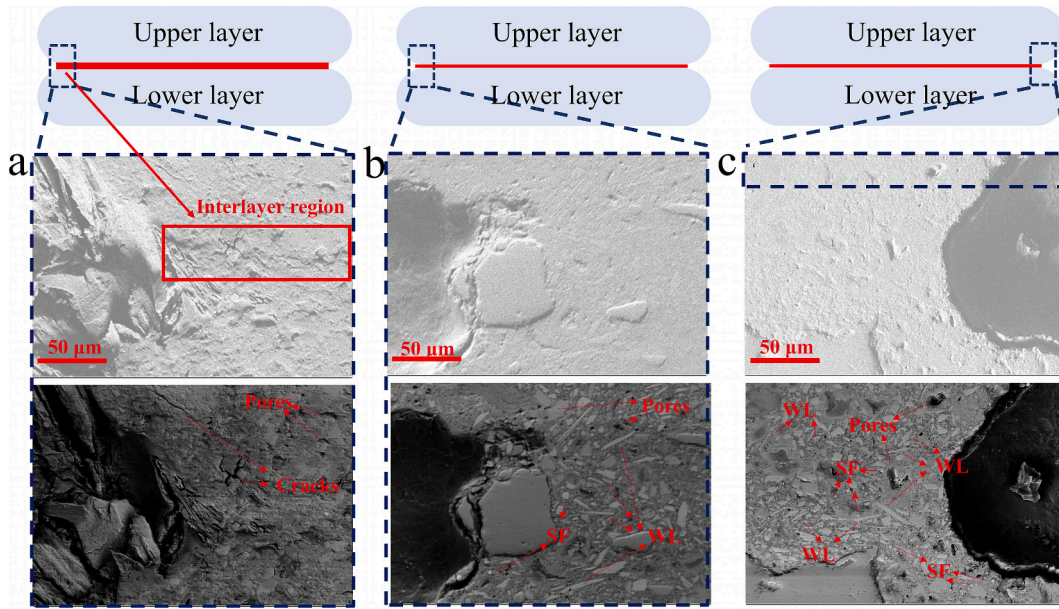


Fig. 15. SE2 (top) and BSE (bottom) observations of the interlayer regions in (a) C100, (b) C80W12S8 and (c) C50W30S20 at 28 d.

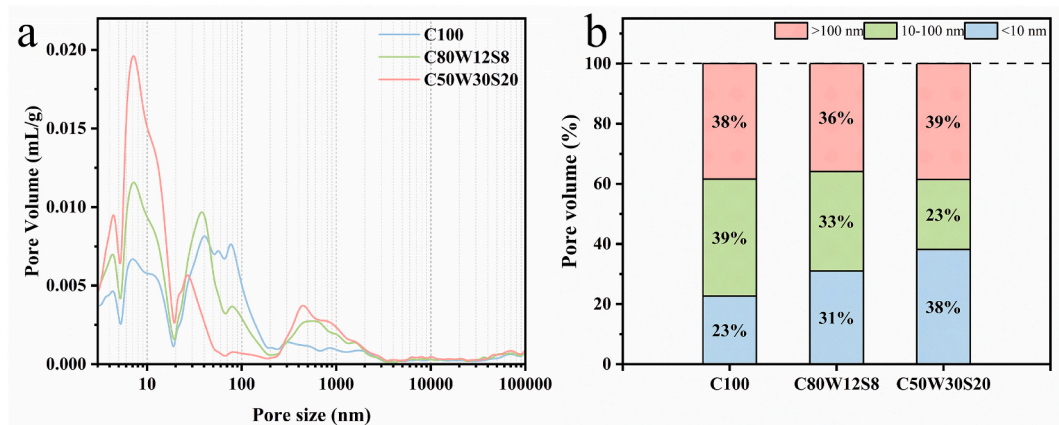


Fig. 16. Pore structure evaluation of C100, C80W12S8 and C50W30S20 at 28 d. (a) differential intrusion curves; (b) quantification of normalized pore-volume fraction in various size ranges.

increase the accessibility of aggressive species.

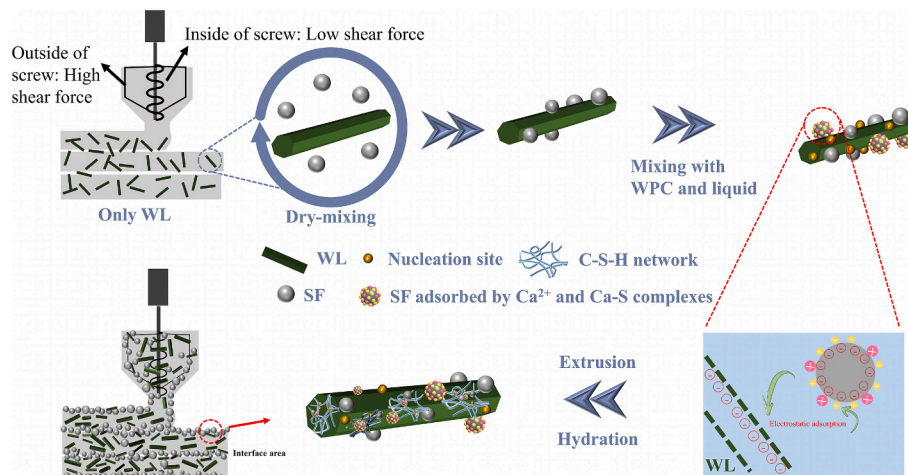


Fig. 17. Proposed mechanism for WL-SF-induced interfacial network formation and mechanical anisotropy reduction in 3DP-WCMs.

3.4. Link between microstructure and mechanical anisotropy of 3D printed samples

The alleviation of mechanical anisotropy in 3DP-WCMs can be interpreted through a multi-scale physicochemical mechanism spanning fresh-state extrusion kinetics and subsequent hardened microstructural evolution, as systematically illustrated in Fig. 17. During extrusion, the rotation of the screw creates a non-uniform shear field within the nozzle, characterized by elevated shear gradients at the periphery and lower shear conditions internally. Previous studies have shown that such shear gradients can drive water and ultrafine particles to migrate toward the nozzle boundaries, forming a particle-rich lubricating layer (Li et al., 2022; Yang et al., 2022). Accordingly, it is reasonable to propose that fine SF and WL particles may also preferentially accumulate near the outer region of the extruded filament. This interpretation is qualitatively supported by the BSE observation in Fig. 15, where WL and SF deposition were observed near the interface of C80W12S8 and C50W30S20. During early hydration, the process-induced particle enrichment may facilitate coupled physical and chemical interactions at the contact zones. The separate-hydration results suggest that the zeta potential trends in the simplified model are consistent with the possible contribution of Ca^{2+} -mediated electrostatic association between negatively charged WL surfaces and SF particles. The SEM observations further indicate that SF adsorption and hydration products develop around WL surfaces, suggesting that WL can provide a rigid micro-scale framework while SF contributes additional filling and hydration-related refinement around the interfacial region.

After hardening, the microstructural changes contribute to the directional mechanical response through several concurrent mechanisms. The high-aspect-ratio WL fibers construct a micro-scale bridging skeleton to bridge the weak zones, while SF-related filling and hydration-product development may improve microstructural continuity around the inter-filament region. Consistent with these observations, MIP results showed a redistribution of small-to-medium capillary pores population toward finer pores after WL and SF addition. The findings suggest that moderate WL and SF incorporation improves interfacial microstructural continuity and resistance to crack propagation, thereby reducing mechanical anisotropic coefficients, whereas excessive replacement introduces competing effects associated with clinker dilution, incomplete SF reaction, and pore heterogeneity. Consequently, C80W12S8 provides the most favorable balance among printability, mechanical strength, and mitigation of mechanical anisotropy within the investigated mixtures.

3.5. Sustainability of 3DP-WCMs

The embodied carbon values of C100, C80W12S8, and C50W30S20 were 1339.81, 1130.49, and 724.64 kg $\text{CO}_2\text{-eq}/\text{m}^3$, respectively, corresponding to 15.6% and 45.9% reductions for C80W12S8 and C50W30S20 compared with C100, as presented in Table 8. This reduction was mainly attributed to the decreased WPC content, since WPC had a substantially higher emission factor than the other constituents. Although the fresh density increased with increasing WL and SF replacement, the reduction in WPC consumption per unit volume remained the dominant factor controlling the overall embodied carbon (Anderson and Moncaster, 2020).

After normalization by the 28 d compressive strength, the carbon-

strength ratio also decreased in all A, B, and C loading directions, indicating that the carbon-reduction benefit was retained after accounting for directional mechanical performance. C80W12S8 exhibited the highest compressive strength and thus provided a favorable balance between mechanical performance and embodied-carbon reduction, whereas C50W30S20 showed the lowest carbon-strength intensity due to its substantially reduced WPC content.

3.6. Limitations and perspectives

Overall, the combined incorporation of WL and SF not only contributed to alleviating the mechanical anisotropy of the 3D printed cementitious composites, but also reduced their embodied carbon and carbon-strength intensity. This indicates that the proposed physicochemical strategy has the potential to simultaneously improve directional mechanical performance and environmental efficiency. However, the 3 d mechanical strength decreased at higher WL and SF replacement levels, indicating a trade-off between early-age strength development and the dilution of the WPC, although the 28 d mechanical performance was improved. The long-term durability of the modified composites also remains to be evaluated.

More importantly, several methodological limitations should also be acknowledged. The separate-hydration setup was designed as a simplified mechanistic model based on the literature, and its composition differed from that of the actual printed mixtures. Therefore, the observed interactions between WL and SF should be interpreted qualitatively rather than quantitatively extrapolated to the 3DP-WCMs. Additionally, the present assessment was limited to a simplified embodied-carbon calculation and did not explicitly consider specific transportation, mixing and printing energy, curing, or other life-cycle stages. In addition, the coupled effects of printing parameters, including printing speed, nozzle geometry, layer interval and nozzle height, on mechanical anisotropy were not systematically examined. Future work will consider a more comprehensive life-cycle assessment incorporating these factors and focus on the interaction among different printing conditions, rheological behavior and directional mechanical performance.

4. Conclusions

In this study, a physicochemical strategy based on the combined incorporation of wollastonite (WL) and silica fume (SF) was investigated to alleviate the mechanical anisotropy of 3D printed white cementitious materials (3DP-WCMs). This study linked rheological regulation with directional mechanical performance and interfacial development. Based on the experimental and mechanistic results, the following main conclusions can be drawn:

- (1) The strategic coupling of WL and SF modified the early structural build-up and viscoelastic properties of fresh 3DP-WCMs to guarantee macro-geometric consistency. Oscillatory rheological tests showed that appropriate addition of WL and SF effectively extended the LVR, delayed the flow point and increased the elastic modulus. The increase in c was consistent with enhanced particle interlocking and structural recovery, whereas the overall decrease in G_{rigid} indicated slower hydration-related structural build-up with increasing WPC replacement.
- (2) The mechanical anisotropy was substantially alleviated by the combined incorporation of WL and SF. For the optimal C80W12S8, the 28 d flexural anisotropy coefficient (I_f) fell by 55.17%, while the compressive anisotropy coefficient (I_c) decreased from 0.52 for C100 to 0.40. In addition, the 28 d flexural strength in the weakest C direction climbed by 46.4% over C100.
- (3) The separate-hydration, zeta potential and microstructural observations support a possible physicochemical mechanism for the

Table 8
Embodied carbon and carbon-strength ratio of 3DP-WCMs.

Mixture	E_c (kg $\text{CO}_2\text{-eq}/\text{m}^3$)	CS_A (kg $\text{CO}_2\text{-eq}/(\text{m}^3 \cdot \text{MPa})$)	CS_B	CS_C
C100	1339.81	37.95	51.14	62.90
C80W12S8	1130.49	26.23	29.44	38.85
C50W30S20	724.64	19.53	23.92	29.34

reduced anisotropy. During extrusion, fine WL and SF particles may preferentially accumulate near filament boundary regions, while Ca^{2+} -mediated electrostatic interactions may contribute to adsorption of SF on WL surfaces. The interactions, together with the nucleation and pozzolanic effects of SF, are proposed to facilitate the development of a more continuous interfacial C-S-H network. The improved interfacial continuity and bridging effects provide a plausible explanation for the reduced mechanical anisotropic coefficients of the 3D printed composites.

- (4) The partial replacement of WPC by WL and SF also decreased the embodied carbon of C80W12S8 and C50W30S20 by 15.6% and 45.9%, respectively, compared with C100. Furthermore, the reductions in carbon emissions per unit strength were even more pronounced, indicating that the proposed strategy can provide environmental benefits while simultaneously improving mechanical strength.

Beyond the specific mixed compositions investigated in this study, the results suggest that coordinated control of particle morphology, interfacial interactions and rheological properties provides a cleaner-material design route for simultaneously alleviating mechanical anisotropy and reducing the carbon emissions of 3D printed cementitious materials. Such a strategy may be applicable to architectural decoration, customized elements and other geometrically complex components owing to their white appearance and compatibility with 3D printing technology.

CRedit authorship contribution statement

Yuan Jin: Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis, Conceptualization. **Zhaoyang Sun:** Software, Methodology. **Mingxu Chen:** Writing – review & editing. **Ming Sun:** Software, Resources. **Xiangming Zhou:** Software, Investigation. **Binmeng Chen:** Writing – review & editing, Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

Data will be made available on request.

References

- Abdel Wahab, M., Abdel Latif, I., Kohail, M., Almasry, A., 2017. The use of Wollastonite to enhance the mechanical properties of mortar mixes. *Constr. Build. Mater.* 152, 304–309.
- Anderson, J., Moncaster, A., 2020. Embodied carbon of concrete in buildings, part 1: analysis of published EPD. *Buildings and Cities* 1 (1), 198–217.
- Arunothayan, A.R., Nematollahi, B., Ranade, R., Bong, S.H., Sanjayan, J.G., Khayat, K.H., 2021. Fiber orientation effects on ultra-high performance concrete formed by 3D printing. *Cem. Concr. Res.* 143, 106384.
- Ateeq, M., Shafique, M., Azam, A., Rafiq, M., 2023. A review of 3D printing of the recycled carbon fiber reinforced polymer composites: processing, potential, and perspectives. *J. Mater. Res. Technol.* 26, 2291–2309.
- Bong, S.H., Nematollahi, B., Xia, M., Ghaffar, S.H., Pan, J., Dai, J.-G., 2022. Properties of additively manufactured geopolymer incorporating mineral wollastonite microfibers. *Constr. Build. Mater.* 331.
- Chen, M., Li, L., Zheng, Y., Zhao, P., Lu, L., Cheng, X., 2018. Rheological and mechanical properties of admixtures modified 3D printing sulphoaluminate cementitious materials. *Constr. Build. Mater.* 189, 601–611.
- Chen, B., Qiao, G., Hou, D., Wang, M., Li, Z., 2020. Cement-based material modified by in-situ polymerization: from experiments to molecular dynamics investigation. *Compos. Part B* 194, 108036.
- Chen, Y., Jia, L., Liu, C., Zhang, Z., Ma, L., Chen, C., Banthia, N., Zhang, Y., 2022. Mechanical anisotropy evolution of 3D-printed alkali-activated materials with different GGBFS/FA combinations. *J. Build. Eng.* 50, 104126.
- Chen, M., Xu, J., Yuan, L., Zhao, P., Li, Q., Lu, L., Wang, L., 2024. Use of creep and recovery protocol to assess the printability of fibre-reinforced 3D printed white Portland cement composites. *Virtual Phys. Prototyp.* 19 (1), e2331201.
- Deng, Z., Jia, Z., Zhang, C., Wang, Z., Jia, L., Ma, L., Wang, X., Zhang, Y., 2022. 3D printing lightweight aggregate concrete prepared with shell-packing-aggregate method - printability, mechanical properties and pore structure. *J. Build. Eng.* 62.
- Dong, E., Jia, Z., Rao, S., Zhao, X., Yu, R., Zhang, Z., Gao, Y., Wang, W., Zhang, Y., Chen, Y., Banthia, N., 2024. Modeling fiber alignment in 3D printed ultra-high-performance concrete based on stereology theory. *Cem. Concr. Compos.* 154, 105786.
- Flatt, R.J., Wangler, T., 2022. On sustainability and digital fabrication with concrete. *Cem. Concr. Res.* 158, 106837.
- Gan, X., Li, Y., Li, W., Li, L., Lu, L., Cheng, X., 2024. Enhancing the mechanical properties and hydration of cement paste by incorporating GO/nano in(OH)3 composite with high specific surface area. *Cem. Concr. Compos.* 149, 105514.
- Gan, X., Zhang, H., Lu, Z., Ma, K., Chen, X., Lu, L., Li, L., 2025. Effect of aluminum dihydrogen phosphate in enhancing mechanical properties and water resistance of magnesium phosphate cement. *Cem. Concr. Res.* 192.
- Hamada, H.M., Abed, F., Binti Katman, H.Y., Humada, A.M., Al Jawahery, M.S., Majidi, A., Yousif, S.T., Thomas, B.S., 2023. Effect of silica fume on the properties of sustainable cement concrete. *J. Mater. Res. Technol.* 24, 8887–8908.
- He, Z., Shen, A., Lyu, Z., Li, Y., Wu, H., Wang, W., 2020. Effect of wollastonite microfibers as cement replacement on the properties of cementitious composites: a review. *Constr. Build. Mater.* 261, 119920.
- He, J., Cheng, C., Zhu, X., Li, X., 2022a. Effect of silica fume on the rheological properties of cement paste with ultra-low water binder ratio. *Materials (Basel)* 15 (2).
- He, L., Li, H., Chow, W.T., Zeng, B., Qian, Y., 2022b. Increasing the interlayer strength of 3D printed concrete with tooth-like interface: an experimental and theoretical investigation. *Mater. Des.* 223, 111117.
- Hou, P., Qian, J., Cheng, X., Shah, S.P., 2015. Effects of the pozzolanic reactivity of nanoSiO₂ on cement-based materials. *Cem. Concr. Compos.* 55, 250–258.
- Irani, M., Razavi, S.M.A., Abdel-Aal, E.M., Hucl, P., Patterson, C.A., 2019. Viscoelastic and textural properties of canary seed starch gels in comparison with wheat starch gel. *Int. J. Biol. Macromol.* 124, 270–281.
- Jiao, D., De Schutter, G., 2022. Insights into the viscoelastic properties of cement paste based on SAOS technique. *Constr. Build. Mater.* 357, 129320.
- Jin, Y., Zhou, X., Chen, M., Zhao, Z., Huang, Y., Zhao, P., Lu, L., 2022. High toughness 3D printed white Portland cement-based materials with glass fiber textile. *Mater. Lett.* 309, 131381.
- Khan, M., McNally, C., 2024. Recent developments on low carbon 3D printing concrete: revolutionizing construction through innovative technology. *Cleaner Mater.* 12, 100251.
- Lapeyre, J., Kumar, A., 2018. Influence of pozzolanic additives on hydration mechanisms of tricalcium silicate. *J. Am. Ceram. Soc.* 101 (8), 3557–3574.
- Le, T.T., Austin, S.A., Lim, S., Buswell, R.A., Law, R., Gibb, A.G.F., Thorpe, T., 2012. Hardened properties of high-performance printing concrete. *Cem. Concr. Res.* 42 (3), 558–566.
- Li, Q., Hou, W., Liang, J., Zhang, C., Li, J., Zhou, Y., Sun, X., 2022. Controlling the anisotropy behaviour of 3D printed ceramic cores: from intralayer particle distribution to interlayer pore evolution. *Addit. Manuf.* 58, 103055.
- Li, Y., Wu, H., Xie, X., Zhang, L., Yuan, P.F., Xie, Y.M., 2024. FloatArch: a cable-supported, unreinforced, and re-assemblable 3D-printed concrete structure designed using multi-material topology optimization. *Addit. Manuf.* 81, 104012.
- Li, Y., Liu, T., Cheng, H., Zhang, C., Xiang, Y., Sun, G., 2026. Void-free internal curing via reactive hydrogel precursor solution: mechanism of water regulation and shrinkage mitigation in AMPS-modified cement-based materials. *Cem. Concr. Res.* 206.
- Liu, Q., Wang, Q., Sun, C., Singh, A., Tam, V.W.Y., Jia, B., 2025. Compressive performance and damage evolution of concrete short columns with shell-filling structure confined by continuous fiber reinforced 3D printed mortar. *Constr. Build. Mater.* 475.
- Ma, S., Qian, Y., Kawashima, S., 2018. Experimental and modeling study on the non-linear structural build-up of fresh cement pastes incorporating viscosity modifying admixtures. *Cem. Concr. Res.* 108, 1–9.
- Maierdan, Y., Armistead, S.J., Mikofsky, R.A., Huang, Q., Ben-Alon, L., Srubar, W.V., Kawashima, S., 2024. Rheology and 3D printing of alginate bio-stabilized earth concrete. *Cem. Concr. Res.* 175, 107380.
- Marchment, T., Sanjayan, J., 2020. Mesh reinforcing method for 3D concrete printing. *Autom. Constr.* 109, 102992.
- Mechtcherine, V., Nerella, V.N., Will, F., Näther, M., Otto, J., Krause, M., 2019. Large-scale digital concrete construction – CONPrint3D concept for on-site, monolithic 3D-printing. *Autom. Constr.* 107, 102933.
- Moelich, G.M., Kruger, P.J., Combrinck, R., 2022. Mitigating early age cracking in 3D printed concrete using fibres, superabsorbent polymers, shrinkage reducing admixtures, B-CSA cement and curing measures. *Cem. Concr. Res.* 159, 106862.

- Mostafa, A.M., Yahia, A., 2016. New approach to assess build-up of cement-based suspensions. *Cem. Concr. Res.* 85, 174–182.
- Nair, N.A., Sairam, V., 2021. Research initiatives on the influence of wollastonite in cement-based construction material- a review. *J. Clean. Prod.* 283, 124665.
- Nguyen-Van, V., Li, S., Liu, J., Nguyen, K., Tran, P., 2023a. Modelling of 3D concrete printing process: a perspective on material and structural simulations. *Addit. Manuf.* 61, 103333.
- Nguyen-Van, V., Liu, J., Li, S., Zhang, G., Nguyen-Xuan, H., Tran, P., 2023b. Modelling of 3D-printed bio-inspired bouligand cementitious structures reinforced with steel fibres. *Eng. Struct.* 274.
- Nguyen-Van, V., Dai, M., Dang, B.-L., 2026. 3D concrete printing for digital construction: materials, process integration, and emerging performance paradigms. *Mater. Emerg. Technol. Sustain.* 02 (01).
- Qiao, G., Zhao, Y., Wang, P., Hou, D., Chen, B., 2025. Unraveling the modulation art of ordered nanomesh-like hydration product on fiber-cement interfaces: insights from fishing net inspiration. *Compos. Part B* 291.
- Robayo Salazar, R., Vargas, A., Martínez, F., Mejía de Gutiérrez, R., 2024. Utilization of powders and fine aggregates from the recycling of construction and demolition waste in the 3D printing of Portland-based cementitious materials. *Cleaner Mater.* 11, 100234.
- Sanjayam, J.G., Nematollahi, B., Xia, M., Marchment, T., 2018. Effect of surface moisture on inter-layer strength of 3D printed concrete. *Constr. Build. Mater.* 172, 468–475.
- Shahmansouri, A.A., Zhang, Z., AzariJafari, H., Shi, X., 2025. Biochar-amended high-strength engineered cementitious composites. *Cem. Concr. Compos.* 164.
- Soliman, A.M., Nehdi, M.L., 2012. Effect of natural Wollastonite microfibers on early-age behavior of UHPC. *J. Mater. Civ. Eng.* 24 (7), 816–824.
- Tiwari, N., Rondinella, F., Satyam, N., Baldo, N., 2023. Experimental and machine learning approach to investigate the mechanical performance of asphalt mixtures with silica fume filler. *Appl. Sci.* 13 (11).
- van den Heever, M., du Plessis, A., Kruger, J., van Zijl, G., 2022. Evaluating the effects of porosity on the mechanical properties of extrusion-based 3D printed concrete. *Cem. Concr. Res.* 153, 106695.
- Wang, L., Ma, G., Liu, T., Buswell, R., Li, Z., 2021. Interlayer reinforcement of 3D printed concrete by the in-process deposition of U-nails. *Cem. Concr. Res.* 148, 106535.
- Wang, Y., Chen, M., Zhang, T., Zhang, M., 2024. Hardening properties and microstructure of 3D printed engineered cementitious composites based on limestone calcined clay cement. *Cem. Concr. Compos.* 152, 105641.
- Wang, Y.-S., Sun, F., Lee, B.-S., Zhang, H., Xuan, M.-Y., Wang, X.-Y., 2026. Development of low-carbon UHPC based on recycled oyster shell powder and wollastonite microfibers: autogenous shrinkage reduction, strength improvement, and carbon emission reduction. *J. Build. Eng.* 127.
- Xu, B., Lothenbach, B., Winnefeld, F., 2020. Influence of wollastonite on hydration and properties of magnesium potassium phosphate cements. *Cem. Concr. Res.* 131, 106012.
- Xu, J., Li, Y., Lu, L., Cheng, X., Li, L., 2023. Strength and durability of marine cement-based mortar modified by colloidal nano-silica with epoxy silane for low CO₂ emission. *J. Clean. Prod.* 382, 135281.
- Xu, C., Yang, R., Wen, R., Dai, Y., Lu, X., Fang, S., Peng, Y., Zhang, Z., Zhou, C., Zeng, Q., Li, Q., Xu, S., 2025. Woven cement slurry. *Adv. Mater.* 37 (10), e2418158.
- Xue, H., Wang, G., Hu, M., Chen, B., 2015. Modification of wollastonite by acid treatment and alkali-induced redeposition for use as papermaking filler. *Powder Technol.* 276, 193–199.
- Yang, R., Zeng, Q., Peng, Y., Wang, H., Wang, Z., 2022. Anomalous matrix and interlayer pore structure of 3D-printed fiber-reinforced cementitious composites. *Cem. Concr. Res.* 157, 106829.
- Yang, R., Xu, C., You, X., Li, X., Xue, H., Peng, Y., Zeng, Q., Wang, Z., 2026. Saddle stitching-enabled interfacial toughening in 3D printed concrete. *Cem. Concr. Compos.* 165.
- Ye, J., Cui, C., Yu, J., Yu, K., Dong, F., 2021. Effect of polyethylene fiber content on workability and mechanical-anisotropic properties of 3D printed ultra-high ductile concrete. *Constr. Build. Mater.* 281, 122586.
- Ye, J., Teng, F., Yu, J., Yu, S., Du, H., Zhang, D., Ruan, S., Weng, Y., 2023. Development of 3D printable engineered cementitious composites with incineration bottom ash (IBA) for sustainable and digital construction. *J. Clean. Prod.* 422.
- Yu, Q., Zhu, B., Li, X., Meng, L., Cai, J., Zhang, Y., Pan, J., 2023. Investigation of the rheological and mechanical properties of 3D printed eco-friendly concrete with steel slag. *J. Build. Eng.* 72, 106621.
- Zelic, J., Rusic, D., Veza, D., Krstulovic, R., 2000. The role of silica fume in the kinetics and mechanisms during the early stage of cement hydration. *Cem. Concr. Res.* 30, 1655–1662.
- Zhang, Y., Nerella, V.N., Krishna, A., Wang, S., Zhang, Y., Mechtcherine, V., Banthia, N., 2021. Mix design concepts for 3D printable concrete: a review. *Cem. Concr. Compos.* 122, 104155.
- Zhang, Y., Zhu, Y., Ren, Q., He, B., Jiang, Z., Van Tittelboom, K., De Schutter, G., 2023. Comparison of printability and mechanical properties of rigid and flexible fiber-reinforced 3D printed cement-based materials. *Constr. Build. Mater.* 400, 132750.
- Zhao, D., Williams, J.M., Park, A.-H.A., Kawashima, S., 2023. Rheology of cement pastes with calcium carbonate polymorphs. *Cem. Concr. Res.* 172, 107214.
- Zhao, Y., Sun, Z., Qiao, G., Fang, X., Chen, B., 2024a. Microstructure evolution of alite in-situ carbonated by aminated mesoporous silica nanoparticles. *Constr. Build. Mater.* 453.
- Zhao, H., Wang, Y., Liu, X., Wang, X., Chen, Z., Lei, Z., Zhou, Y., Singh, A., 2024b. Review on solid wastes incorporated cementitious material using 3D concrete printing technology. *Case Stud. Constr. Mater.* 21.
- Zhao, Z., Cai, X., Chen, F., Gong, Y., Xiao, P., Lu, L., 2025. Effect of wollastonite content on rheology and mechanical properties of 3D printed magnesium potassium phosphate cement-based material of MgO-SiO₂-K₂HPO₄. *Constr. Build. Mater.* 458, 139729.
- Zheng, D., Yang, H., Feng, W., Fang, Y., Cui, H., 2023. Modification mechanism of cellulose nanocrystals in cement. *Cem. Concr. Res.* 165.
- Zheng, T., Ming, X., Tang, J., Gu, Y., Sun, G., 2026. Rheology-dependent multistage performance evolution of foam cement. *Constr. Build. Mater.* 518.
- Zhou, Y., Jiang, D., Sharma, R., Xie, Y.M., Singh, A., 2023. Enhancement of 3D printed cementitious composite by short fibers: a review. *Constr. Build. Mater.* 362.
- Zukoski, C.F., Saville, D.A., 1986. The interpretation of electrokinetic measurements using a dynamic model of the stern layer. *J. Colloid Interface Sci.* 114, 45–53.