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3D Printing of Fiber-Reinforced Calcined Clay-Limestone-Based Cementitious Materials: From Mixture Design to Printability Evaluation

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Abstract: Sustainability and limitations in embedded reinforcement are the main obstacles in digital fabrication with concrete. This study proposed a 3D printable fiber-reinforced calcined clay-limestone-based cementitious material (FR-LC³). The binder systems incorporating calcined clay (CC) and limestone filler (LF) were optimized by determining the flow characteristics and water retention ability of the paste. The effect of fiber volume on the key fresh and mechanical properties of the fiber-reinforced mortars made with the optimized binder was evaluated. A combination of offline assessments and inline printing were employed to investigate the printability of the FR-LC³ with various binder systems and viscosity-modifying admixture (VMA) dosages. The results revealed that the binary system with 20% CC and the ternary system containing 30% CC and 15% LF were highly advantageous, with enhanced packing density, robustness, and water retention ability. Incorporating 2% 6-mm steel fiber contributed to the highest 28-day compressive and flexural strengths and toughness without significantly compromising the fluidity. Finally, the developed FR-LC³ mixtures were successfully printed using an extrusion-based 3D printer. The LF addition in the ternary system decreased the maximum buildable height of a single-wall printed object while reducing the SP/VMA ratio significantly increased the height due to enhanced yield stress and thixotropy.

Keywords: calcined clay-limestone-based cementitious materials; digital fabrication; fiber reinforcement; packing density; printability; rheology



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1. Introduction

Digital fabrication with concrete is an innovative construction method that allows the concrete structure to be built without formwork but only via a 3D printer system. It enables the creation of intricate structures using less material and dramatically lowers costs due to material optimization and being formwork-free [1]. Demonstrated engineering applications encompass the prefabrication of structure components with a high-quality level and the on-site construction of structural elements such as walls and columns for buildings or other infrastructures by means of printing facilities [1,2]. Unlike traditional casting methods, an extrusion-based printer can make concrete materials extrude through the hopper and nozzle, and the layer-wise object can be obtained based on the input design. A material used for printing should maintain sufficient fluidity during extrusion but exhibit a high yield stress to retain its shape and withstand the additional weights resulting from subsequent layers. To this end, a high content of cement or paste is required; moreover, adding aggregate and limiting the maximum aggregate size to achieve better printability without nozzle clogging. The estimated cement content used in 3D concrete printing (3DCP) was reported to be approximately 40% higher than that in conventional cast concrete [3], potentially increasing the environmental impact. One practical solution is substituting cement with traditional supplementary cementitious materials (SCMs) such as fly ash,

slag, and silica fume [4,5]. However, these widely used SCMs are facing a shortage due to regulatory restrictions and changes in industrial manufacturing practices. Consequently, the search for alternative binder sources for use in 3DCP has become urgent. Limestone calcined clay cement (LC³) represents an emerging low-carbon cement option, comprising a mixture that includes calcined clay (CC), limestone filler (LF), and gypsum. This blend allows for a reduction of up to 50% in cement clinker content while maintaining essential performance characteristics [6]. Chen et al. [7] reported that replacing 60% cement content with CC and LF reduced the flowability, extrudability, and printable window but improved strong buildability. The authors also evaluated the extrudability of LC³, incorporating different dosages of viscosity-modifying admixture (VMA) using a ram-type extruder. They reported that a higher dosage of VMA could increase the elongational yield stress, flow consistency, and shear yield stress [8].

Non-reinforced 3D-printed elements typically display brittleness like conventional concrete and possess a relatively low tensile-to-compressive strength ratio, particularly with interlayers after layer-wise deposition [9]. Contemporary reinforcement techniques involve inserting a steel rebar immediately after printing to comply with building codes and structural requirements at a stage when the printed concrete has not yet hardened. This process, however, introduces new, unpredictable forces that could potentially compromise the integrity of the printed structure [10]. On the other hand, 3DCP can serve as a supportive framework similar to traditional concrete construction, where the reinforcement must be completely set up and secured within the formwork before placing [11]. An alternative technique involves the simultaneous additive manufacturing of concrete and reinforcement, where the reinforcing cable is embedded within the concrete filament and deposited concurrently with the concrete through extrusion [12]. However, this approach carries a significant risk of cable slippage. Short fiber-reinforced 3DCP is a competing strategy since the fibers can be incorporated into 3DCP to enhance tensile strength and toughness while also mitigating the risk of shrinkage-induced cracking and reducing crack width [13]. A 3D printable ultra-high-performance concrete (UHPC) that incorporated 6-mm steel fiber at a 2% addition was successfully developed by Arunothayan et al. [14] that presented excellent extrudability and buildability. The authors also confirmed that the mechanical properties depended on the loading direction in printed samples but were least affected by the steel fiber content. Regardless of the testing direction, incorporating steel fibers improved both the compressive and flexural strengths of the cast and printed samples [15]. Yang et al. [16] found that 10-mm steel fibers were more likely to clog the nozzle than 6-mm steel fibers, indicating that longer fibers hinder extrusion, making shorter steel fibers more suitable for use in 3DCP. The printability of PVA and PP fiber-reinforced composites was studied by Chen et al. [17]. They discovered that fiber incorporations above 1.25% for PVA fibers and 1.5% for PP fibers impeded extrusion, thus reducing extrudability. Moreover, different fiber types had varying effects: steel fibers (rigid fibers) caused the lowest extrusion pressure, while PVA and PP fibers (flexural fibers) led to higher pressures. Chu et al. [18] showed that incorporating fibers slightly reduced the extrusion pressure without adversely affecting the extrusion properties when adjusting the SP amount to maintain constant flow consistency. However, it is crucial to acknowledge the existence of a critical fiber concentration threshold, representing a compromise between the hardened mechanical properties and fresh properties. Beyond this point, the extrudability of the printable mixture is severely compromised, primarily due to the fiber clumps. These clumps obstruct concrete flow during extrusion through narrow channels in the hopper and nozzle, thereby hindering its application and compromising its homogeneity in the printed elements [19].

This research aimed to reduce overreliance on cement using CC and LF to formulate 3D printable mixtures and enhance the mechanical properties and cracking resistance by using short fiber reinforcement. The effect of CC and LF content as well as their combinations on packing density, robustness, and water retention ability was evaluated. The optimization of fiber content was then carried out by considering both the key fresh and mechanical properties, with the aim of determining the optimal threshold that ensures extrudability

without compromise. A fiber-reinforced calcined clay-limestone-based cementitious material (FR-LC³) was developed specifically for 3D printing (3DP) applications. In addition, the rheological properties and shape retention abilities of the developed FR-LC³ with different binder systems and varying SP-to-VMA (SP/VMA) ratios were evaluated. The printability of these mixtures was validated through inline printing using an extrusion-based screw-type 3D printer.

2. Experimental Program

2.1. Materials and Mixture Proportions

Type III cement (PC) with a Blaine fineness of 625 m²/kg was used in this study to promote early strength development, thereby enhancing the structural build-up of 3DCP. The alternative binder materials consisting of CC and LF were used to replace cement. The relatively low-grade CC with 65% metakaolin was obtained from kaolinitic clay heated up to 650 °C. Fine limestone powder with a spherical shape was used as filler. Table 1 illustrates the chemical composition and median particle size (d₅₀) of the investigated binder materials. The CC had the coarsest particles, followed by PC and LF in size.

Table 1. Chemical composition, physical characteristics, and 7-day cumulative heat of the binder materials.

	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	K ₂ O	d ₅₀ (μm)	Calculated SSA (m ² /cm ³)	7-Day cum. Heat (J/g SCM)
PC	19.1	3.88	3.51	68.4	1.8	0.46	8.5	1.12	340
CC	57.1	35.3	3.3	0.73	0.68	1.04	16.6	0.8	381
LF	2.97	0.7	0.17	95.33	0.18	0.13	3.5	2.33	76

The specific surface area (SSA) of the binder materials was determined from their particle size distribution (PSD) curves, assuming that the particles were spherical. The SSA per unit volume can be estimated as follows, and the calculated SSA of each binder material is listed in Table 1.

$$SSA = \sum_{i=1}^j \frac{6V_i}{D_i}, \quad (1)$$

where V_i and D_i represent the volume fraction between two successive particle sizes and the average particle diameter (μm), respectively. j denotes the number of fractions in the grading curve.

The pozzolanic reactivity of alternative binders was examined using the rapid, relevant, and reliable (R³) test with isothermal calorimetry (I-Cal) measured at 40 °C. Their 7-day cumulative heats are also detailed in Table 1. It can be observed the CC showed higher pozzolanic reactivity than cement, while LF exhibited the lowest 7-day cumulative heat below 100 J/g SCM, which can be regarded as an inert material [20].

To enhance the packing density of the granular skeleton, two types of sand with maximum particle sizes of 4.75 and 2.0 mm were used. A polycarboxylate-based superplasticizer (SP) with a solid content of 30% and a cellulose-based VMA were combined to ensure adequate flowability and stability during extrusion and promote the structural build-up rate after layer-wise deposition. Additionally, a striated straight steel fiber limited to 6 mm long was used to prevent a 30-mm nozzle clogging in the 3DP process [21].

The paste mixtures that incorporated incremental contents of CC and LF as well as their combinations were investigated. The nomenclature for binary systems incorporating CC with replacement levels ranging from 20% to 50% and LF with substitution rates from 5% to 30% is denoted as CC20–CC50 and LF5–LF30, respectively. In addition, the CC30LF15 and CC40LF20 binders are categorized as ternary systems. Given the varying solid densities of the binder materials, the paste mixtures were formulated based on volume replacements to ensure a consistent volume of solid particles.

The fiber-reinforced mortars made with the optimized binder combination were evaluated with varying fiber contents. The addition of a 6-mm steel fiber was selected up to

a maximum of 2%, which was determined to be the optimal fiber content without causing clogging in a 30-mm circular nozzle [15,22]. The water-to-binder ratio (w/b) of 0.3 and sand-to-binder ratio (s/b) of 1.5 was set to prepare the 3D-printed mortar. The cellulose-based VMA with two dosage levels, 0.25% and 0.5% by mass of binder, were added.

2.2. Test Methods

2.2.1. Flow Characteristics and Water Retention Ability for Binder Optimization

The flow characteristics of the paste mixtures made with various binder systems were evaluated using the mini-slump cone based on the method detailed in [22]. This test can be used to determine the effect of CC and LF content as well as their combinations on packing density and robustness, and the test procedure is illustrated in Figure 1. The relative spread area of the slump flow linearly increases with the water-to-powder volume ratio (V_w/V_p) of the paste mixture. The intercept of the relation refers to the minimum water content (MWC) necessary to start the flow, and the slope represents the relative water demand (RWD) that reflects the flow's sensitivity to changes in water content. Assume that no air is entrapped in the paste mixtures; a low MWC indicates high packing density, while a high RWD indicates high robustness. The paste mixtures were formulated with incremental V_w/V_p varying from 1.1 to 1.5. A constant superplasticizer (SP) dosage of 0.1% by mass of the binder was added to secure the complete dispersion of binder particles, corresponding to the saturation point for the binder systems [23].

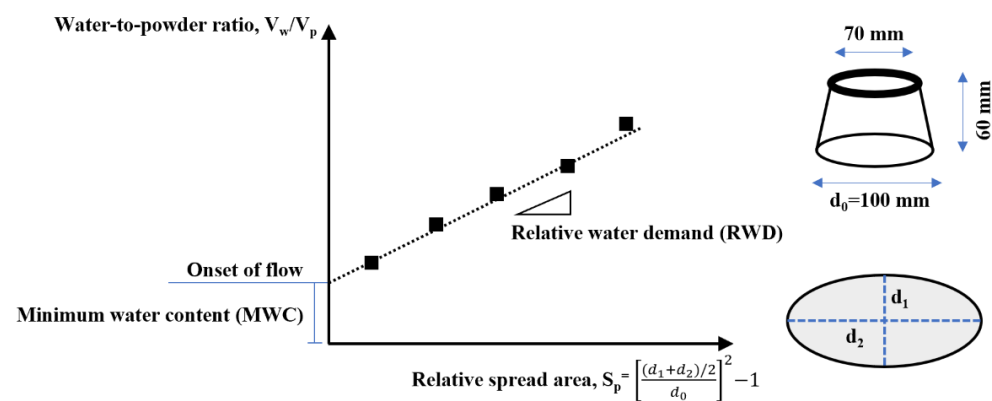


Figure 1. The test principle for measuring the flow characteristics of the paste mixtures.

For optimal performance in 3DCP, maintaining the stability and homogeneity of the ink materials under a given pumping or extrusion pressure is essential. This stability is crucial to avoid dynamic bleeding and segregation, which can cause blockages in the pumping pipe or extruder and detrimentally affect the material's properties after extrusion, potentially undermining the structural integrity of the printed objects [24]. The fluid filtration test was employed to determine the water retention ability of the paste mixtures made with various binder combinations under a given air pressure. The test setup is illustrated in Figure 2a including a container filled with samples, a filtrate tube, and a hold connected to an air pump. The rate of water bleeding from the filtrate tube under pressure can be measured and characterized as the forced bleeding capacity. Figure 2b shows the test diagram of the reference paste made with pure cement, and the slope of the relative amount of filtrated water versus the square root of testing time refers to the forced bleeding capacity. A low forced bleeding capacity indicates a high water retention ability, reflecting greater extrudability. Approximately 700 g of the sample was placed in the filtration cylinder, and the test duration was 20 min. An air pressure of 160 KPa was selected to ensure that the entrained water could be filtered out.

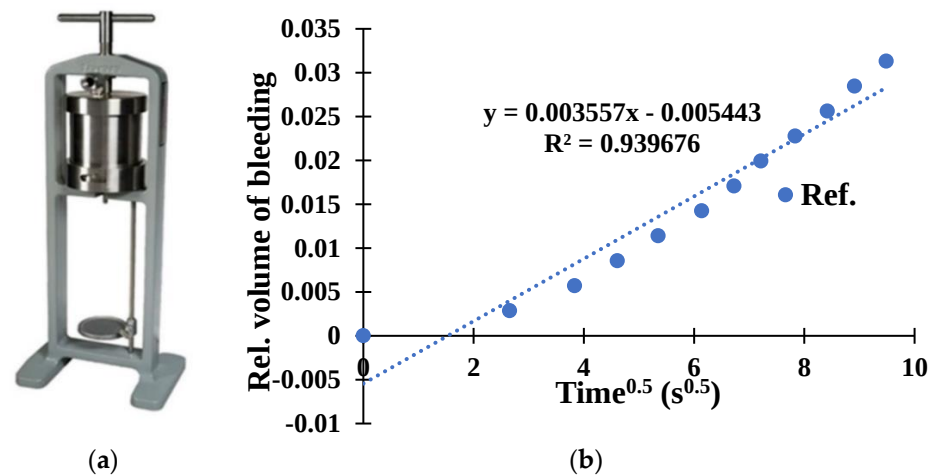


Figure 2. Fluid filtration test for measuring the water retention ability. (a) Test setup; (b) forced bleeding capacity obtained from the slope of the typical diagram.

2.2.2. Fresh and Mechanical Properties for Fiber Content Optimization

The mini-slump flow was used to determine the SP demand of fiber-reinforced mortar with varying fiber contents to achieve a constant slump flow of 170 ± 10 mm after 25 jolts, considered an appropriate fluidity for 3DCP [25]. In addition, the fluid filtration test was used to investigate the effect of fiber content on the water retention ability of the fiber-reinforced mortar made with the selected binder systems. The same test procedure was employed as introduced in Section 2.2.1, but the air pressure was increased to 240 KPa to fit the denser matrix induced by fiber bridging.

The compressive and flexural strengths as well as the toughness of the fiber-reinforced mortar mixtures were determined using the unconfined uniaxial compression test and third-point loading test, as shown in Figure 3. The 50-mm cubic samples and $405 \times 75 \times 75$ mm³ prismatic beams were tested for compressive and flexural strengths, respectively. The samples were cured under lime-saturated water for 28 days before testing. An average of three cubes and two beams was obtained to secure proper reproducibility.

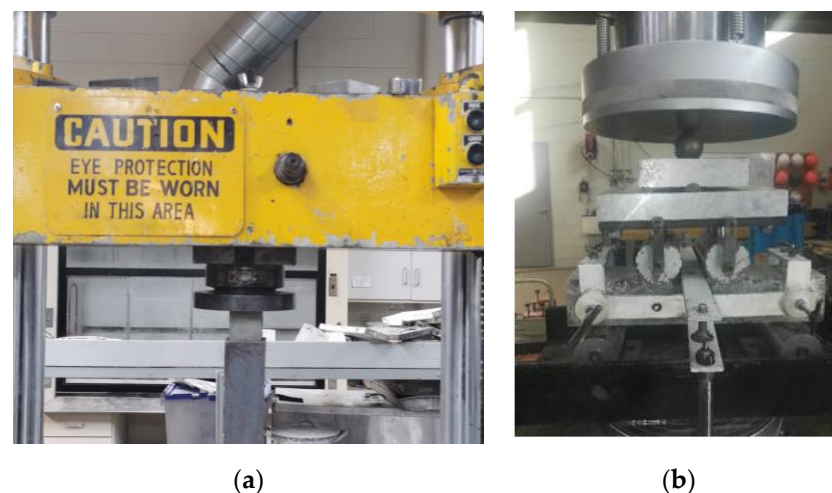


Figure 3. Testing setup for measuring hardened properties: (a) compression test; (b) third-point loading test.

For the bending test, the loading rate was set at 0.1 mm/min, with displacement meticulously tracked by two transducers (LVDTs) positioned on both sides of the beams. In the event of a fracture occurring within the middle of the beam, flexural strength was

derived using Equation (2). The area beneath the load–deflection curve, from 0 to 2 mm deflection, was quantified as energy absorption, serving as blows:

$$f = \frac{PL}{bd^2}, \quad (2)$$

where f , P , and L represent the flexural strength (MPa), the maximum load (N), and the span length (mm), respectively. d and b represent the depth and width of the beam (mm), respectively.

2.2.3. Offline and Inline Printing Assessments

Offline evaluations including dynamic and static rheological tests and axial deformation tests were employed to evaluate the fresh properties and shape stability of the fiber-reinforced mixtures. The effect of binder systems and chemical admixture contents was emphasized.

A Contec 5 coaxial rheometer was used to measure the rheological properties. The dynamic rheological properties were determined based on the steady-flow measurement protocol introduced in [22]. Each mortar was replicated two times for accuracy. Data points at rotational velocities of 0.45 and 0.5 rps were removed after verifying signs of equilibrium. The Reiner–Riwlin model was used to calculate the dynamic yield stress and plastic viscosity, as shown in Equations (3)–(5).

$$T = G + H \cdot N, \quad (3)$$

$$\tau_0 = \frac{\left(\frac{1}{R_i^2} - \frac{1}{R_o^2}\right)}{4\pi h \ln \frac{R_o}{R_i}} G, \quad (4)$$

$$\mu = \frac{\left(\frac{1}{R_i^2} - \frac{1}{R_o^2}\right)}{8\pi^2 h} H, \quad (5)$$

where T and N represent the torque (N·m) and rotational velocity (m/s), respectively. G and H refer to the intercept and slope of the linear regression of the flow curves, respectively. τ_0 and μ refer to the dynamic yield stress (Pa) and plastic viscosity (Pa.s), respectively. h is the height (m) of the inner bob immersed in the mortar. R_i and R_o denote the radius of the inner bob (0.1 m) and outer cylinder (0.15 m), respectively.

The static yield stress (τ_s) of the fiber-reinforced mixtures was evaluated at each resting period of 5, 15, and 30 min, respectively. The mortars were subjected initially to a shallow rotational velocity of 0.05 rps for 120 s without any pre-shear to determine the static yield stress, which can be calculated as follows. The evolution of yield stress within 30 min was characterized using the thixotropic index (A_{thix}), which refers to the slope of linear evolution.

$$\tau_s = \frac{T_{\max}}{2\pi^2 h R_i^2}, \quad (6)$$

$$A_{\text{thix}} = \frac{\tau_{s,30 \text{ min}} - \tau_{s,5 \text{ min}}}{t}, \quad (7)$$

The immediate axial deformation test was used to evaluate the shape stability proposed by [26]. The test measured the axial strain of a 700 g fiber-reinforced mortar filled within a 15 mm thick ring plate subjected to incremental loads representing the applied weight of the added layers at different time intervals during 3DP. Four dial gauges were attached to measure the average axial strains under each incremental load. The selected time intervals corresponded to the rest periods of 5, 10, 15, 20, 25, and 30 min.

Inline printing via an extrusion-based 3D printer was used to evaluate the printability of the fiber-reinforced mortar mixtures. The screw-type printer is illustrated in Figure 4. An extruder mounted with an auger and nozzle was installed on a gantry frame measuring

$1.5 \times 1.5 \times 1.5 \text{ m}^3$. Guided by a controller, the three-axis movement of the extruder, powered by an actuator, was meticulously configured to fabricate the designed printing objects. For the X, Y, and Z axes, the moving speed of the extruded was customized at 50 mm/s, 50 mm/s, and 5 mm/s, respectively. A circular 30-mm nozzle was selected, and the rotational speed of the augur was set at 0.8 rounds per second constantly during extrusion.



Figure 4. Apparatus of the extrusion-based screw-type 3D printer.

The one-layer filaments with a continuous length of 300 mm and conform width of 40 mm were extruded to examine the extrudability of fiber-reinforced mortar mixtures made with varying fiber contents. Once the optimized fiber content was determined, the buildability of the fiber-reinforced mortar mixtures was achieved by adjusting the admixtures and evaluated via the maximum buildable height. A 300-mm single-wall structure was built layer-wise under the same printing parameters until failure occurred.

3. Results and Discussion

3.1. Flow Characteristics and Water Retention Ability of Paste Mixtures

The effect of CC and LF content as well as their combinations on the flow characteristics (i.e., MWC and RWD) are illustrated in Figure 5. The MWC decreased with the addition of CC, while the RWD slightly increased when CC replaced the 20% cement content. As the replacement level of the CC increased, the MWC of the paste increased, but the RWD decreased. The CC50 mixture exhibited a 34% reduction in RWD compared to the reference paste made with pure cement. This indicates that a low replacement level (less than 20% by volume) of CC can increase both the packing density and robustness of the blend binders. Instead, a higher CC addition resulted in a significant reduction in the flow characteristics. Replacing an excessive amount of cement with CC particles that are twice as large (shown in Table 1) could lead to a wall effect that will disrupt the original packing of cement particles at wall-like boundaries [27].

The LF addition led to a slight increase in MWC but a drop in RWD. With the increase in the LF replacement, the MWC decreased while the RWD slightly increased. Due to the micro-filling effect induced by the ultrafine LF, the entrapped water inside the granular skeleton of cement particles could be released and filled by the LF [28]. On the other hand, the MWC and RWD significantly decreased when the CC and LF were combined, most notably in the CC40LF20 mixture with the lowest MWC and RWD values, which were reduced by 6% and 48%, respectively, compared to the reference paste.

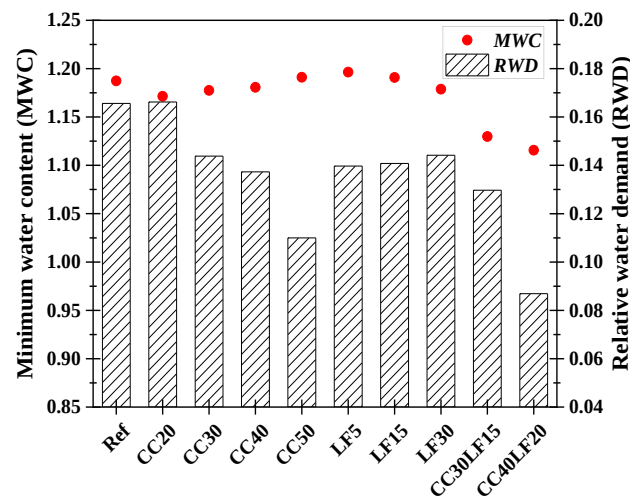


Figure 5. MWC and RWD of the paste mixtures made with various binder systems.

The effect of the SSA of the blend binders on the flow characteristics of the paste mixtures was also discussed. Table 2 shows the estimated SSA of the investigated binder systems, as calculated based on Equation (8).

$$SSA_{com} = \frac{V_c \times (SSA_c) + V_\alpha \times (SSA_\alpha) + \dots + V_\eta \times (SSA_\eta)}{V_c + V_\alpha + \dots + V_\eta} \quad (8)$$

where V_c , V_α , and V_η refer to the volume fractions of the cement and other alternative binders, respectively.

Table 2. SSA calculated for the blend binders of each mixture.

	Blend Binders	SSA _{com} (m ² /cm ³)
Binary systems	CC20	1.06
	CC30	1.02
	CC40	0.99
	CC50	0.96
	LF5	1.18
	LF15	1.30
	LF30	1.48
Ternary systems	CC30LF15	1.21
	CC40LF20	1.23

As shown in Table 2, the CC addition adversely affected the SSA of the binary binders, which can be reflected in the drop in the RWD as the replacement of CC increases. For instance, the decrease in SSA from 1.06 to 0.96 m²/cm³ led to a reduction in RWD from 0.17 to 0.11. However, a low replacement level of the CC (i.e., CC20) exhibited a slightly higher RWD than the reference paste made with 100% cement, which can be explained by the subhedral flaky habit and clustered structure of CC particles [7]. Conversely, although the LF addition enhanced the SSA of the binary binders, the RWD of paste mixtures that solely utilized LF as a replacement showed a marked reduction; this can be attributed to the inherently inert nature of LF (shown in Table 1). Incorporating a high content of LF may lead to an increased distance between particles due to the loosening effect of fine particles, causing larger particles to be displaced further apart [23]. This inert material functions predominantly as a filler, negatively affecting the rheological characteristics but improving the mechanical properties and durability [29]. As for the ternary systems, the

combined addition of LF and CC amplified the adverse impact of both binder materials on the RWD of the past mixtures. However, the increased SSA in the ternary binders reduced the MWC to initiate flow, consistent with the observation reported by [23], indicating a higher packing density of the combination of cement, CC, and LF particles.

The effect of the CC and LF content and their combinations on the water retention ability is shown in Figure 6. The highest and lowest forced bleeding capacity corresponded to the binary systems containing 30% LF and 50% CC, respectively. It can be concluded that the CC addition increased the water retention ability of the paste mixtures made with the blend binders, while the LF negatively affected it. Hatch et al. [30] noted that clay minerals, particularly the kaolinite utilized in this study, exhibited a high capacity for water adsorption due to the uptake of water within their interlayer spaces. On the other hand, the micro-filling effect resulting from the addition of LF can release water trapped between cement particles, thereby diminishing the water retention capacity of binary systems. Additionally, ternary binders incorporating both CC and LF exhibited reduced forced bleeding capacities; moreover, the CC30LF15 binder showed superior water retention compared to the CC40LF20 binder. This shows that the adverse effect of LF outweighed the water uptake capacity of the CC particles.

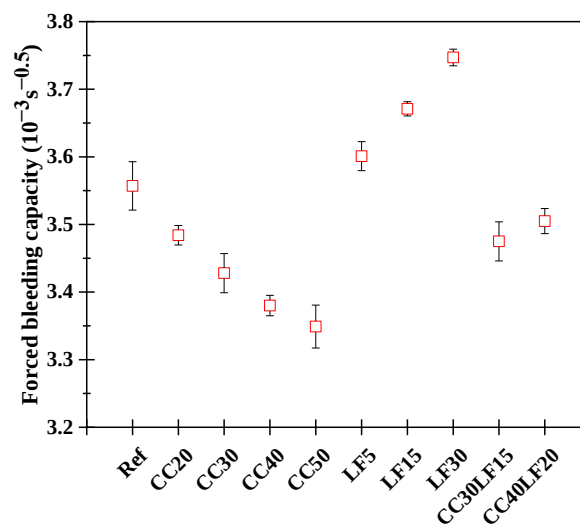


Figure 6. Forced bleeding capacity of the paste mixtures made with various binder systems.

In light of the above analysis of the experimental results, the CC20 binder, noted for its highest packing density, robustness, and comparatively high water retention ability within binary systems, together with the CC30LF15 binder, was selected for the following evaluation.

3.2. Fresh and Hardened Properties of Fiber-Reinforced Mixtures

Based on the optimization of blend binders, the sand and steel fibers were incorporated, and the effect of the fiber contents on the fresh and hardened properties of the fiber-reinforced mortars incorporating the CC30LF15 binder was investigated. The SP demand to maintain a constant slump flow after 25 jolts and the forced bleeding capacity of fiber-reinforced mortars with varying fiber contents are shown in Figure 7. The fiber incorporation slightly increased the SP demand while reducing the forced bleeding capacity, indicating a negative impact on fluidity but a contribution to water retention ability. For example, the incorporation of 2% steel fiber exhibited the highest SP demand but the lowest forced bleeding capacity, 30% higher and 50% lower, respectively, compared to the mortar without the steel fiber. This can be attributed to the physical interlock between fibers and the fiber and aggregate, which can restrain the flow of mortars and, in the meanwhile, enhance their resistance to the dynamic pressure, mitigating the water bleeding. It is

noticeable that the increased SP demand induced by the 6-mm steel fiber was far below the negative effect of the 13-mm fiber [31] while maintaining a competent water retention ability.

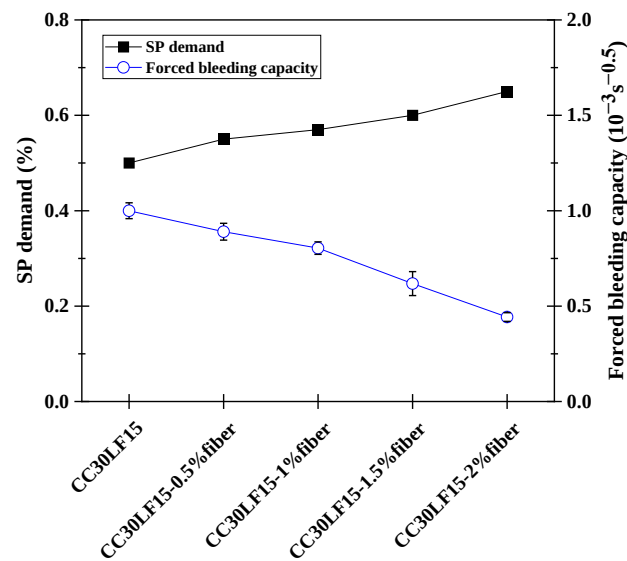


Figure 7. SP demand and forced bleeding capacity of mortars with varying fiber contents.

Figure 8 illustrates the influence of the volume fraction of fiber on the 28-day mechanical properties of fiber-reinforced mortars made with the optimized CC30LF15 binder. A significant contribution of the steel fiber on hardened mechanical properties could be observed, most notably in the flexural strength. The 2% fiber addition led to 43%, 275%, and 240% increases in the 28-day compressive and flexural strengths and toughness, respectively. The presence of steel fibers can constrain lateral deformation under compression loads; moreover, the strong physical bonding between the fiber and density matrix can provide support. In addition, the increased flexural strength and toughness can be explained by the strain-hardening behavior induced by the fiber-bridging effect [32]. Mechanical fiber interlock, frictional sliding between fiber–matrix interfaces, and fiber debonding will contribute to the energy dissipation under loads and thus the toughness [31]. Therefore, the 2% steel fiber with optimal mechanical and moderate fresh properties was selected for further evaluation.

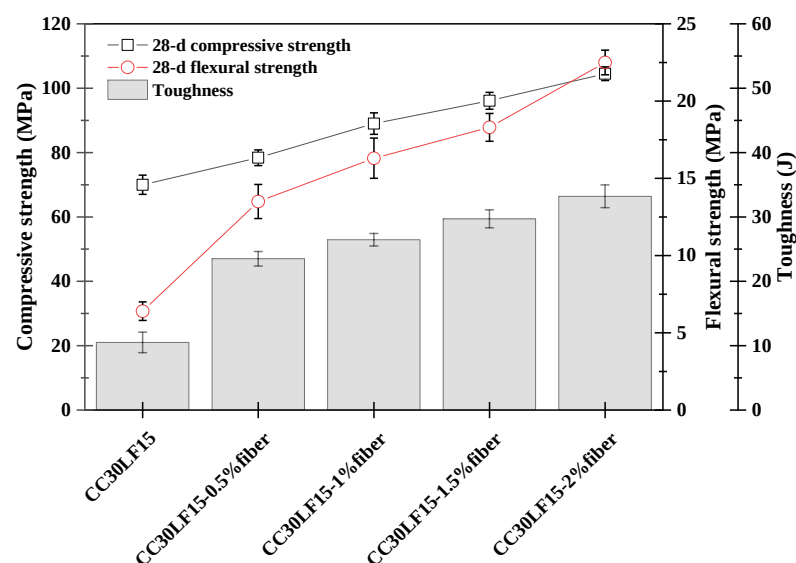


Figure 8. The 28-day compressive and flexural strengths as well as the toughness of mortars with varying fiber contents.

3.3. Offline and Inline Printing Validation for Extrudability and Shape Stability

Based on the optimization of binder systems and fiber content, the printability of fiber-reinforced mortars incorporating two different binder systems and 2% steel fibers was evaluated using offline and inline printing assessments. Two VMA contents, SP/VMA ratios of 2.6 and 1.3, were added as a thixotropic modifier to improve the structural build-up rate for better buildability.

The dynamic rheological properties (i.e., dynamic yield stress and plastic viscosity) of the investigated fiber-reinforced mortars are shown in Figure 9. It should be noted that the SP dosage was varied for two different binder systems as the constant slump flow of 170 ± 10 mm after 25 jolts was maintained. Therefore, the dynamic yield stress values of the CC20 and CC30LF15 fiber-reinforced mortars were more or less the same, as shown in Figure 9. However, the plastic viscosity of the mortar made with the CC20 binder was significantly higher than that of the mortar with the CC30LF15 binder. This was consistent with the results of the flow characteristics in Section 3.1. The enhanced packing density, attributed to the micro-filling effect of the LF, led to an increase in the availability of water molecules acting as lubricants, which in turn decreased the viscous dissipation and thus the viscosity. On the other hand, adding VMA while maintaining the same SP dosage resulted in a rise in both the dynamic yield stress and plastic viscosity. For instance, the CC30LF15-2%fiber+1.3SP/VMA mortar exhibited the highest values of 306 Pa and 29 Pa·s, respectively, which was increased by 16% and 28% compared to the CC30LF15 fiber-reinforced mortar without any VMA. Without competing absorption that can interfere with the effect of VMA resulting from the extra SP dosage, the addition of high molecular weight VMA is capable of effectively trapping water within the pore solution [33], thereby resulting in an increase in viscosity.

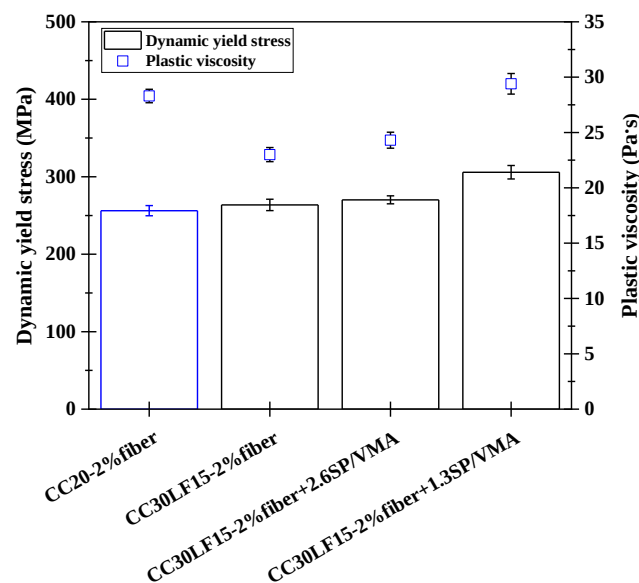


Figure 9. Dynamic yield stress and plastic viscosity of mortars with different binder systems and VMA contents.

Figure 10 illustrates the static rheological properties (i.e., static yield stress at 5 min and thixotropic index from 5 to 30 min) of the investigated fiber-reinforced mortars. The initial offset in static yield stress observed in the mortar was primarily attributed to the network formation resulting from particle colloidal flocculation, considering the negligible impact of very early-age hydration after mixing such as 5 min [34]. The CC20 fiber-reinforced mortar showed the highest static yield stress, indicating pronounced particle flocculation. This suggests that a significant number of flocs formed, particularly in the absence of the LF. Moreover, the intrinsic layered particle structure of the CC resulted in a high water demand and increased flocculation. Muzenda et al. [29] reported that the CC was identified

as a critical factor that enhanced plastic viscosity and static and dynamic yield stress, while the LF, even in smaller quantities, tended to mitigate these rheological properties slightly, thus helping adjust the workability. In addition, the addition of VMA led to an increase in static yield stress. This can be attributed to the long polymer chains in the VMA, which can entangle each other, forming a relatively robust network [33].

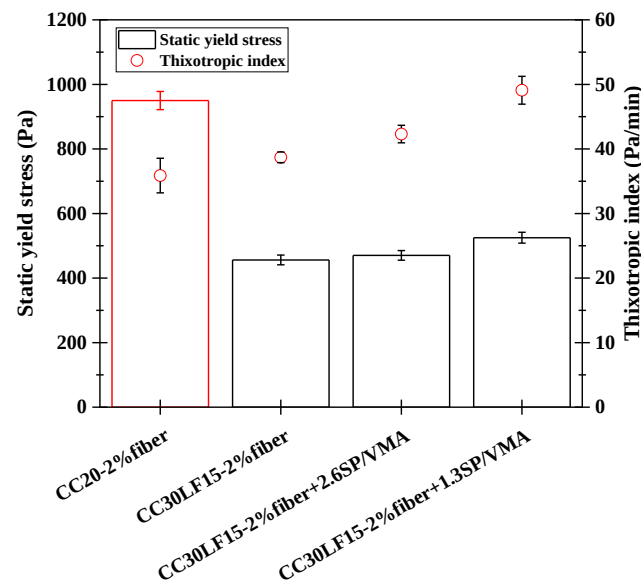


Figure 10. Static yield stress and thixotropic index of mortars with different binder systems and VMA contents.

The network formed by particle colloidal flocculation becomes progressively rigid as a result of the nucleation of early-age hydration products at the contact points of binder particles [35]. The bridging effect plays a crucial role in elucidating the thixotropic behavior (A_{thix}), as illustrated in Figure 10, and should be considered in conjunction with particle flocculation [36]. Despite the CC20 binder displaying a higher static yield stress than the CC30LF15 binder, the A_{thix} was lower. This can be attributed to the much finer LF providing more nucleation sites, which enhanced the bridging effect of early-age hydration products. Additionally, the increase in VMA dosage from 0.25% to 0.5% led to a 9% and 27% increase in A_{thix} , respectively. This heightened A_{thix} is likely due to the entanglement of long polymer chains, which was also reported by Le et al. [37].

The axial strains of the investigated fiber-reinforced mortars under incremental loads corresponding to the resting period of 5, 10, 15, 20, 25, and 30 min are shown in Figure 11. The CC20 fiber-reinforced mortar exhibited the lowest strain of 0.7% when weight was added at 5 min, aligning with the findings of the highest static yield stress observed. The significant colloidal flocculation exhibited by the CC20 binder enhanced its weight resistance capacity, which is particularly crucial at the beginning of printing layers. This helps prevent plastic collapse failure due to the inadequate yield strength, thereby improving buildability. Moreover, the incorporation of VMA also contributed to lower axial strains. When the SP/VMA ratio was decreased from 2.6 to 1.3, there was a noticeable reduction in axial strains by 26% and 33% at 5 min, in comparison to the CC30LF15 fiber-reinforced mortar that did not contain any VMA. With the number of loads increasing at 5-min intervals, the rate at which axial strains increased in the CC30LF15 fiber-reinforced mortar containing VMA was significantly lower compared to the CC20 fiber-reinforced mortar. This reflects the higher A_{thix} resulting from the addition of VMA, which provided greater yield stress for the printed filament over time, enabling it to withstand more weight from the upper layers for up to 30 min. For instance, the CC30LF15 fiber-reinforced mortar incorporating a SP/VMA ratio of 1.3 showed the lowest axial strain at 30 min, which decreased by 13% compared to the pure CC30LF15 fiber-reinforced mortar.

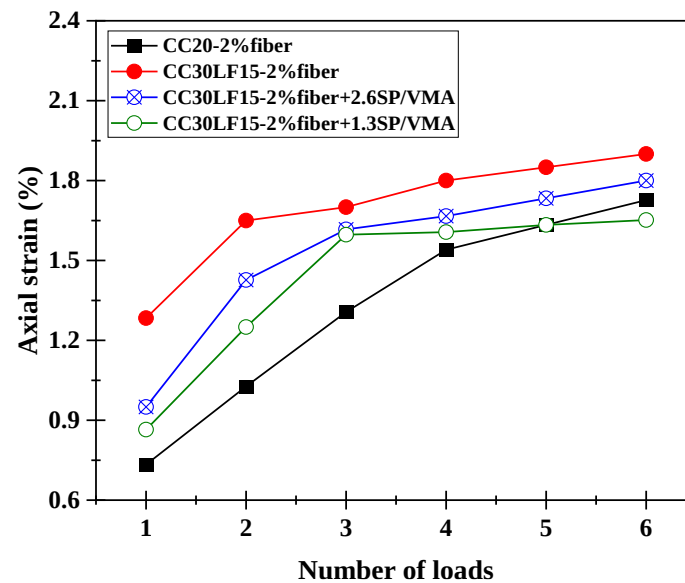


Figure 11. Immediate axial strain of mortars with different binder systems and VMA contents.

Following the offline assessments, which included evaluations of the rheological properties and axial strains of the fiber-reinforced mortar with two different binder systems and SP/VMA ratios, inline 3D printing was employed to validate their printability. The feasibility of printing with the maximum fiber content was assessed by conducting a visual inspection of a 300-mm continuous length of the extruded filament with a designated width of 40 mm, as shown in Figure 12. It can be observed that an increase in fiber content led to a decrease in filament width, attributed to diminished fluidity due to the interlocking of fibers.

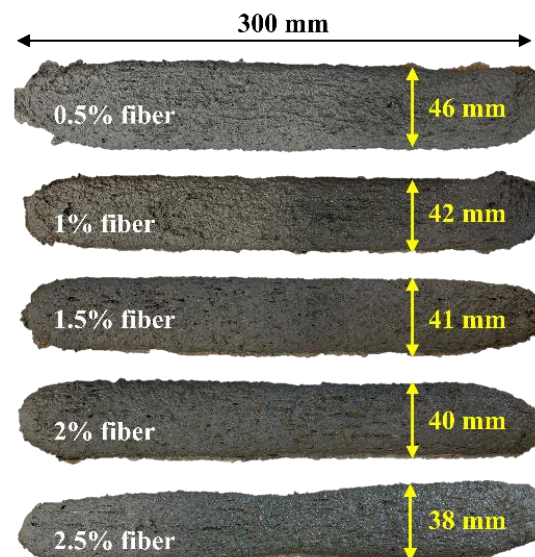


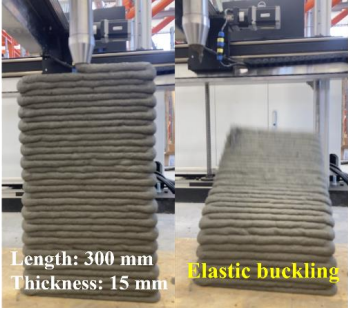
Figure 12. Visual inspection of a continuous length of extruded filament with varying fiber contents.

When adding up to 2.5% steel fiber, the filament showed noticeable unconformity and discontinuity, likely due to the nozzle becoming clogged by an excess of fibers. The nozzle is more likely to clog due to the restrained flow and physical interlocking when the aggregate volume or fiber factor (fiber aspect multiplied by fiber volume) is high. It should be noted that the printer size and printing parameters can also affect the printing quality. The large-scale 3D printer equipped with a higher printing capacity and larger nozzle size is capable of printing objects that incorporate coarse aggregates with a maximum size of 10 mm [38,39] and can even handle 2.5% 13-mm steel fiber [40] without significantly compromising

printability. The filament incorporated with 2% 6-mm steel fiber was extruded smoothly and continuously without clogging issues. It also maintained a conforming shape aligned with the target width when using the lab-scale extrusion-based 3D printer with a 30-mm nozzle in the research lab.

Table 3 presents a printability evaluation of the fiber-reinforced mortar incorporating two distinct binder systems or SP/VMA ratios. It details the maximum layer/height achievable for continuously building up a 300-mm slender wall with a layer thickness of 15 mm, alongside the first layer's deformation after printing. The ink materials that can be built up to achieve higher layers within the open time have a greater potential for scaling up in large-scale printing operations. All printing objects made with the investigated fiber-reinforced mortars showcased outstanding printability without bleeding, segregation, or excessive deformation. Nevertheless, elastic buckling failures were observed at the relatively high layers. Elastic buckling primarily depends on the geometric features of the printed object, especially in slender vertical structures as well as the strength and stiffness of the materials used in printing [41]. It can be expected that, below a given height, plastic failure would be critical while, above this given height, buckling would become critical.

Table 3. Maximum buildable height and deformation of the first layer after printing.

	Failure Mode	Maximum Buildable Layer/Height (mm)	Deformation Strain of First Layer
CC20-2%fiber		32/480	Negligible
CC30LF15-2%fiber		30/445	33.3%
CC30LF15-2% fiber+2.6SP/VMA		36/538	13.3%
CC30LF15-2% fiber+1.3SP/VMA		40/600	Negligible

The fiber-reinforced mortar using the CC20 binder showed a marginally greater capacity for layer deposition, while the deformation strain of the initial layer remained negligible compared to the use of the CC30LF15 binder. This can be attributed to the higher static yield stress of the CC20 fiber-reinforced mortar resulting from the higher particle flocculation of the CC20 binder systems. The enhanced static yield stress was sufficiently greater than the gravitational force, allowing it to withstand the weight of the subsequent upper layers without compromise. On the other hand, reducing the SP/VMA ratio from 2.6 to 1.3 in the CC30LF15 fiber-reinforced mortar reduced the deformation of the initial layer; moreover, 6 to 10 additional buildable layers could be achieved. This enhanced shape retention ability is likely due to the increased static yield stress and A_{thix} with the addition of VMA, as shown in the above-mentioned rheological results.

4. Conclusions

A printable FR-LC³ was methodically developed, starting from the mixture design to the printability evaluation. Detailed conclusions are drawn as follows:

1. Replacing cement with a high content of CC decreased the packing density and robustness of the blend binders while enhancing their capacity to retain water. In contrast, the LF addition improved the packing density but compromised robustness and water retention. However, a low replacement level (below 20% by volume) of CC and the combinations of CC and LF with increased SSA enhanced the packing density and water retention ability.
2. The addition of steel fibers slightly increased the SP dosage, but it significantly enhanced the ability to prevent bleeding. Moreover, it considerably improved the

compressive and flexural strengths and toughness of the fiber-reinforced mortars. However, the optimal fiber content threshold should be aligned with the printer's capacity, aiming to prevent nozzle blockages and guarantee uninterrupted extrusion quality.

3. When maintaining a fixed slump flow, the fiber-reinforced mortar that incorporated the binary CC20 binder exhibited increased plastic viscosity and static yield stress as well as shape retention ability, but a lower A_{thix} compared to the ternary CC30LF15 binder. In addition, incorporating VMA effectively improved the static rheological properties of the fiber-reinforced mortars, contributing to a higher structural build-up rate and thereby enhancing buildability.
4. The developed FR-LC³ mixtures within the 2% steel fiber threshold were extruded smoothly, exhibiting no bleeding, segregation, or excessive deformation. A decrease in the SP/VMA ratio notably increased the maximum buildable height of a single-wall printed object up to the layer of elastic buckling failure.

This study mainly focused on the development of fiber-reinforced eco-friendly mixtures for use in 3DCP and the printability evaluation using offline and inline approaches. However, the anisotropic mechanical characteristics and long-term durability of the printed elements made with the developed FR-LC³ mixtures are equally important for scaling them to large-scale printing operations, which will be explored in further research. Moreover, the life cycle assessment (LCA) can be considered to quantitatively evaluate energy saving and CO₂ emission reductions achieved by substituting cement with CC and LF in 3DCP and comparing these metrics between conventionally cast and printed elements. Additionally, to enhance the sustainability of digital fabrication with fiber-reinforced mixtures, future research could explore the effect of using recycled fibers or other waste materials on the printability and hardened properties of 3DCP.

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