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LinkedIn Revista Ingeniería de Construcción

Vol. 39
Special Issue 2024
DOI: 10.7764/RIC.00131.21

Advancements in low carbon emission cements for 3D printing: a state-of-the-art review

Avances en cementos de bajas emisiones de carbono para impresión 3D: revisión del estado del arte

Silvestro, Laura *; Scoczynski Ribeiro, Rodrigo *; Navarrete, Iván **[1]

* Federal University of Technology – Paraná (UTFPR), Guarapuava, Brasil.
** Departamento Ingeniería y Gestión de la Construcción, Escuela de Ingeniería, Pontificia Universidad Católica de Chile, Santiago, Chile.

Fecha de Recepción: 20/10/2024
Fecha de Aceptación: 24/11/2024
Fecha de Publicación: 30/12/2024
PAG: 1-10

Abstract

Concrete production, heavily reliant on Ordinary Portland Cement (OPC), is a significant contributor to global CO_2 emissions, responsible for about 8% of human-related emissions. While past efforts to reduce the carbon footprint of cement focused on improving energy efficiency and incorporating supplementary cementitious materials (SCMs), recent innovations have shifted towards low-carbon binders like calcium sulfoaluminate (CSA) cement, limestone calcined clay (LC3) cement, and geopolymers. These alternatives offer considerable reductions in CO_2 emissions during production. However, the adoption of these materials faces challenges, particularly in 3D-printed concrete (3DPC), an emerging construction method that demands specific rheological and mechanical properties. Research into these low-carbon binders shows that CSA cement provides rapid strength development, LC3 mixtures offer promising environmental benefits and structural integrity, and geopolymers achieve high compressive strength but require optimization for durability. This study summarizes a review of the effects of these binders on the fresh properties, mechanical performance, and durability of concrete, emphasizing their potential for use in 3DPC. The findings underscore the importance of optimizing material properties for enhanced performance and sustainability in the construction industry.

Keywords: Calcium sulfoaluminate cement; limestone calcined clay; geopolymers; printability.

Resumen

La producción de hormigón, que depende en gran medida del cemento Portland ordinario (OPC), contribuye significativamente a las emisiones globales de CO2, y representa aproximadamente el 8% de las emisiones relacionadas con el hombre. Si bien los esfuerzos anteriores para reducir la huella de carbono del cemento se centraron en mejorar la eficiencia energética e incorporar materiales cementosos suplementarios (SCM), las innovaciones recientes se han desplazado hacia materiales cementicios bajos en carbono como el cemento de sulfoaluminato de calcio (CSA), el cemento de arcilla calcinada (LC3) y geopolímeros. Estas alternativas ofrecen reducciones considerables en las emisiones de CO2 durante la producción. Sin embargo, la adopción de estos materiales enfrenta desafíos, particularmente en el concreto impreso en 3D (3DPC), un método de construcción emergente que exige propiedades reológicas y mecánicas específicas. La investigación sobre estos materiales cementicios con bajo contenido de carbono muestra que el cemento CSA proporciona un rápido desarrollo de resistencia, las mezclas con LC3 ofrecen beneficios ambientales e integridad estructural prometedores, y los geopolímeros logran una alta resistencia a la compresión, pero requieren optimización para su durabilidad. El presente estudio resume una revisión de los efectos de estos materiales cementicios sobre las propiedades en estado fresco, el rendimiento mecánico y la durabilidad del hormigón, enfatizando su potencial para su uso en 3DPC. Los hallazgos destacan la importancia de optimizar las propiedades de los materiales para mejorar el rendimiento y la sostenibilidad en la industria de la construcción.

Keywords: Cemento de sulfoaluminato de calcio; arcilla calcinada de piedra caliza; geopolímeros; imprimibilidad.

Corresponding author: iinavarr@uc.cl
Departamento Ingeniería y Gestión de la Construcción, Escuela de Ingeniería, Pontificia Universidad Católica de Chile, Santiago, Chile.

ISSN: 0718-5073

Vol. 39
Special Issue 2024
DOI: 10.7764/RIC.00131.21

1. Introduction

Concrete is the most widely produced material globally (Rashid et al., 2020), with production expected to grow from 14 billion m3 today to 20 billion m3 by 2050 (World Economic Forum, 2024). The key ingredients include a binder, aggregates, water, and chemical admixtures. The most frequently used binder is Ordinary Portland Cement (OPC), which consists of roughly 95% clinker and 5% gypsum by mass (Mehta and Monteiro, 2014). Clinker is created by heating a mixture of limestone (CaCO3) and clays rich in silicon, aluminum, and iron oxides in a rotary kiln (Kosmatka et al., 2011). Achieving the high temperatures necessary for clinker formation requires the combustion of fossil fuels. Combined with the decarbonation of CaCO3 during this process, the production of OPC becomes a major contributor to CO_2 emissions. In fact, the OPC production process is responsible for approximately 8% of global CO_2 emissions from human activities (Shao et al., 2024).

In response to these environmental concerns, the concrete industry has developed several innovations. Early efforts concentrated on improving energy efficiency in clinker production, and most modern kilns now operate close to their theoretical efficiency limits (Martínez et al., 2024); (Ramirez-Amaya et al., 2023); (Sharma et al., 2021). Another common practice has been to partially replace cement with supplementary cementitious materials (SCMs), like fly ash (Rivera et al., 2015), slag (Ustabaş and Kaya, 2018), and natural pozzolans (Rodríguez-Camacho and Uribe-Afif, 2002), and today, most concrete worldwide uses a combination of OPC and SCMs as binders (Mehta and Monteiro, 2014).

However, over the last two decades, further reductions in OPC content have stagnated, and it seems unlikely that additional reductions can be achieved using the currently available SCMs (Dhandapani et al., 2021); K. L. (Scrivener and Nonat, 2011). For these reasons, research has been growing into low-carbon binders. Among these, calcium sulfoaluminate (CSA) cement (Cau Dit Coumes et al., 2009); (Nie et al., 2022), limestone calcined clay (LC3) cement (Sharma et al., 2021); (Yılmaz and Ediz, 2008), and geopolymers (Abbas et al., 2022); (Álvarez-Ayuso et al., 2008) have shown promising results. The production of OPC requires temperatures of 1450°C, releasing about 0.54 tons of CO2 per ton of clinker produced. Conversely, CSA is produced at a lower temperature of 1250°C, resulting in a 50% reduction in CO2 emissions (Yuan et al., 2021). Similarly, an LC3 system composed of 50% Portland clinker, 15% limestone, 30% calcined clay, and 5% gypsum can reduce CO2 emissions by up to 40% and production costs by up to 30% compared to traditional OPC production (Sánchez Berriel et al., 2016). Additionally, calcined clay requires activation at lower temperatures (~800°C), significantly below the temperatures used in rotary kilns in cement plants. Although geopolymers are considered cement-free binders, studies have indicated that the activator solution used in their formulation substantially impacts CO_2 emission, directly influencing the environmental impact of the material (Bajpai et al., 2020); (Salas et al., 2018).

Beyond the environmental aspect, it is crucial to consider issues related to the scalability of production and market adoption of these alternative types of cement. Given these factors, attention has increasingly turned to LC3 types of cement, as they do not require significant modifications to existing cement plants and have a wide availability of raw materials such as clay and limestone. This contrasts with CSA cement, which requires bauxite reserves, and geopolymers, which typically use precursors like fly ash and blast furnace slag. It is well-known that these materials currently do not meet the global demand for composite cement (e.g., pozzolanic cement) and will likely fall short of meeting the needs for entirely alternative cement based on supplementary cementitious materials (K. Scrivener et al., 2018).

In addition to its significant contribution to global CO_2 emissions, the concrete construction industry is responsible for over 33% of all waste generated and consumes 40% of the world's energy (European Commission, 2020). Moreover, the sector faces numerous challenges tied to the green and digital transition, including an aging workforce and difficulties attracting younger workers with advanced digital skills (Bakhshi et al., 2024); (Kasztler et al., 2016); (Li et al., 2024).

In recent years, 3D printed concrete (3DPC) has gained significant attention for its potential to revolutionize the industry, improving productivity while reducing environmental impacts. 3DPC offers several advantages over traditional construction methods, such as minimizing workplace injuries, speeding up construction processes, reducing material waste, and allowing for greater architectural flexibility (Bhattacherjee et al., 2021); (De Schutter et al., 2018); (Mohan et al., 2021). These benefits could result in up to 805 reductions in costs, 60% reductions in material waste, and 70% reductions in construction time (Ahmed, 2023); (De Schutter et al., 2018); (Nodehi et al., 2022); (Wangler et al., 2019).

However, despite its promise, 3DPC is still an emerging technology facing several challenges before widespread adoption. One of the primary issues is that 3DPC mixtures require nearly double the amount of binder compared to traditional concrete to achieve the necessary fresh-state properties for printing (De Schutter et al., 2018); (Han et al., 2021); (Silvestro et al., 2024). To mitigate the environmental impact, the use of low-

ISSN: 0718-5073

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DOI: 10.7764/RIC.00131.21

carbon binders is a promising solution. Preliminary research has explored the use of LC3 types of cement (Al-Noaimat, Chougan, et al., 2023), CSA cement (Chen et al., 2020), and geopolymers (Al-Noaimat, Ghaffar, et al., 2023) in 3DCP.

This study aims to provide an overview of the effects of the most common low-carbon binders on the fresh properties, mechanical behavior, and durability of concrete. Additionally, it will evaluate the key benefits of incorporating low-carbon binders in 3DPC.

2. Fresh properties

The rheological behavior of fresh cementitious materials resembles that of yield stress fluids (Tattersall and Banfill, 1983), where the material only begins to flow once the applied shear stress surpasses its yield stress threshold (Feys et al., 2017). After this flow is initiated, the material's viscosity becomes the primary factor governing its behavior (Mahmoodzadeh and Chidiac, 2013). Additionally, fresh cementitious materials exhibit both dynamic and static yield stress (Perrot et al., 2012). Dynamic yield stress refers to the minimum stress needed to sustain flow, while static yield stress is the stress required to initiate flow (Qian and Kawashima, 2018). Notably, the static yield stress increases as the material remains at rest, a process known as structural build-up (Roussel et al., 2012).

This rheological behavior plays a critical role in determining the suitability of cementitious materials for 3D printing applications. Structural build-up significantly influences key factors such as buildability (Joh et al., 2020), interlayer strength (Hager et al., 2022), and the open time (Zhang et al., 2021) of 3DPC. Meanwhile, viscosity directly impacts extrudability and pumpability (Ma and Wang, 2018) for efficient 3D printing operations.

2.1 Calcium sulfoaluminate (CSA) cement

Limited research has been conducted on the rheological behavior of CSA cement mixtures. In a study by Chen et al. (Chen et al., 2020), the rheological properties of CSA and OPC pastes with similar water-to-binder ratios were compared. The results revealed that while both pastes exhibit similar dynamic yield stress, CSA pastes demonstrate significantly higher viscosity than OPC pastes (Silvestro et al., 2024). This increased viscosity can lead to challenges in compaction, potentially causing air entrapment during mixing and pumping when using CSA cement (Ke et al., 2020).

Additionally, CSA cement hydrates much faster than OPC, largely due to the presence of a phase called ye'elimite (Mohan et al., 2021). Unlike the primary phases of OPC, which experience an initial period of slow reaction due to the presence of gypsum (Schöler et al., 2017), ye'elimite does not exhibit such a delay (Bogner et al., 2020). In fact, it is not possible to control the ye'elimite hydration with gypsum as in the case of tricalcium aluminate (C3A) (Winnefeld and Lothenbach, 2010), gypsum actually accelerates ye'elimite hydration (Sahu et al., 1991). As a result, CSA cement has a shorter setting time compared to OPC (Wang et al., 2023).

These characteristics make CSA cements particularly well-suited for 3DPC, offering high buildability and rapid setting times. However, CSA cements also pose challenges in 3DPC applications, such as a significantly shorter open time, which can be mitigated with the use of retarders, and the need for higher pumping pressure due to its elevated viscosity.

2.2 Limestone calcined clay (LC3) cements

Previous studies (Mandal et al., 2023); (Muzenda et al., 2020); (Nair et al., 2020) have demonstrated that the use of LC3 types of cement reduces the workability of concrete. As a result, higher dosages of high-range water-reducing admixtures (HRWRA) are necessary for LC3 mixtures compared to those made with OPC (Mandal et al., 2023). Furthermore, the individual effects of calcined clay and limestone in LC3 have been thoroughly investigated (Muzenda et al., 2020). The inclusion of calcined clay has been found to increase dynamic yield stress, viscosity, and structural build-up, likely due to its high specific surface area and layered structure, which leads to a higher water demand. In contrast, the limestone fraction in LC3 types of cement tends to lower dynamic yield stress, viscosity, and structural build-up in the cementitious material.

Optimizing the rheological properties of LC3 mixtures for 3D printing typically involves the use of chemical admixtures (Al-Noaimat, Chougan, et al., 2023); (Chaves Figueiredo et al., 2019); (Chen et al., 2019); (Marchon et al., 2018). Most studies on 3DPC using LC3 types of cement have incorporated HRWRA and viscosity-modifying agents (VMA) (Al-Noaimat et al., 2023). HRWRA decreases both dynamic yield stress and viscosity, improving the mixture's pumpability and extrudability (Whiting and Dziedzic, 1992). VMAs further enhance rheological performance and reduce

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the pumping pressure needed for extrusion (Chen et al., 2020). However, its inclusion may reduce the open time of the mixture (Marchon et al., 2018).

Furthermore, the printability of LC3 mixtures is primarily influenced by the replacement ratio of OPC with LC3 cement (Al-Noaimat, Chougan, et al., 2023); (Long et al., 2021). A previous study (Long et al., 2021) demonstrated that increasing the OPC replacement level from 40 to 50% enhanced the buildability of the 3DPC mixture. However, this improvement alone was insufficient to maintain the structure's shape, resulting in significant deformations. The study further revealed that incorporating silica fume effectively enhanced the buildability of LC3 mixtures and helped retain the object’s shape, providing better structural integrity during the printing process. Moreover, silica fume exhibits pozzolanic activity, influencing hydration reactions, specifically the consumption of portlandite and the formation of additional C-S-H.

2.3 Geopolymers

Fresh behavior of geopolymers is influenced by interparticle interactions, particle solubility, and viscous interactions with alkaline activators (K. Chen et al., 2024). The elemental composition of the silico-aluminate material plays a key role in controlling the rheological behavior of geopolymer mixtures (Ilcan et al., 2023); (Kaze et al., 2022). Silico-aluminate materials generate substantial gelling material under alkaline activation, forming grid-like structures that require higher shear stress to break down. During shear, these structures disintegrate, reducing internal friction and lowering shear stress in the descending stage of the process.

Printability is also greatly influenced by the structural build-up of geopolymers (K. Chen et al., 2024). Generally, finely ground particles with a narrow size distribution further increase structural build-up (Navarrete et al., 2020). This property aids in shear thinning, improving extrudability and pumpability during 3D printing. Additionally, a proper structural build-up enhances interlayer bond strength and buildability. The type and concentration of alkali activators also impact the structural build-up, further influencing the geopolymer's suitability for 3D printing (Ilcan et al., 2023).

3. Mechanical behavior

3.1 Calcium sulfoaluminate (CSA) cement

CSA cement is characterized by its rapid strength development and excellent early-age mechanical performance. This accelerated strength gain is primarily attributed to the formation of ettringite, which facilitates the early densification of the microstructure during hydration and significantly impacts the fresh-state behavior, as previously discussed. Compared to OPC, CSA cements demonstrate significantly superior mechanical properties within the first 24 hours, making them particularly suitable for applications requiring quick setting times, such as structural repairs and precast concrete elements. Compressive strengths of approximately 40 MPa at 1 day have been reported for CSA-based mortars with a water-to-cement ratio 0.5 (García-Maté et al., 2015).

The high early strength of CSA also makes them highly suitable for 3D concrete printing, enabling faster layer deposition and construction. Their fast-setting properties and dimensional stability also ensure precision and prevent deformation during the printing process. An interesting approach for 3D printing cement-based materials is a binary system composed of OPC and CSA. For instance, (Khalil et al., 2017) found an optimal composition for 3D printing of 93% OPC and 7% CSA with a water-to-cement ratio of 0.35, sand-to-cement ratio of 2, and 0.26% of superplasticizer by binder weight, which resulted in the superposing more than 25 layers. The CSA small addition did not affect the early compressive strength of the material, although its incorporation modifies the material's stiffening behavior during the induction period, enhancing the 3D printability. Wang et al. (2021) also reported that increasing the CSA content in binary compositions with OPC results in a shorter setting time and, therefore better buildability. Moreover, the mixture with 80% CSA cement showed higher early strength and 1d compressive strength higher than 30 MPa.

3.2 Limestone calcined clay (LC3) cements

In the field of LC3 types of cement, one of the pioneering studies was conducted by Antoni et al. (2012). Their results demonstrate that the combined substitution of metakaolin and limestone yields excellent mechanical performance. Specifically, a mixture containing up to 45% substitution of Portland cement (PC) with a 2:1 ratio of metakaolin to limestone exhibited superior mechanical properties at both 7 and 28 days, compared to OPC. Scrivener et al. (2018) further indicated that a kaolinite content of 40%, in a system where 50% of OPC is replaced by 30% calcined clay, 15% limestone, and 5% gypsum, is necessary to achieve mechanical properties comparable to OPC at 7 days. However, more recent studies have shown that calcined clays with lower kaolinite content (≤ 25.0 wt.%) can still replace up to 30% of OPC by weight while maintaining

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similar mechanical performance to OPC formulations (Cardinaud et al., 2021). Additionally, emerging research suggests that, beyond kaolinite content, the presence of impurities such as iron in these materials contributes to a higher specific surface area, which significantly influences hydration kinetics and, consequently, the development of compressive strength in these types of cement (Matschei et al., 2023).

Regarding applying this type of alternative cement for 3D printing, research is still in its early stages. (Jin et al., 2024) suggest that LC3 is a promising solution for mitigating greenhouse gas emissions in the context of 3D concrete printing, as it offers both mechanical strength and an improved structuration rate. The authors also highlight that special attention must be given to the location of calcined clay plants, particularly concerning the calcined clay content. Wang et al. (2024) assessed LC3-based engineered cementitious composites (ECC) for 3D printing applications. The authors reported that the pore structure of the printed LC3-ECC is denser than that of the cast LC3-ECC, which resulted in mechanical properties significantly higher than those of the cast specimens. Furthermore, the authors draw attention to the anisotropy of flexural and compressive strength, especially during the later stages of the curing process.

3.3 Geopolymers

Geopolymeric binders are produced by activating aluminosilicate materials with an alkaline solution. Depending on the precursors used, usually materials such as fly ash or blast furnace slag, and the alkaline activator employed, these materials can achieve compressive strengths comparable to or even exceeding those of Portland cement. Moreover, the composition of geopolymers allows for incorporating various industrial by-products, providing a sustainable disposal solution for these wastes. Recent studies have explored using binary, ternary, and quaternary blends of waste materials for geopolymer production (Ruviaro et al., 2024). This highlights the broad potential for combining different residues in developing geopolymers, contributing to creating more environmentally sustainable construction materials.

Regarding 3D-printed geopolymer, its compressive strength is influenced by several factors, including the quality of the materials used for printing, the specific printing conditions, and the curing process employed. In general, 3D-printed geopolymer concrete exhibits compressive strengths ranging from 20 to 70 MPa, with typical values around 30 MPa (Shilar et al., 2023). Chen et al. (K. Chen et al., 2024) reviewed works concerning the mechanical properties of 3D printing geopolymers, reporting values within the range of 20 MPa to 56.8 MPa.

4. Durability

The durability of 3D-printed materials is a growing concern, particularly for long-term applications. While efforts to reinforce 3D-printed materials with traditional steel rebar are ongoing, challenges such as shrinkage cracking, reduced carbonation resistance, and vulnerability to chloride ingress remain significant. These durability issues are especially pronounced due to the permeability at the interfaces between layers. Additionally, the increased risk of creep, delamination under high temperatures, and susceptibility to chemical attacks further exacerbate these concerns, with variations in durability depending on the direction of load and environmental exposure. These problems are primarily associated with the higher porosity and presence of microcracks at layer interfaces (Ler et al., 2024).

Wang et al. (2021) investigated the drying shrinkage and carbonation resistance of cementitious composites of CSA and OPC. The results demonstrated that CSA effectively enhanced the drying shrinkage resistance. A blend of 80% CSA and 20% OPC exhibited 60% lower drying shrinkage. Nevertheless, the compositions with higher OPC content demonstrated greater resistance to carbonation, which can be attributed to their denser microstructures and higher alkalinity. For example, the mixture containing 80% OPC + 20% CSA exhibited a carbonation depth of only 3.5 mm after 28 days of CO_2 exposure, compared to a depth of 11.0 mm for a composition with 20% OPC and 80% CSA under the same conditions.

In terms of evaluating the durability of LC3-based materials for 3D printing, no studies have yet been found in the literature, highlighting a broad area for future research. However, general studies on the durability of these binders for other applications reported that LC3 has shown enhanced resistance to sulfate attack, making it well-suited for use in environments with elevated sulfate concentrations and an increased service life against chloride ingress (Barbhuiya et al., 2023). However, raising concerns regarding LC3 carbonation resistance was highlighted in previous research and should be further investigated (Y. Wang et al., 2023).

Research on the durability of 3D-printed geopolymers remains limited. As with OPC-based printed materials, the permeability at layer interfaces is a key factor influencing the durability of 3D-printed geopolymer structures (Raza et al., 2022). Mermerdaş et al. (2024) evaluated the effects of the sodium silicate-to-sodium hydroxide ratio and molarity on the durability of mold-cast and 3D-printed geopolymers, including water

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absorption, sorptivity index, and chloride penetration test. The authors reported that the durability of 3D-printed specimens was inferior to those of mold-cast specimens, largely due to weak interfacial bonding between the printed layers. The study also demonstrated that increasing the sodium hydroxide molarity positively influenced both the strength and durability of the composites, suggesting that optimizing this parameter can help improve the performance of 3D-printed materials.

5. Conclusions

The effect of the most used low-carbon binders (i.e., calcium sulfoaluminate cement (CSA), limestone calcined clay (LC3) cement, and geopolymers) on the fresh properties, mechanical performance and durability of concrete, and their applicability for 3D printing are reviewed in this paper. Important conclusions from this work are listed below:

1. The composition and particle size of low-carbon binders significantly affect the rheological properties of cementitious mixtures. These materials influence viscosity and structural build-up of mixtures, which are critical for achieving optimal 3D printing performance.
2. CSA cement provides rapid strength development and dimensional stability, making it suitable for 3D concrete printing. Its fast setting and high early strength enable efficient layer deposition, though a higher CSA content may reduce setting time, enhancing buildability.
3. LC3 cements combining metakaolin and limestone, offers excellent mechanical performance and sustainability. While still in early research for 3D printing, LC3 mixtures show promise in reducing greenhouse gas emissions and providing improved structuration rates with denser pore structures.
4. Geopolymeric binder offers compressive strengths comparable to or exceeding OPC. 3D printed geopolymer concrete exhibits compressive strengths ranging from 20 to 70 MPa, depending on printing and curing conditions.
5. Although considered more sustainable, types of cement such as LC3, CSA, and geopolymers face certain challenges related to durability, particularly carbonation for CSA and LC3. Specifically for applications in 3D-printed structures, each material presents unique advantages and limitations, with permeability at layer interfaces being a critical durability issue for this type of application.

6. Acknowledgment

The authors acknowledges the funding provided by CNPq/MCTI Call No. 10/2023 and ANID/CONICYT FONDECYT Iniciación 11240179.

7. Notes on Contributors

Laura Silvestro, Federal University of Technology – Paraná (UTFPR), Guarapuava, Brasil. **ORCID** https://orcid.org/0000-0002-6437-3047	**Rodrigo Scoczynski Ribeiro,** Federal University of Technology – Paraná (UTFPR), Guarapuava, Brasil. **ORCID** https://orcid.org/0000-0002-3409-2041
Iván Navarrete, Departamento Ingeniería y Gestión de la Construcción, Escuela de Ingeniería, Pontificia Universidad Católica de Chile, Santiago, Chile. **ORCID** https://orcid.org/0000-0002-2918-7900	

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ISSN: 0718-5073