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Limestone–Calcined Clay: An Alternative Binder for 3D Concrete Printing

Nada Oulad Moussa [1]*, *Mohamed* El Haim[1], and *Loubaba* Rida[2,3]

[1]Civil Engineering, Mechanics and Energy Research Team: Modeling and Experimentation (GC2-ME), National School of Applied Sciences of Al Hoceima, Abdelmalek Essaadi University, Al Hoceima, Morocco
[2] Laboratory of Systems, Control, and Decision (LSCD), New Science School of Engineering (ENSI), Tangier, Morocco
[3] Mechanical and Civil Engineering Laboratory, FST of Tangier, Abdelmalek Essaadi University, Tangier, Morocco

Abstract. Additive manufacturing in construction has emerged as a promising alternative to conventional form-work-based building processes. However, most printable mixtures are rich in ordinary Portland cement OPC, which is associated with high embodied carbon and energy consumption. Active 3D concrete printing researchers are exploring limestone-based binders, specifically Limestone–Calcined Clay Cement LC3, as substitutes to OPC for a low carbon footprint. This literature review summarises recent research on LC3 as a low-carbon binder for 3D printing. Primary sources show calcined clay improves static and dynamic yield stress and buildability but reduces flowability, while limestone filler acts as a fine filler, improving particle packing and early-age hydration. Optimised LC3 mixtures achieve high yield stress and thixotropy that enable the printing of up to 23 layers while keeping good pumpability and extrudability. Fibre reinforcement increases 28-day compressive and flexural strength without clogging the nozzle. Studies report that LC3-based concretes can attain 28-day compressive strengths of 30–50 MPa while achieving up to 30–50 % reductions in CO_2 emissions relative to OPC. This review discusses rheological behaviour, mechanical properties, sustainability benefits, and mixture-optimization strategies of LC3, also pointing out research gaps and future directions for climate-positive 3D printing.

1 Introduction

Building and construction contribute around 38 % of global energy-related carbon dioxide emissions and consume 40 % of the world's energy [1]. Production of conventional concrete is a very carbon-intensive activity. Producing one tonne of clinker requires temperatures of around 1450–1500 °C and releases 900 kg CO_2 per Metric ton of clinker [2], [3]. Extrusion-based 3D concrete printing can reduce material waste by 30–60 %, 50-80% in labor costs, and construction time by 50–70 % due to the fact that material is deposited only where needed [4]. However, most printable mixtures currently contain high OPC contents of 400–600 kg m^{-3} to attain sufficient green strength, which nullifies the possible environmental advantages they may offer [5]. For example, a review into 3D-printed concrete mix designs determined that most mixtures contain high cement contents due to the low aggregate-to-binder ratio required for pumpability and buildability [6].

LC3 is a ternary binder composed of approximately 50 % clinker, 30 % calcined clay, 15 % limestone, and 5 % gypsum. Calcined clay and limestone are abundant, inexpensive, and globally distributed; calcination of low-grade clay occurs at 700–850 °C—significantly lower than the temperatures required for clinker production—resulting in CO_2 emissions of only 0.25–0.37 kg CO_2 kg^{-1} [7]. Calcined clay, or metakaolin, and limestone react with the hydration products of clinker to form C-A-S-H and carboaluminate phases, which densify the microstructure, reduce porosity, and increase mechanical strength [8]. These characteristics render LC3 an attractive low-carbon binder for 3DCP. This review intends to provide a summary of the most recent findings on the rheology, printability, mechanical performance, and sustainability of LC3-based mixtures, as well as optimization studies for 3D printing applications.

2 Composition and Chemistry of LC3

2.1 Component roles

When using LC3, a four-component binder, you're looking at 50% regular Portland cement clinker, 30% calcined kaolinitic clay, also known as metakaolin, 15% finely ground limestone, and 5% gypsum to slow down the setting process [9]. The metakaolin part of the mix is what makes the magic happen.

Coming hotfooting into the mix, metakaolin is an extremely reactive material, sending the yield stress and the thickness of the paste through the roof [10].

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* Corresponding author: nadaom.professionnel@gmail.com

Research tests show that when adding metakaolin from 0% to 10%, the yield stress rises by a whopping 77% and the plastic viscosity almost doubles by 140% [11]. This is basically because the particles in the mix are forming a tighter network, resisting flow [12]. Early stiffening is also given a boost; metakaolin speeds up the formation of new phases, C-(A)-S-H and carbo-aluminate, and gets the whole paste hardening faster [12].

Gypsum supplies sulphate, which is primarily utilized as a regulating agent in early hydration as well as an ettringite-forming agent, while the main hydraulic phases, namely C_3S and C_2S, responsible for strength, are supplied by the clinker [13], [14]. The common LC 3 mix proportion requires a water-to-binder ratio ranging from 0.34 to 0.40, along with admixtures such as superplasticizers, viscosity-modifier substances, fibers, or accelerators [15].

2.2 Environmental benefits

LC3 has a smaller carbon footprint because the calcined clay fraction is processed at a much lower temperature than Portland cement, so only a small decomposition of limestone is required [16]. In a very recently published review on limestone-calcined clay cement, it is confirmed that for a 3D-printable LC3 mortar that reduced-environmental-impact levels of 30–50 % compared to ordinary Portland cement formulations can be achieved, while maintaining equivalent strength properties [17]. A life cycle assessment of an LC3 3D printing formulation quantifies life cycle savings as a 36% decrease in the climate change impact score compared to a conventional 30 MPa 3D printing cement, with up to 15% variability based on supply chain sourcing [16]. A further review emphasizes the fact that the calculated CO_2 equivalent kg CO_2 kg^{-1} emissions values of the calcined clay component can be as low as 0.25–0.37, as opposed to approximately 0.9 kg CO_2 kg^{-1} for Portland cement [18].

Together, these studies confirm that substituting approximately 50% of clinker with calcined clay and limestone can reduce embodied carbon by roughly one-third to one-half, making LC3 a promising low-carbon binder for 3D-printed construction.

3 Fresh Properties and Printability

3.1 Rheology and flowability

The mixtures for 3D printing of Limestone Calcined Clay Cements (LC3) display a noticeable trade-off between shape stability (green strength), which increases due to improved static/dynamic yield stress and plastic viscosity, and fluidity [19]. With increasing metakaolin-based calcined clay content, there's a dramatic increase in static/dynamic yield stress components [19]. For example, using finer high metakaolin-based calcined clay in place of coarser clay increases dynamic yield stress values from 66.23 ± 1.93 to 347.92 ± 5.29 Pa after 10 minutes of resting time [20]. In addition to improved green strength properties due to increased static/dynamic yield stress components (rheological stability), there's a simultaneous enhancement in structuration times (thixotropy) [20]. These high-clay binders result in raised static values of yield stress, to the extent that a mere 0.7% deformation strain was recorded after 5 minutes for a 20% calcined clay (without limestone) mortar, indicating excellent layer stability [21]. Nevertheless, this comes with reduced workability: to reach a comparable slump-flow of 170 mm or extrusion flow, more superplasticizer was required, maintaining a substantially higher plastic viscosity than a mix proportionate to limestone [21], [22]. In fact, increasing the proportion of calcined clays/limestone binder greatly decreases open times (printable times) dramatically, raising the cement replacement percentage from 45% to 75% and 90% in one study [8]. The workable time is reduced from 120 min to 45 min and 25 min, respectively [8]. By contrast, introducing finely ground limestone filler tends to **diminish** yield stress and viscosity, thereby improving flowability [19]. For example, compared to a 20% clay (0% limestone-containing) counterpart with similar slump values, an LC3 mortar containing 30% calcined clay + 15% limestone demonstrated much lower rheological resistance (reduced viscosity) [21]. The fine limestone particles function both as inert-filling materials to counteract the flocculation properties of clays (and slightly resist structuration), provided with additional nucleation sites whose influence could extend to thixotropy[21], [23]. Quantitatively, printable LC3 mortars typically reach dynamic yield stress on the order of a few hundred Pascals and plastic viscosity on the order of 10–30 Pa·s [21], [22]. One specific example involving a fiber-reinforced LC3 mix (with composition 30% clay + 15% limestone) demonstrated dynamic yield stresses achieved 260 Pa with viscosity values of 23 Pa·s (with sufficient flow), whereas adding a viscosity-modifier pushed these up to ~306 Pa and 29 Pa·s (further increasing build stability) [21], [22].

In summary, increased amounts of calcined or metakaolin improve mechanical properties (greater initial strength), while higher limestone content improves fresh fluidity and pumpability by reducing those rheological parameters [24].

3.2 Printable structures with waste-clay and recycled aggregates

In recent studies within the past few years, there have been investigations into the possibility of using lower-grade waste clay materials and recycled aggregate to make 3D-printed Limestone Calcined Clay Cement (LC3). In an article published by Zhao et al., a low-carbon binder was designed for 3D concrete printing. Ground Granulated Blast Furnace Slag (GGBS) and Clay Brick Powder (CBP), which function to replace Ordinary Portland Cement (OPC), were utilized [25]. The fluidity was improved by 45.5% when 65% of OPC was replaced with GGBS [25] [26]. The chloride migration coefficient in 3D-printed mortar was between 8-13% compared to ordinary mortar for layers. It was 24-30% for interlayers [25].

In another study, Al-Noaimat et al. calcinated excavated-waste clays at a temperature of 800 °C to create an LC3 mixture that was mixed with 1% superplasticizer and 0.6% viscosity-modifying agent (VMA) [27]. The mixture was used to create a 23-layer cylindrical object (150 mm high), which was successful. In addition, after 28 days, its compressive strength was around 15.9 MPa, whereas its flexural strength was around 5.5 MPa [27]. The use of 100% natural aggregate mixed with recycled aggregate made from recycled bricks in the LC3 mix increased its compressive strength after 28 days by 36-62% compared to conventional concrete [28]. Freeze-thaw resistance improved by up to 25% with water absorption values largely similar to reference batches up to 70% replacement [28]. These experimental results prove that LC3 can recycle clays to/from printed concrete elements [25], [26], [27], [28].

3.3 Integrating Biotechnology into LC³-Based 3D-Printed Concrete

Recently, there has been interest in bio-based admixtures or biotechnologies to enhance the fresh/hardened properties of 3D-printed concretes, notably those based on LC3 binder or similar. Biopolymers (xanthan, alginate, or cellulose gums), biotechnologies, or biotechnology-based admixtures show promise to enhance rheology (yield stress, thixotropy, or constructability), with little or no environmental burden. As an example, Maierdan et al. studied a binder based on xanthan gum, as a binder for clay-based earth concrete, demonstrating that the addition of 0.6–5 wt% of clay drastically modifies rheology [29]. At a concentration above 0.6% XG, a biopolymer-based three-dimensional network enhances yield stress related to initial conditions (after pre-shearing), improving constructability. At 5% XG, the mix achieves ideal viscosity/yield stress for 3D printing [29]. More recently, biotech-based rheology modification has focused on temperature-induced bioGels using temperature-sensitive nanoparticles (gelatin/carrageenan), which allow larger processing windows to create complex structures [30]. These pastes made with 3% carrageenan are reinforced with 50% solids (suspended particles), which develop compressive strengths comparable to common construction materials after setting [30]. In hardened concrete, there are microbial techniques that provide self-healing properties and strength gains, demonstrating bacterial-mineralization, enabling binding printed layers to self-heal cracks detrimental to structural functionality [31]. Bacteria or enzymes added to the LC3-based 3D concrete display self-healing crack sizes down to millimeter resolution. Also, interface bonding improved by multiple folds [32]. These advancements contribute to both the sustainability by enabling cement reduction and carbon sequestration, and the durability of 3D-printed constructions [29], [30], [32].

4 Mechanical Properties of Printed LC3 Concrete

In recent studies focusing on 3D-printed LC3 (limestone calcined clay cement) concrete, it was found that printing technique and mixture design are significant parameters in determining mechanical properties. As an example, one study reported that for extruded LC3 concrete, cylinders made using extrusion attained a compressive strength of around 73% compared to equal-sized cubes cast using traditional methods after 28 days [33]. In addition to that, splitting tension strength after 28 days was found to be around 88% compared to cast LC3 concrete [33]. Cast concretes possess greater compressive strength due to lower porosity compared to layer-printed concretes [34]. Additive fiber reinforcement was found to check post-peak failure mechanisms within printed LC3 concrete due to bridging action initiated due to crack development, but there exists considerable anisotropy in terms of strength values depending on orientation between layers [21], [35]. In other words, compress strength values show differences based on orientation [35]. Fortunately, compress strength values attain between 30-50 MPa after 28 days for fiber-reinforced mixtures, and flexure strength values attain 10 MPa. These values are similar to or surpass those attainable using conventional OPC-based 3D-printables. Notably, ternary LC3 blends based on 30% calcined clay, 15% LC (limestone), mixed with 6 mm steel fibers (amounting to 2% of the entire mixture's value), CC30+LF15 attained maximum compressive strength values compared to other mixtures .

In addition to that, a "set-on-demand" system based on $Ca(NO_3)_2$ accelerators was proven advantageous for preparing low-clinker concretes [36]. As demonstrated in another study, compress strength values for the previously mentioned low-OPC LC3 mixture are well above 30MPa [36]. In other words, accelerated reactions are advantageous for upgraded initial hydration to attain elevated compressive strength values [36]. These studies clearly demonstrate that LC3 concretes printed using 3D printing techniques possess sufficient compressive strength to match traditional concretes. In other words, these concretes possess remarkably lower carbon footprints.

5 Sustainability and Environmental Impact

Recent life-cycle assessment studies verify that limestone calcined clay cement (LC3) strongly reduces carbon footprint effects by comparison with ordinary Portland cement (OPC). Thus, for instance, CO_2 emissions due to clay calcination amount to only about 0.25-0.37 kg CO_2 per kg material, opposed to 0.9-1.0 kg CO_2 per kg for conventional clinker [37]. Thus, with this cement combining much lower amounts of conventional raw materials, such as clinker replaced by calcined clay and limestone, overall global warming effects decrease by about 30-50% [38]. Indeed, Haverkamp et al. indicate that LC3 concrete mortars can decrease climate change effects by up to 42.6% compared with OPC [38]. Also, by substituting 50% by weight of concrete binder with LC3, CO_2 emissions can decrease by about 29.9% for equivalent strength by concrete, found by Yu et al [39]. Similarly, by using low-ranking excavated clay instead of standard clay in this cement formulation, overall CO_2 emissions decreased by about 39% compared with OPC concrete, as demonstrated by Al-Noaimat et al [27].

Finally, by recycling construction waste for incorporation with this cement, one can produce concrete with about 36-62% improved strength because it resulted in "a more environmentally friendly concrete mixture," involving a complete substitution for virgin aggregate with recycled bricks [28]. Incidentally, any recycling of construction waste for subsequent combination with this cement can obviously greatly minimize the concrete's CO_2 footprint [37].

6 Research Gaps and Future Directions

Standardized testing protocols: None of these studies on LC3 for 3D printing have common approaches for assessing "fresh-state printability". Reviews have pointed out a "lack of standardized testing methods for assessing properties such as extrudability and interlayer build-up [17].

Scale-up and pumping effects: Presently, nearly all experiments involving 3D printing with LC3 have been small-scale samples and lab-scale printers. Outstanding problems related to scale have not been considered. These problems include, for example, assessing how long-distance pumping can affect LC3 material properties and testing whether extruded LC3 can have dimensional stability on a larger scale [17].

Long-term durability: The performance behavior in environments of the LC3 in 3D printed structures has yet to be investigated. Freeze-thaw resistance and long-term durability have been mentioned in one recent literature survey for LC3 [17].

Fiber reinforcement and anisotropy: The function of fibres in 3DLC3 is unclear. Specifically, the interaction between fibre orientation (determined by print path) and anisotropic strength has not been elucidated. Recent studies on fibre-reinforced 3D printed materials remark on the "limited understanding of fibre orientation effects" and identify a need for investigation on hybrid systems of fibres [40].

Digital mix-design tools: At present, there are no published approaches for automatic optimizations in LC3 mixes for 3D. Several authors have argued for designing a digital/design-of-experiments (DoE) platform or a machine-learning model for the determination of ratios for required strength characteristics with low environmental effects [41].

7 Conclusion

Recent research shows that limestone calcined clay cement is an emerging low-carbon binder for extrusion-based 3D concrete printing. Additionally, calcinated clay enhances both yield stress and thixotropy properties. On the other hand, using limestone filler enhances particle packing properties. Based on these observations, optimized LC3 mix designs consisting of a mixture of 20-30% calcinated clays with a 10-20% addition of limestone filler possess static yield stress values high enough to resist up to 23 layers. In addition to using viscosity-modifying agents _, accelerators, and short steel fibers, LC3 possesses 28-day compressive strength values between 30-50 MPa with maximum flexural strength values up to 10 MPa. Environmental analyses emphasize that using LC3 cuts down carbon emissions by approximately 30-50 percent compared to using other printable concretes. Finally, prospects include normalizing printability test procedures. In addition, large-scale applications and LC3 mix design optimization are areas yet to be explored.

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