

## Durability and sustainability performance of Portland limestone cements.

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### ABSTRACT

This paper reviews the mechanical, microstructural, and durability performance of Portland-limestone cement (PLC) as a sustainable alternative to traditional Portland cement. A systematic analysis of literature spanning almost five decades was conducted, focusing on compressive strength, porosity, chloride ingress, and carbonation in mortars and concretes. Results indicate that PLC generally provides adequate early-age strength, while porosity, chloride diffusion, and carbonation increase with higher LS content; these effects can be mitigated by optimizing water-to-cement ratio and incorporating supplementary cementitious materials. Limitations include inconsistent experimental protocols, variable curing durations, and limited long-term data. The originality lies in synthesizing PLC performance across multiple durability metrics and highlighting research gaps. Overall, the study emphasizes the need for integrated, performance-based evaluation to ensure sustainable and durable concrete applications.

**Keywords:** Portland limestone cement; compressive strength; porosity; chloride ingress; carbonat.

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### Contribution of each author

In this work, author DMSC contributed with the activities: original ideas, data collection, results, results discussion, and writing of the work. Author AATA contributed with activities: original ideas, data collection, results discussion, and writing review/editing. Author BMP contributed with activities: original ideas, data collection, results discussion, and writing review/editing.

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## Desempeño en durabilidad y sostenibilidad de cementos Portland con remplazo de caliza.

### RESUMEN

Este trabajo hace una revisión del desempeño mecánico, microestructural y de durabilidad del cemento Portland-caliza (PLC) como alternativa sostenible al cemento Portland tradicional. Se realizó un análisis sistemático de la literatura de casi cinco décadas, centrándose en resistencia a compresión, porosidad, ingreso de cloruros y carbonatación en morteros y concretos. Los resultados indican que el PLC proporciona generalmente una resistencia adecuada a edades tempranas, mientras que la porosidad, la difusión de cloruros y la carbonatación aumentan con mayores contenidos de caliza; estos efectos pueden mitigarse optimizando la relación agua-cemento e incorporando materiales puzolánicos. Las limitaciones incluyen protocolos experimentales inconsistentes, variaciones en los tiempos de curado y escasez de datos a largo plazo. La originalidad radica en sintetizar el desempeño del PLC considerando múltiples métricas de durabilidad y en resaltar brechas de investigación. En general, se enfatiza la necesidad de evaluaciones integradas y basadas en desempeño para asegurar aplicaciones de concreto sostenibles y duraderas.

**Palabras clave:** cemento Portland con caliza; resistencia a la compresión; porosidad; penetración de cloruros; carbonatación.

### Desempenho em durabilidade e sustentabilidade de cimentos Portland com substituição de calcário.

### RESUMO

Este artigo faz a resenha o desempenho mecânico, microestrutural e de durabilidade do cimento Portland-calcário (PLC) como alternativa sustentável ao cimento Portland tradicional. Foi realizada uma análise sistemática da literatura de quase cinco décadas, com foco na resistência à compressão, porosidade, entrada de cloretos e carbonatação em argamassas e concretos. Os resultados indicam que o PLC geralmente proporciona resistência adequada em idades precoces, enquanto a porosidade, a difusão de cloretos e a carbonatação aumentam com maiores teores de calcário; esses efeitos podem ser mitigados otimizando a relação água-cimento e incorporando materiais pozolânicos. As limitações incluem protocolos experimentais inconsistentes, variações nos tempos de cura e escassez de dados de longo prazo. A originalidade reside em sintetizar o desempenho do PLC considerando múltiplas métricas de durabilidade e em destacar lacunas de investigação. Em geral, enfatiza-se a necessidade de avaliações integradas e baseadas no desempenho para garantir aplicações de betão sustentáveis e duradouras.

**Palavras-chave:** cimento Portland com calcário; resistência à compressão; porosidade; ingresso de cloretos; carbonatação.

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## 1. INTRODUCTION

Portland Limestone Cement (PLC) has seen widespread international adoption as a sustainable alternative to Ordinary Portland Cement (OPC), primarily because it can reduce CO<sub>2</sub> emissions by partially substituting clinker with finely ground limestone (LS). Initially limited to modest replacement levels of 5–15% (ASTM International, 2022; CSA, 2003), several countries and regulatory bodies have expanded the allowable substitution level to 35% (ONNCCE, 2017), further promoting PLC's market presence. While the environmental benefits of reduced clinker content are compelling, these assessments often overlook the broader implications of PLC's performance in reinforced concrete systems, where the concrete-steel interaction governs long-term durability. Concrete efficiency must be evaluated not only at the binder level but also within the context of the complete composite material, especially in reinforced concrete applications. Increased LS content often results in higher porosity and permeability (Courard et al., 2005; Palm et al., 2016), which not only affects the concrete matrix but also compromises the protective environment around the embedded reinforcing steel. This is a significant concern, as reinforcement corrosion remains one of the most critical degradation mechanisms in reinforced concrete structures worldwide (Elgalhud et al., 2017). Although PLC is promoted for its lower CO<sub>2</sub> emissions, those benefits are uncertain if it compromises the durability of reinforcing steel. This is especially relevant given that steel production emits approximately twice as much CO<sub>2</sub> per ton as clinker (Elhacham et al., 2020). Therefore, any reduction in clinker emissions may be outweighed by premature corrosion-induced repairs or structural maintenance.

Moreover, PLC production typically requires finer grinding to achieve comparable performance, thereby increasing energy demand during manufacturing. Additionally, PLC systems often depend more heavily on chemical admixtures to maintain workability and strength development, especially at higher LS contents. These factors contribute to hidden environmental costs that are frequently excluded from basic CO<sub>2</sub> accounting.

From a materials science standpoint, the aging performance of PLC systems differs notably from that of OPC-based ones, and even more so from systems incorporating supplementary cementitious materials (SCMs). When subjected to varying water-to-cement (w/c) ratios, PLC concretes often exhibit a slower development of mechanical and durability-related properties over time. This trend is particularly evident in low w/c systems, where the development of properties occurs at a slower pace. Conversely, in high w/c mixtures, performance improvements tend to plateau early or even stagnate, resulting in limited long-term enhancement. This disparity in the evolution of key properties complicates the assessment of PLC's long-term durability and sustainability, especially in real-world applications where environmental exposures and reinforcement corrosion are critical concerns.

## 2. HISTORICAL EVALUATION OF CEMENTS WITH LS REPLACEMENT

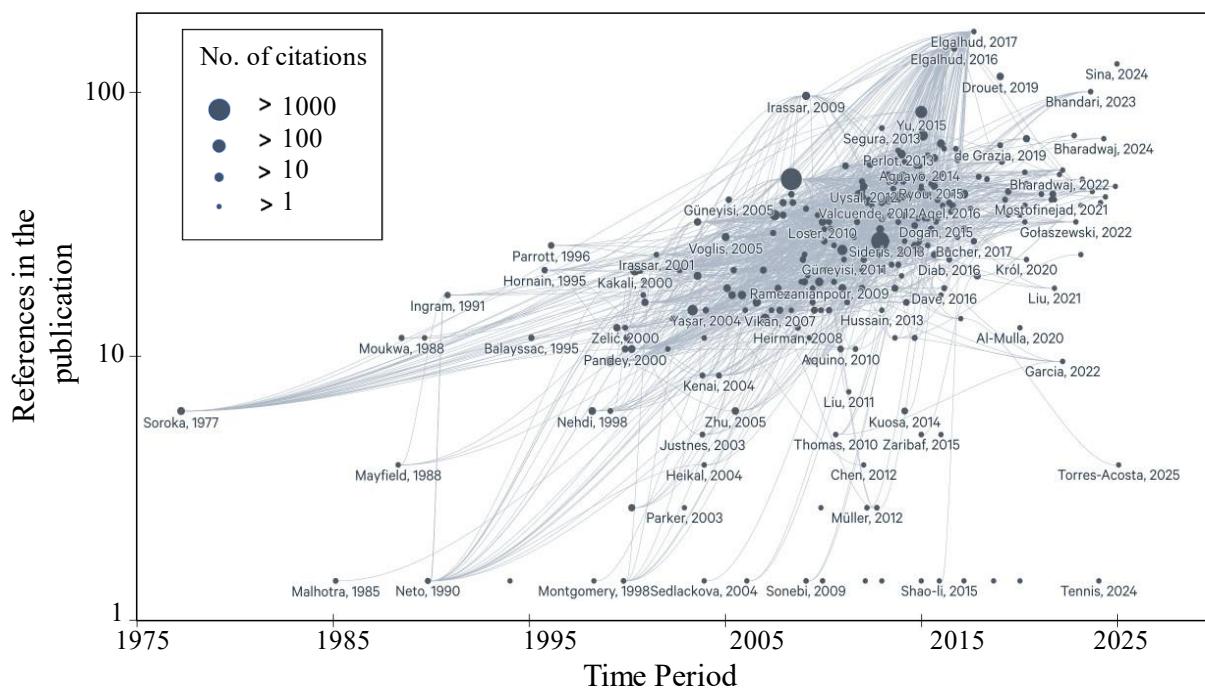
The interaction between calcium carbonate (CaCO<sub>3</sub>) and cement hydration products has been under investigation for nearly nine decades, with one of the earliest mechanistic hypotheses proposed by Bessey in 1938 regarding the formation of calcium carboaluminates in the presence of CaCO<sub>3</sub>; this hypothesis was documented by Soroka and Setter in 1977 (Soroka and Setter, 1977). The authors noted that the formation of calcium carboaluminates through the reaction of CaCO<sub>3</sub> with aluminate phases such as C<sub>3</sub>A and C<sub>4</sub>AF had been increasingly substantiated by experimental data in the preceding decades. Since that foundational period, the field has seen significant advancement, particularly in the characterization of the chemical and physical properties of PLCs, across varying LS replacement levels and water-to-cement ratios.

The scope of these previous studies has traditionally focused on fresh-state rheology, hydration

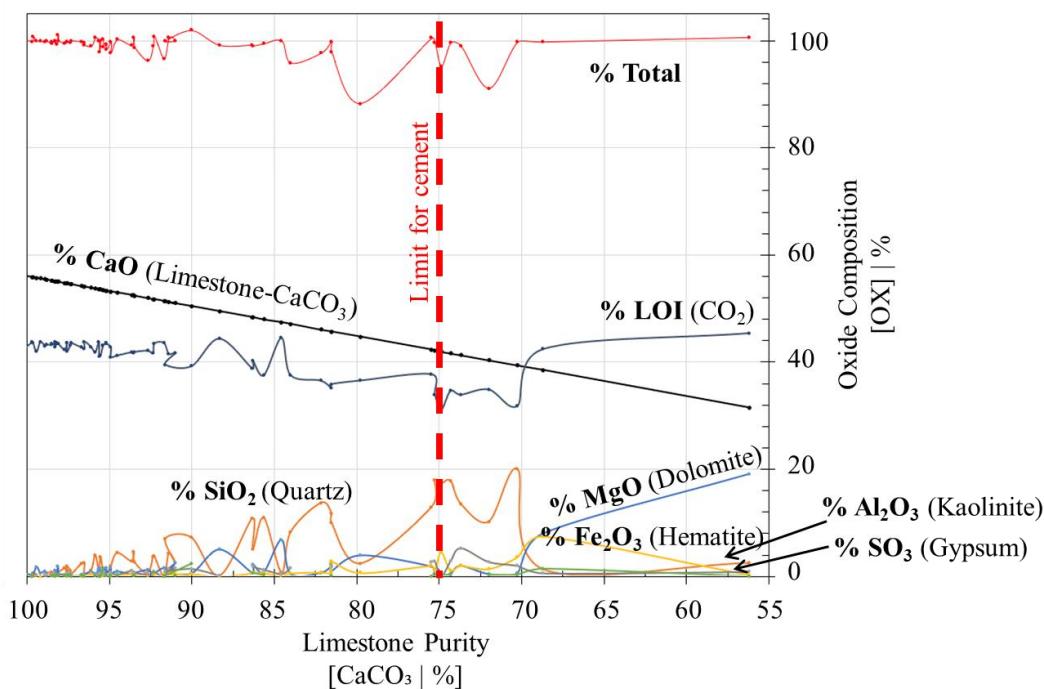
kinetics, and mechanical performance—primarily compressive strength at 28 days (Soroka, I., and N. Setter, 1977; Neto, Claudio S., and Vicente C. Campiteli, 1990; Adams, Lawrence D., and Ronald M. Race, 1990; Nehdi et al., 1996). However, the exploration of long-term durability metrics has historically remained comparatively limited. Critical aspects such as chloride ingress, carbonation resistance, freeze-thaw performance, and internal swelling reactions have only recently garnered broader research attention. This shift in focus has become particularly relevant in the past five years, as regulatory frameworks and sustainability agendas in several countries have accelerated the adoption of PLCs as the baseline binder system in structural concretes. Despite 87 years since the earliest mechanistic proposals and more than 49 years of systematic research following Soroka and Setter in 1977, the long-term durability of PLCs remains insufficiently established, and a broad international consensus on their extended-age performance has yet to emerge.

Building on this unresolved gap in long-term performance knowledge, the present review undertakes a structured effort to consolidate and interpret the existing evidence base on LS-containing cementitious systems. To address the persistent fragmentation of results across decades, mixtures, and testing protocols, a comprehensive bibliometric database was assembled (Figure 1), encompassing more than 200 peer-reviewed publications and institutional reports that directly examine LS incorporation in cement binders or concrete applications. This collection spans studies on pastes, mortars, and concretes, includes a wide range of replacement levels, finenesses, and clinker chemistries, and covers both mechanical performance and durability-related metrics. By systematically analyzing the historical data contained within this body of literature, the aim is to identify areas of agreement, clarify points of divergence, and ultimately establish a coherent interpretation of how LS-containing cements perform across the breadth of modern structural applications.

The first stage of the global bibliometric analysis effort involved establishing a unified chemical baseline for all retrieved publications, beginning with a systematic evaluation of the LS powders' oxide composition derived from XRF data. This variable was prioritized as the initial point of analysis because it represents the most consistently reported material property across the literature and provides a critical reference frame for interpreting the broad spectrum of performance outcomes. The resulting chemical mapping in Figure 2 reveals substantial variability in  $\text{CaCO}_3$  purity, impurity profiles, and overall oxide distributions among the studied LSs—factors that, although often underemphasized, may exert influence on hydration mechanisms, microstructural development, and durability behavior.



**Figure 1.** Node-based network representation of the comprehensive bibliometric database compiled for studies on LS in cementitious systems.



**Figure 2.** Chemical composition from the database compiled for studies on LS in cementitious systems.

Given the magnitude and diversity of publications captured in the bibliometric dataset, a fully exhaustive examination of every study falls beyond the scope of a single review. Moreover, the accelerating global demand for concrete systems that meet both durability and sustainability imperatives underscores the need for a focused and analytically coherent synthesis. Accordingly, this review adopts a selective but methodologically guided approach, concentrating on a curated

set of representative studies that directly investigate the durability-relevant variables most critical to LS-containing cementitious systems. The selection criteria prioritize research with clearly reported material characterizations, controlled mixture parameters, and explicit evaluation of performance indicators across relevant time scales. These representative investigations form the analytical foundation for the discussions that follow, enabling a structured synthesis of the current state of knowledge regarding mechanical and durability responses in PLC-based materials.

Building on this curated foundation, the review critically examines three interrelated domains essential to assessing the durability performance of PLCs: carbonation depth, porosity evolution, and chloride transport mechanisms, including both non-steady-state migration and bulk diffusion approaches. These topics have emerged as central to understanding the long-term behavior of LS-containing concretes under aggressive environmental exposures. However, the existing body of literature exhibits substantial heterogeneity in chemical composition as shown in Figure 2, physical properties, sample types (paste, mortar, or concrete), curing regimes, water-to-cement ratios, and LS replacement levels. Such variability complicates direct comparison of findings and poses significant challenges to developing broadly applicable conclusions.

Within this context, the following sections synthesize the state-of-the-art on the influence of LS replacement on compressive strength development, porosity characteristics, resistance to chloride ingress, and carbonation behavior. In addition to consolidating experimental findings to date, the review seeks to delineate persistent inconsistencies and identify critical gaps in durability research. Together, these analyses underscore the need for a comprehensive, data-driven framework that supports predictive model development and guides future experimental design for LS-based binder systems.

### 3. PURITY OF LS ACROSS STUDIES

As a first step toward establishing a consistent chemical baseline, LS purity across the collected studies was quantified through oxide compositions reported by X-ray fluorescence (XRF) in these studies: Ahmad et al. (2020), Ali et al. (2013), Barker and Hobbs, (1999), Burgos Montes (2012), Burgos Montes (2013), Cam et al. (2010), Celik et al. (2015), Chen et al. (2012), Da Silva and De Brito (2016), De Weerdt et al. (2011), Dhir et al. (2007), El-Didamony et al. (1994), Elkhadiri et al. (2002), Ferreiro et al. (2018), Ghrici et al. (2007), Golaszeski (2022), Guemmadi et al. (2008), Hartshorn et al. (1999), Hartshorn et al. (2002), Heikal et al. (2000), Jin et al. (2024), Kakalis et al. (2000), Li et al. (2019), Li et al. (2020), Liu and Yan (2010), Liu et al. (2021), Lothenbach et al. (2008), Machner et al. (2017), Malhotra and Garette (1985), Marzouki et al. (2013), Meddah et al. (2014), Moir et al. (1999), Montgomery et al. (1998), Moukwa (1989), Pandey and Sharma (2000), Rahhal et al. (2012), Soroka and Setter (1976), Torresan et al. (2000), Tsivilis (1999), Tsivilis (2000), Tsivilis (2003), Yilmaz and Olgun (2008), and Zhang et al. (2000). For comparability, all CaO% values were converted to equivalent CaCO<sub>3</sub>% content using molar mass balance, assuming that all CaO% present originates from calcium carbonate. This normalization enables cross-study interpretation independent of reporting format and permits direct comparison of LS powders used across five decades of experimental work.

When plotted across the bibliometric dataset (Figure 2), the resulting CaCO<sub>3</sub> distribution reveals three distinguishable purity categories. The first corresponds to high-grade LSs (> 92% CaCO<sub>3</sub>), in which no major impurity oxides appear in significant proportion; these samples align with materials generally classified in industry as high-purity fillers suitable for performance-driven cement formulations. A second category encompasses LSs where XRF profiles show recognizable secondary oxides. Within this group, two dominant impurity types are observed: a) silica-bearing LSs, where elevated SiO<sub>2</sub> suggests quartz, and b) MgO-bearing LSs frequently associated with dolomitic phases. Although dolomite is the most plausible carrier of MgO in this dataset, the presence of other magnesium-bearing minerals (e.g., magnesite) or minor accessory phases such

as magnetite cannot be excluded without complementary mineralogical characterization (XRD, TGA, or SEM-EDS). These chemically hybrid LSs, together with high-purity LS, constitute a considerable proportion of the literature.

A third and less frequently acknowledged class includes low-purity LSs approaching the lower limit defined in ASTM C150/C595, which permits up to 25% non-calcium-carbonate minerals (i.e., a minimum ~75%  $\text{CaCO}_3$  purity).

Together, these observations illustrate that LS used in cement systems cannot be treated as chemically uniform. Variability in  $\text{CaCO}_3$  content, impurity profiles, and mineralogical associations introduces a significant source of divergence within the literature. Establishing LS purity through XRF-based  $\text{CaCO}_3$  quantification, therefore, provides not only a consistent reference for bibliometric synthesis but also a necessary framework.

#### 4. EFFECT ON COMPRESSIVE STRENGTH

This section presents the historical review of the compressive strength performance in LS-containing cements as a function of curing time and w/c ratio. The reviewed studies show that PLCs typically exhibit accelerated early-age strength performance compared to OPC and SCMs blended cements. However, this initial advantage tends to diminish over time as the material ages. At lower w/c ratios, PLCs may continue to gain strength, but at a slower pace than OPC, while at higher w/c ratios, strength development in PLCs tends to plateau, a performance that aligns partially with that of traditional OPC systems. A critical observation is that, at extended curing ages, OPC frequently surpasses PLC in compressive strength, suggesting a shift in performance trajectory beyond the early stages. It is important to note that although factors such as aggregate size, Blaine fineness, sample shape, and air entrainment may influence strength, the current review primarily focuses on cement content, water content, and curing duration as the main variables.

The studies by Malhotra and Carette (1985), Nedhi and Mindess (1986), Montgomery et al. (1998), Moir and Kelham (1999), Tsivilis et al. (1999), Heikal et al. (2000), Irassar et al. (2006), Dhir et al. (2007), Pourkhorshidi et al. (2008), Thomas et al. (2010), De Weerdt (2011), Sezer (2012), Chen et al. (2012), Neithalath and Cam (2012), Githachauri (2012), Diab et al. (2015), and Ahmad (2022) comprise a variety of sample types, including cement pastes, mortars, and concretes. While each of these materials exhibits distinct fracture mechanics and microstructural characteristics, due to the presence or absence of aggregates and differences in interfacial transition zones, such distinctions are not consistently specified across all source studies with respect to the material's age. This differentiation is critical, particularly when analyzing mechanical properties like compressive strength, as the scale and composite behavior influence stress distribution and failure mechanisms.

The earliest study on concrete samples with LS addition to the mix and sand replacement was conducted by Malhotra and Carette (1985). The study reported clinker phases of the OPC sample ( $\text{C}_3\text{S} = 53.8\%$ ,  $\text{C}_2\text{S} = 19.6\%$ ,  $\text{C}_3\text{A} = 6.5\%$ ,  $\text{C}_4\text{AF} = 8.6\%$ ), the w/c ratio (0.4, 0.53, and 0.7), the LS replacement with respect to sand (ranging from 0 to 20%), and the superplasticizer (ranging from 0 to 1.4% by cement mass) in the mixture design. The properties of the fresh concrete reported were slumps from 60 to 90 mm, densities from 2,295 to 2,340 kg/m<sup>3</sup>, and air content from 4.6 to 6.2%. The results for the 0.40 w/c ratio indicate that the control mix would surpass the strength of the 20% LS dust mix between 28 and 90 days, reaching approximately 43 MPa. For the 0.53 and 0.70 w/c ratios, the 20% replacement would not exceed the strength of the OPC reference mix.

Nedhi and Mindess (1986) studied mortar samples based on OPC, LS, and silica fume with a w/c ratio of 0.5 over a 90-day period. The clinker composition was  $\text{C}_3\text{S} = 51\%$ ,  $\text{C}_2\text{S} = 23\%$ ,  $\text{C}_3\text{A} = 7\%$ , and  $\text{C}_4\text{AF} = 10\%$ . Silica fume replacement was 10%, while LS replacement was 6.3% or 18.8%. Only the comparison between the reference and the cement + LS mixes is analyzed. Their results show that the mix with the highest strength was OPC + 10% SF + 6.3% LS, while the mix with the

lowest strength was OPC + 18.8% LS.

Montgomery et al. (1998) studied both mortar and high-performance concrete samples using two different cement types and three LS fillers. Cement PC1 had a composition of  $C_3S = 63\%$ ,  $C_2S = 12\%$ ,  $C_3A = 2.5\%$ , and  $C_4AF = 17\%$ , while PC2 had  $C_3S = 64\%$ ,  $C_2S = 11\%$ ,  $C_3A = 4.5\%$ , and  $C_4AF = 14\%$ . For mortar mixes, the LS content ranged from 0 to 20%. At 28 days, the highest strength was obtained with mix M25, with 0% LS using PC2, whereas the lowest strength was for mix M6, which contained 20% LS with PC1. For the high-performance concrete mixes, the water-to-binder ratio ranged from 0.30 to 0.35, the cement content from 439 to 488 kg/m<sup>3</sup>, and the LS content was 10% for all mixes except the reference. The highest strength at 28 days was observed in mix CC3A with 90.1 MPa, which had a w/b of 0.30 and 10% LS. The lowest strength (73.4 MPa) was recorded for mix CC2 with a w/b of 0.35 and 10% LS.

Moir and Kelham (1999) evaluated different SCMs and Portland cements with 20% LS replacement. The OPC composition was  $C_3S = 55\%$ ,  $C_2S = 23\%$ ,  $C_3A = 6\%$ , and  $C_4AF = 11\%$ , while the mineralized cement had  $C_3S = 68\%$ ,  $C_2S = 7\%$ ,  $C_3A = 7\%$ , and  $C_4AF = 10\%$ . The study covered ages from 3 to 90 days. The results showed that the control reference samples outperformed all LS-containing mixes at every age.

Tsivilis et al. (1999) examined cement paste samples using two clinkers, each combined with 5% gypsum to produce four cements with Blaine fineness values ranging from 2,800 to 3,900 cm<sup>2</sup>/g. Clinker C1 consisted of  $C_3S = 65.15\%$ ,  $C_2S = 13.32\%$ ,  $C_3A = 7.54\%$ , and  $C_4AF = 10.92\%$ ; while clinker C2 consisted of  $C_3S = 57.99\%$ ,  $C_2S = 18.60\%$ ,  $C_3A = 11.74\%$ , and  $C_4AF = 6.26\%$ . LS replacement ranged from 0 to 35%. For cements C1L1, the highest strength occurred at 5% LS replacement with a Blaine fineness of 3,490 cm<sup>2</sup>/g, while the lowest strength occurred at 35% replacement with a Blaine of 5,150 cm<sup>2</sup>/g. For C1L2, the highest strength occurred at 10% LS and 3,900 cm<sup>2</sup>/g, and the lowest at 35% LS and 5,080 cm<sup>2</sup>/g.

Heikal et al. (2000) studied cement pastes with a constant Blaine fineness of 3,100 cm<sup>2</sup>/g composed of OPC, Homra, and LS during a period of 100 days. OPC was fixed at 80%, while Homra and LS varied from 0 to 20%. Only the reference versus LS mixes are analyzed. The cement with 0% LS achieved the highest compressive strength, and strength decreased as LS replacement increased across all ages.

Irassar et al. (2006) evaluated compressive strength using concrete cylinders (100×200 mm) from 3 to 360 days. All mixtures used a water-to-cementing materials ratio (w/cm) of 0.50, a cementitious material content of  $350 \pm 10$  kg/m<sup>3</sup>, and a sand-to-total-aggregate ratio of 0.45. The slump was  $70 \pm 20$  mm, and LS replacement levels were 0%, 12%, and 18%. At 28 days, the compressive strengths were similar (36.0, 34.5, and 35.2 MPa). At 360 days, both LS concretes showed strength reductions of up to 8%.

Dhir et al. (2007) examined cement combinations including a 100% PC reference (0% LS), 85% PC–15% LS (Type II/A-L range), 75% PC–25% LS, 65% PC–35% LS (Type II/B-L range), and 55% PC–45% LS. Aggregates were crushed granite (20–10 mm and 10–5 mm) and natural sand. PC concretes were proportioned to achieve cement contents from 235 to 410 kg/m<sup>3</sup> at 40–50 kg/m<sup>3</sup> increments, with a fixed water content of 185 L/m<sup>3</sup> (w/c = 0.45–0.79) and a slump of 75 mm. Coarse aggregate content was fixed at 800 and 400 kg/m<sup>3</sup>, while fine aggregate content was adjusted to maintain yield. Air-entraining admixture was used to achieve  $5.0 \pm 0.5\%$  air. Water-to-cement ratios tested were 0.46, 0.54, 0.62, 0.67, and 0.79. The reference mix with 0% LS had the highest strength at all ages and at all w/c ratios.

Pourkhorshidi et al. (2008) studied the mixture design and proportions of OPC concrete and PLC concrete using two cement categories: ASTM Type II Portland cement and Type II/A-LL PLC. The compressive strength was measured using 150-mm concrete cubes, and the specimens were tested after 7, 28, and 90 days of curing. The PLC specimens had 14% LS. The slump ranged from 55 to 75 mm, the densities from 2,295 to 2,330 kg/m<sup>3</sup>, and the air content from 2.6 to 2.8%. The water-to-cement ratio varied from 0.35 to 0.55. The cement content was 350 kg/m<sup>3</sup>, with fine

aggregates at 960 kg/m<sup>3</sup> and coarse aggregates at 850 kg/m<sup>3</sup>. The PLC mix with a w/c ratio of 0.35 had higher compressive strength than the reference with 0.45; the rest of the mixes (PLC with 0.45 and 0.55 w/c ratio) had a lower strength at all ages.

Thomas et al. (2010) reported field results from Quebec, Alberta, and Nova Scotia. Since all cements used in Nova Scotia incorporated SCMs, no analysis was conducted for this location. Only the results for the reference mixes—including those containing a small amount of LS and the GUL cement—are discussed (the cements with SCMs are not included in the analysis). The total cementitious materials content for all mixtures was 355 kg/m<sup>3</sup>, and the w/cm ranged from 0.44 to 0.45. This corresponds to approximately 91 to 92% clinker for the control mix produced with PC and no SCMs (PC contains about 3 to 4% LS and 5% gypsum). Type GU cement used in Quebec and Alberta contains 3–4% LS and 91% clinker, while Type GUL cement used in the same regions contains 12% LS and 83% clinker. For Quebec, the GU cement had a Blaine fineness of 373 m<sup>2</sup>/kg and the GUL had 453 m<sup>2</sup>/kg. For Alberta, the GU had a Blaine fineness of 399 m<sup>2</sup>/kg and the GUL had 510 m<sup>2</sup>/kg. The results at the Quebec site showed that the PLC had a higher strength than PC at 3, 7, and 28 days, while the PC-based mixture had the highest strength, surpassing the PLC mixture at 56 days. For the field data in Alberta, the GUL-0 had a better performance across all ages than with respect to GU-0.

De Weerdt et al. (2011) investigated composite cements in which the LS powder was ground separately. The clinker interground with 3.7% gypsum by mass is referred to as OPC. The experimental matrix consisted of a reference mix: 100% OPC (mix 1), and subsequently, OPC was gradually replaced by LS powder in 5% increments up to 35% (mixes 2–8). For each testing age and material combination, three mortar prisms (40×40×160 mm) were prepared. In addition, corresponding cement pastes with a water-to-binder ratio of 0.5 were produced for all mixes using a Renfert vacuum mixer. Hydration of the samples stopped after 1, 28, 90, and 140 days. The results showed that the compressive strength of the LS-blended cements decreased with increasing LS powder replacement at all ages from 1 to 140 days. The main compressive strength gain occurred during the first 28 days of curing. The compressive strength did not appear to change significantly after 28 and 90 days of curing, respectively.

Sezer (2012) investigated the effect of silica fume and LS on the compressive strength of mortars. Mortar mixes containing LS at 5%, 20%, and 35% by weight of cement were prepared using CEM I 42.5 type normal Portland cement. The Blaine surface area of the LS was 420 m<sup>2</sup>/kg. Compressive strengths were measured at 2, 7, 28, 90, and 180 days. For the tests, 50×50×50 mm cube mortar specimens were produced using 450 g of cement, 1,350 g of standard sand, and a water-to-binder ratio of 0.5. Only the results comparing the reference mix with the cement + LS mixes are analyzed. Compared with the control mortar mixture, the inclusion of LS reduced compressive strength at all ages (2–180 days), and the reductions increased with increasing LS content.

Chen et al. (2012) investigated the effect of adding finely ground LS filler (LF) as a cement-paste replacement (not cement replacement) in high-flowability concrete (slumps ranging from 205 mm to 275 mm) while keeping the w/c ratio constant. The study used OPC and explored a wide range of mixture designs that had w/c ratios between 0.35 and 0.60, adjusted in small increments. The LS filler was incorporated at varying volumes depending on the w/c ratio. For mixes with lower w/c ratios, the LF volume was limited to avoid overly dry mixes, while at higher water-to-cement ratios, additional LF levels were evaluated. Across all mixes, the total paste volume (cement paste + LF) was held constant at 34%, meaning that adding LF directly reduced the cement paste volume but did not change the w/c ratio. The aggregate contents were also kept constant, and a superplasticizer was added incrementally to ensure high flowability in every mix. Compressive strengths were assessed at early and later ages. The results consistently showed that adding LS filler increased both 7-day and 28-day strengths, despite the constant w/c ratio. Higher LF volumes generally produced higher strengths under the same w/c condition. In some cases, adding a modest LF volume resulted in strength gains of around 7–11%, whereas increasing LF further yielded gains

ranging from approximately 14% to over 22%, depending on the cement and water proportions. Neithalath and Cam (2012) examined how replacing a portion of cement with coarse LS powder affects mechanical properties while keeping the overall water-to-powder ratio constant. Their mixtures used a baseline cement content of about 400 kg/m<sup>3</sup>, and cement was replaced by mass with LS powder at levels of 10% and 20%, alongside a reference mixture with no replacement. All mixtures were proportioned with a fixed water-to-powder ratio of 0.40, ensuring that changes in performance could be attributed only to the LS substitutions rather than water availability. To evaluate mechanical behavior, concretes were cast in cylindrical specimens of 100-mm diameter and 200-mm height. The study monitored compressive strength development over a 90-day period for both the reference concrete and the LS-modified concretes. The results showed a clear trend: because coarse LS powder acts primarily as an inert filler, its incorporation reduced compressive strength relative to the plain concrete at the same water-to-powder ratio. This reduction was observed consistently at all ages tested. The reference mix exhibited the highest strength at each hydration age, while the mixtures with 10% and 20% LS replacement showed progressively lower strengths, with the larger replacement producing the most significant reduction.

Githachuri and Alexander (2013) investigated how a commercially produced Portland–LS cement (PLC) performs in concrete compared with a conventional Portland cement (OPC), focusing on compressive strength at two binder contents and two water-to-binder ratios (w/b). Ten concrete mixtures were prepared at w/b = 0.40 and w/b = 0.55. The reference binder was a CEM I 42.5N OPC sourced from the Cape Town region. The PLC binder was a CEM II A-L 32.5R containing 20% LS filler interground with clinker. All mixtures used the same aggregates: 19 mm tillite as coarse aggregate, and a fine aggregate blend of river sand with 10% pit sand. To control workability and isolate the effect of the cement type, the mixtures were proportioned with a fixed water content of 165 L/m<sup>3</sup>, resulting in total binder contents of 412 kg/m<sup>3</sup> at a 0.40 w/b ratio and 300 kg/m<sup>3</sup> at a 0.55 w/b ratio. The coarse aggregate content was also kept constant at 1,070 kg/m<sup>3</sup>, and a polycarboxylate superplasticizer was used to maintain slump in the 65–95 mm range. Across both water-to-binder ratios, the reference OPC mixtures exhibited higher compressive strength at 28 and 91 days compared with their PLC counterparts.

Diab et al. (2016) evaluated the mechanical performance of concrete mixtures incorporating LS powder as a partial cement replacement. Twenty-one mixtures were produced using Type I Portland cement and LS powder, with replacement levels ranging from 0% to 25%. All mixtures were proportioned with a cement content of 400 kg/m<sup>3</sup> and a fixed water-to-binder ratio of 0.45. Compressive strength was measured at 3, 7, 28, and 365 days using 150-mm concrete cubes. The results showed that increasing the LS powder content generally led to a decrease in compressive strength. However, this reduction was minimal up to approximately 10% replacement, indicating that low levels of LS addition had only a minor effect on mechanical performance. Beyond 10%, the strength loss became more evident, and mixtures with higher LS contents exhibited more pronounced reductions in compressive strength at all ages.

Ahmad et al. (2022) investigated the effect of LS powder fineness and replacement level on concrete compressive strength. The study used Type I Portland cement with a Blaine fineness of 3,523 cm<sup>2</sup>/g, together with LS powders incorporated at 10%, 15%, and 20% by mass of cement. Three LS fineness levels relative to the cement were evaluated: fine (3,845 cm<sup>2</sup>/g), intermediate (3,215 cm<sup>2</sup>/g), and coarse (2,648 cm<sup>2</sup>/g). All mixtures maintained consistent proportions, including a binder content of approximately 490 kg/m<sup>3</sup>, 645 kg/m<sup>3</sup> of fine aggregate, 1,151 kg/m<sup>3</sup> of coarse aggregate, 164 kg/m<sup>3</sup> of water, and 0.6% polycarboxylate superplasticizer, with a fixed w/c ratio of 0.33. Compressive strength was measured at 1, 3, 7, and 28 days. Results showed that compressive strength decreased with increasing LS replacement level across all ages within each fineness batch.

Méndez-Páramo (2024) evaluated the effect of limestone filler (LS) content on the compressive strength of mortar mixtures produced with PLC containing between 3% and 35% LS. Mortar cubes

measuring  $5 \times 5 \times 5$  cm were fabricated using commercially available cements in Mexico. The LS content was determined indirectly from the standardized loss-on-ignition (LOI%) procedure (ONNCCE, 2017), using a previously established relationship between LS content and LOI%: LS% = 2 · LOI% (Méndez-Páramo, 2024). The cubes were cast and cured in lime-saturated water for either 28 or 90 days. Two sets of cubes cured for 28 days were then exposed to unsheltered, natural urban conditions in Querétaro City, Mexico, for up to one year. The results showed that the compressive strength of the mortars at 28 and 90 days was inversely proportional to the LS content in the PLC. The cubes exposed to the unsheltered urban environment developed surface fissures and exhibited even lower compressive strengths—reduced by more than 25% compared with their 28- and 90-day values. This behavior was unexpected and supports the hypothesis that chemical reactions involving CO<sub>2</sub> or SO<sub>3</sub> may have produced surface microcracking, thereby affecting the long-term mechanical performance of the material.

## 5. EFFECT ON POROSITY

Porosity is a critical parameter in assessing the microstructural development and long-term durability of PLC systems. However, its evaluation across studies (Courard et al., 2005; Matschei et al., 2007; Palm et al., 2016; Bharadwaj et al., 2022; Steiner et al., 2022) has been carried out using a wide range of experimental techniques, sample types, and analytical approaches, resulting in heterogeneity in data interpretation. This section synthesizes historical data from selected sources and categorizes the investigations by porosity type, test method, and the influence of compositional parameters, including LS replacement, alumina content, and w/c ratio.

Courard et al. (2005) analyzed mortar specimens with varying LS replacement levels (0–27%) using water absorption tests, which indirectly reflect total porosity but are sensitive to pore connectivity and capillary refinement. Two series of samples, labelled P and L, were prepared with a constant water-to-binder ratio of 0.45. While P-type mortars contained only LS replacements, L-type mortars also included slag as an SCM. Although the curing duration was not reported, results are assumed to correspond to 28 days. The results indicate that porosity increases with increasing levels of LS replacement, though the rate of increase stabilizes above 15%.

Matschei et al. (2007) presented porosity trends in cement pastes as part of a modelling framework, focusing on the thermodynamic evolution of hydrated phases in systems with LS. The calculated total porosity decreased as the LS content increased to about 2% by weight, but beyond that point it rose linearly with further LS addition. This trend does not specify the clinker phases' content, which may affect the evolution of hydration products, the gypsum content, and the w/c ratio.

In their 2016 study, Palm et al. evaluated porosity in mortar derived from concrete mixes after removing coarse particles larger than 2 mm—about 51 vol.% of the material. The total porosity was estimated from the evaporable water content using a relationship extrapolated from the Powers and Brownyard model (Crundwell et al., 2019). Excess water—defined as 0.27 times the clinker content—was plotted against measured total porosity, yielding a linear relationship. In general, mortars with higher LS content displayed a higher porosity. Although the study focused only on mortars, it links porosity development to effective water content and binder hydration progress, offering an indirect, model-based assessment influenced by assumed hydration kinetics and porosity evolution over time.

Bharadwaj et al. (2022) conducted a parametric analysis of cement pastes with varying levels of LS and alumina. Porosity was estimated using thermodynamic modelling and inferred from the volume of hydration products. The reported relationship between porosity and LS content was similar to that reported by Matschei et al. (2007). The authors plotted porosity for systems containing 0, 2.5, 5, and 7.5% alumina as a function of varying LS content. Results showed that the point of minimum porosity shifts to higher LS content as alumina increases, and the minimum porosity itself decreases. The lowest predicted porosity for pastes was 28%, occurring at 4% LS

and 7.5% alumina, corresponding to an optimal balance that favored the formation of space-filling carboaluminate and ettringite phases (with ~28% of the total volume occupied by hemi-/mono-carbonate phases and 8.5% by ettringite). When alumina exceeds 7.5%, porosity increases due to sulfate depletion, which limits ettringite formation and leads instead to the formation of monosulfate. In the range of  $7.5\% < \text{Al}_2\text{O}_3 < 9\%$  and  $4\% < \text{LS} < 10\%$ , this shift results in reduced space-filling phases. At alumina levels  $> 9\%$ , calcium hydroxide becomes depleted, and strätlingite forms in place of monocarbonates. Under these conditions, the predicted minimum porosity decreases further to 26%, occurring at 10% LS and 30% alumina. Beyond the critical LS content, porosity increases at all alumina levels due to dilution effects. Thus, porosity increased when sulphate became limiting or when dilution effects dominated. The study presents a model-predicted porosity map across a multidimensional compositional space, offering insight into the effects of phase equilibrium on microstructure. However, the absence of direct experimental validation limits its applicability for benchmarking against physical durability indicators.

Steiner et al. (2022) conducted a multi-technique porosity assessment of concrete specimens using gas pycnometry (He-displacement) and mercury intrusion porosimetry (MIP). The gas pycnometer enabled the determination of true volume and true density ( $\rho_T$ ), representing the mass-to-volume ratio of the solid phase, excluding intragranular voids. Bulk volume and bulk density ( $\rho_B$ ) were measured using a GeoPyc 1360 device. Effective porosity was then calculated as a function of  $\rho_T$  and  $\rho_B$  and is considered comparable to water-accessible porosity. Pore size distribution was derived using the Washburn equation (Crundwell et al., 2019) under a cylindrical pore model. This approach captured both total and effective porosity with high resolution across a range of pore diameters. The findings indicate that, at a given water-to-binder ratio, higher LS contents lead to increased concrete porosity, as observed by previous authors. However, the use of concrete specimens (3-cm diagonal cuboids) introduces spatial variability due to coarse aggregates, potentially obscuring binder-matrix-specific trends unless aggregate-free subsamples are analyzed. Across the literature, discrepancies arise from the type of porosity evaluated—total, effective, or capillary—and from methodological approaches used, which range from water absorption, gas pycnometry, and mercury intrusion porosimetry to thermodynamic modelling. Comparative analysis is further complicated by variations in sample types (cement paste, mortar, concrete), w/c ratios, LS replacement levels, and the lack of standardized curing durations. Additionally, few studies track porosity evolution over time, limiting insight into the early-age versus long-term dynamics of porosity. Given this variability, direct comparison between datasets is not viable without normalization procedures.

Méndez-Paramo (2024) presents experimental results from Portland cement mortars in which inert limestone fillers were used to replace 20% to 40% of the clinker. The results include the mechanical (compressive strength), physical (electrical resistivity, total void content, capillary porosity), and chemical (carbonation under natural exposure) performance of these mortars at different ages up to one year. The experimental findings show that adding between 15% and 35% inert limestone filler increased saturated electrical resistivity and decreased ultrasonic pulse velocity. These techniques indirectly determine the porosity of cement-based materials, and both corroborate that increasing the limestone filler content in cement increases the porosity of the mortar mixtures tested.

## 6. EFFECT ON CHLORIDE INGRESS

Chloride ingress is a key durability indicator in reinforced concrete systems, as it directly correlates with the risk of corrosion initiation in embedded steel. The quantification of chloride transport through PLC concrete has been approached in the literature using both natural diffusion and accelerated migration methods. The diversity in experimental conditions, testing protocols, curing procedures, and compositional variables (e.g., LS replacement and w/c ratio) contributes to the

heterogeneity in reported diffusion coefficients. This section compiles the most relevant data from the literature, highlighting the test methodologies, sample configurations, and variables affecting chloride ingress in PLC systems. The reviewed studies reveal broad variation in experimental frameworks, including:

- Test types: apparent diffusion (ASTM C1556), accelerated migration (NT BUILD 492, potential difference methods), and modified electrochemical setups.
- Sample geometry: full-size prisms, cores, or discs ranging from 25 mm to 100 mm in thickness.
- Curing methods: wet, air-dry, and combined regimes with durations rarely consistent across studies.
- Age at testing: from early ages (35 days) to long-term exposure (1 year), introducing variability in binder maturity and pore structure development.
- LS replacement: often variable, limiting direct correlations to chloride transport behavior.

The lack of harmonization in experimental procedures, reporting of compositional details, and time-dependent data makes it challenging to establish generalized trends regarding PLC performance. Despite the variety of experimental approaches, consistent trends indicate that increasing LS content in cement generally leads to higher apparent chloride diffusion coefficients, particularly at elevated w/c ratios (Irassar et al., 2001). However, the interpretation of binder effects is often confounded by variations in testing methodology, exposure duration, preconditioning procedures, and internal moisture profiles. Differences between steady-state and non-steady-state diffusion methods, between accelerated and natural exposure regimes, and in sample geometry further hinder direct comparisons across studies. To advance understanding of chloride transport in PLC systems, future research should emphasize standardizing test protocols and including long-term exposure data that better reflect in-service conditions.

Irassar et al. (2001) investigated chloride ingress in concrete with various LS replacement levels (0%, 10%, and 20%) and three w/c (0.4, 0.5, and 0.6). Concrete prisms measuring 100×150×530 mm were cast and cured in their molds for 24 hours. After demolding, the specimens underwent either wet curing (6 days in lime-saturated water followed by 21 days in laboratory air) or air curing (27 days in laboratory air). At 28 days, all surfaces were sealed with epoxy, except for one face perpendicular to the casting direction, and the prisms were saturated in water for 24 hours before immersion in a 3% NaCl solution. After 180 and 360 days of exposure, a 70-mm slice was cut from the top of each prism; the remaining specimens were resealed with epoxy, and immersion continued for later measurements. Powder samples were then collected at 5 mm intervals using a drilling device to determine the total chloride content. The authors determined surface chloride and apparent diffusion coefficients by fitting the experimental data to a nonlinear regression model using the error function. The results indicated that increasing LS replacement leads to higher apparent chloride diffusion coefficients. Thomas and Hooton (2010) conducted a similar study with less conclusive results; however, the collected data were obtained after only 35 days of immersion. Dhir et al. (2007) investigated chloride diffusion in concrete slices (w/c ranging from 0.45 to 0.65) using an accelerated technique within a two-compartment cell, applying a chloride concentration gradient and a 7.5 V electrical potential across the assembly. The LS replacement levels investigated were 0%, 15%, 25%, 35%, and 45%. The results showed that chloride diffusion coefficients increased with higher LS replacement, particularly at levels above 25%. This effect was further amplified in concrete with higher w/c ratios.

Cam and Neithalath (2010) conducted a non-steady-state migration test in accordance with NT BUILD 492 (Nordtest, 1999) for concrete mixtures with varying w/c (0.34, 0.37, and 0.4) and LS replacement levels (10% and 15%). Concrete specimens, 50 mm thick, were first saturated with a 2% calcium hydroxide solution and then placed in a test cell containing 2 N NaCl in the catholyte chamber and 0.3 N NaOH in the anolyte chamber. The applied voltage was determined from the initial current response when a 30 V potential was imposed across the electrodes. This electrical

potential drove chloride ions from the cell into the specimen. After the 24-hour test, each specimen was split axially, and the exposed surfaces were sprayed with a 0.1 N silver nitrate solution to reveal the depth of chloride penetration. The chloride diffusion coefficient increased when incorporating LS powder into the concrete mixture, although the increase was slightly lower for longer curing times (56 days vs. 28 days). Replacing clinker with 15% LS and 5% silica fume at a low w/c of 0.34 resulted in a similar chloride diffusion coefficient to an OPC mixture with the same w/c. This suggests that optimizing the w/c ratio and incorporating SCMs to PLC can mitigate adverse effects associated with using LS alone.

Meddah et al. (2014) measured chloride-ion ingress through concrete slices (100 mm in diameter, 25 mm thick) extracted from 150×300 mm cylinders. The variables tested were the same as those of Dhir et al. (2007). The specimens underwent two curing regimes—wet curing and dry air curing—for the first 28 days, and results from both conditions were reported. Each slice was placed in a standard two-compartment diffusion cell, with the inner chamber filled with calcium hydroxide solution and the outer face exposed to a chloride solution by partial immersion in a chloride tank. The electrochemical test employed a 7.5 V potential difference across the specimen to drive chloride migration. Chloride transport was monitored through periodic analysis of the liquids in both chambers, and the diffusion coefficient was calculated using Fick's first law. At any given w/c ratio, concretes with higher LS powder replacement show noticeably higher diffusion coefficients than OPC mixtures, demonstrating that increasing LS content leads to reduced resistance to chloride diffusion.

Celik et al. (2015) evaluated the chloride penetration resistance of one-year-old concrete samples using the non-steady-state migration method in accordance with the NT BUILD 492 (Nordtest, 1999). Cylindrical specimens (100×200 mm) were cut into sections 50±2 mm thick and stored in a fog room until testing. In addition to OPC mixtures with 0%, 15%, and 25% LS content, binary and ternary mixtures also containing SCMs were tested. Prior to testing, the samples were vacuumed in a saturated calcium hydroxide solution and placed between a catholyte (10% NaCl) and an anolyte (0.3 N NaOH). A 30-V electrical potential was applied across the specimens for 24 hours. After testing, each sample was split axially, and a 0.1 M silver nitrate ( $\text{AgNO}_3$ ) solution was sprayed onto the freshly exposed surfaces. Chloride-contaminated areas were identified by the formation of white precipitate. The chloride migration coefficient in mixtures with 25% LS replacement was approximately twice that measured for the OPC system. This effect was substantially mitigated in the binary and ternary mixtures.

Palm et al. (2016) conducted accelerated chloride migration tests in accordance with the BAW-Merkblatt Chlorideindringwiderstand von Beton guidelines (Bundesanstalt für Wasserbau, 2012) on concrete mixtures with w/c ratios of 0.35, 0.45, and 0.50 at 35 and 98 days. Disc-shaped specimens measuring 100×50 mm were used, with 0.2 N KOH as the anolyte and 0.2 N KOH containing 10% NaCl as the catholyte. It was observed that mixes with LS fillers (two types) exhibit a wide range of results. Some combinations perform significantly worse, especially at 35 days, while others approach the performance of the control mixture, showing that the type and proportion of LS addition have a notable impact on early-age chloride resistance. The highest coefficients occur in mixtures with high LS content and a w/c of 0.5, whereas the lowest are observed in concrete mixtures with a w/c of 0.35 after 98 days of test, underscoring that both mixture design and age are key factors in limiting chloride ingress.

Da Silva and De Brito (2016) determined the chloride diffusion coefficient following the procedure outlined in NT BUILD 492 (Nordtest, 1999). For each mix (LS contents of 0%, 30%, 60%, and 70%) and age studied (28, 91, and 182 days), three cylindrical specimens (100 mm in diameter and 50 mm thick) were prepared. After undergoing wet curing at 20±2 °C and relative humidity above 95%, the specimens were preconditioned under vacuum and immersed in a saturated calcium hydroxide ( $\text{Ca(OH)}_2$ ) solution made with distilled water. The chloride migration coefficient increases significantly with increasing LS content, although this effect is attenuated at 182 days.

The LS-fly ash mixtures with combined replacement levels of 30% and 60% show migration coefficients comparable to those of the OPC mixture, particularly at 91 and 182 days.

Elgalhud et al. (2022) analyzed chloride ingress in PLC-concrete by drawing on a large dataset extracted from international studies. The authors have reported that chloride penetration increases as LS content rises, though the effect is smaller when PLC mixes are proportioned to match the strength of equivalent OPC concretes rather than simply using the same w/c ratio. The findings suggest that European durability requirements for chloride exposure may need to be reconsidered when LS is incorporated into Portland cement.

## 7. EFFECT ON CARBONATION

Carbonation is a critical deterioration mechanism in reinforced concrete, particularly in PLC systems, where increased LS content can modify the microstructure, reduce pH buffering capacity, and influence carbonation depth. The extent of carbonation depends on several factors, including cement type, curing conditions, w/c, porosity, and environmental exposure (CO<sub>2</sub> concentration, relative humidity, and temperature). This section reviews key carbonation studies on PLC concretes and mortars, identifying differences in test conditions and experimental configurations that influence comparability and interpretation. The reviewed studies exhibit considerable heterogeneity in experimental setups, including:

- CO<sub>2</sub> concentration: ranging from natural (0.035%) to highly accelerated (up to 20%), affecting carbonation front advancement and comparability.
- Relative humidity: generally maintained between 55–65%, but some variations exist that impact the diffusion–reaction balance.
- Sample types and sizes: from small mortar specimens to large prisms and diffusion cells, influencing moisture gradients and carbonation kinetics.
- Preconditioning protocols: drying periods and sealing strategies vary widely, influencing surface microstructure and test realism.
- Exposure duration: from 28 days to multi-year natural exposure, reflecting both short-term kinetics and long-term durability behavior.

Despite the diversity of approaches, consistent findings suggest that the carbonation depth generally increases with higher LS content (Courard et al., 2005; Dhir et al., 2007; Meddah et al., 2014; Bucher et al., 2017; Palm et al., 2016; Steiner et al., 2022), especially under insufficient curing or elevated porosity. However, the impact of w/c ratio, curing quality, and testing configuration often outweighs binder composition in determining carbonation resistance. Future research should aim to harmonize testing protocols and extend exposure durations to better simulate field conditions and enable reliable comparisons across binder types.

Dhir et al. (2007) conducted two series of carbonation tests: (i) concrete samples exposed to a natural carbonation environment using the EN 12390-10:2018 method (storage in a 0.035% CO<sub>2</sub> environment for 365 days at 20°C, 65% RH) and (ii) 100-mm cubes exposed to an accelerated regime (storage in an enriched 4% CO<sub>2</sub> environment for 20 weeks at 20°C, 65% RH). The LS replacement level ranged from 15% to 45%. Independent of w/c ratio, specimens exposed to the natural carbonation environment showed increased carbonation depth with increasing LS content, and this difference was amplified at elevated w/c ratios. Similar observations were made for those concrete samples exposed to an accelerated environment.

Meddah et al. (2014) conducted accelerated carbonation testing on 100-mm cubes that were wet cured for the first 28 days. The specimens were then pre-conditioned by drying in the laboratory for 14 days. Before curing in the carbonation chamber, all sides of the concrete specimens were sealed with bituminous coating paint, except the top side, which was exposed to a CO<sub>2</sub>-enriched atmosphere containing 4% CO<sub>2</sub> at 20°C and 55% RH. After 20 weeks of exposure, carbonation

depths were greater in specimens with higher LS replacement levels, which ranged from 10% to 45%. The effect was more pronounced in the samples with a water-to-binder ratio of 0.65 than in those with a ratio of 0.5.

Bucher et al. (2016) conducted accelerated carbonation tests on prismatic concrete samples with different LS content (0, 15%, 20%, and 25%) after curing times of 28 and 365 days. Between the curing time and the beginning of the carbonation test, the samples were stored in an atmosphere with  $50 \pm 5\%$  RH at  $20^\circ\text{C}$  for 14 days. Accelerated carbonation was conducted in a chamber at  $20^\circ\text{C}$ , 55% RH with a  $4 \pm 0.5\%$  CO<sub>2</sub> content. Carbonation depth measurements were taken at 56, 63, and 70 days by spraying a phenolphthalein solution onto the concrete. A replicate set of samples was also exposed to natural carbonation after 28 days of curing. In this case, the specimens were stored in situ outside the laboratory, with a mean temperature in Toulouse from 8 to  $18^\circ\text{C}$  between winter and summer and an average RH of 75%. The carbonation depth was measured after 1 and 2 years of natural exposure.

The accelerated carbonation tests reveal that LS addition consistently increases carbonation depth, with the magnitude of the effect strongly dependent on the replacement level and binder composition (CEM I and blended CEM II). For CEM I systems, carbonation resistance declines steadily as LS content rises, with a pronounced loss of performance at higher dosages ( $> 20\%$ ), where carbonation depths more than double relative to the clinker-rich control sample—even after 365 days of curing. Extended curing substantially reduces carbonation across all mixtures, yet it does not eliminate the performance gap introduced by LS. In blended CEM II binders, the influence of LS is more nuanced: systems containing slag exhibit only minor changes in carbonation behavior, whereas fly ash-based formulations remain highly susceptible, and the addition of LS further aggravates their already elevated carbonation depths. Overall, the results demonstrate that LS incorporation reduces carbonation resistance in most cases, especially in pure Portland cement and fly ash-containing systems, while slag-bearing blends show a more moderate response. Similar results were observed for the samples exposed to natural carbonation; increasing LS content in CEM I mixtures leads to noticeably greater carbonation depths after 1 and 2 years of exposure, with the rise becoming more pronounced as the replacement level increases. In contrast, the CEM II blends exhibit only modest increases, indicating a less sensitive response to LS additions.

Palm et al. (2016) exposed  $100 \times 100 \times 500$  mm<sup>3</sup> concrete prisms to an accelerated carbonation environment containing 2% CO<sub>2</sub> by volume at  $20 \pm 2^\circ\text{C}$  and  $65 \pm 5\%$  R.H. for 28 days. The authors also reported deeper carbonation fronts in concretes with higher LS replacement levels.

Steiner et al. (2022) employed a CO<sub>2</sub>-diffusion setup comprising a cylindrical measurement cell sealed with a disc-shaped specimen, placed in a carbonation chamber. The diffusion cell is an acrylic cylinder ( $h = 210$  mm,  $\varnothing = 100/94$  mm, wall thickness = 6 mm) composed of three sections. Its lower section, 50 mm in height, holds a saturated NH<sub>4</sub>NO<sub>3</sub> solution to maintain an internal relative humidity of 65%. After sealing the specimen to the cell with wax, the assembly was conditioned at  $20^\circ\text{C}$  and 65% RH until the wax hardened. Before testing, the cell was purged with pure nitrogen for approximately 10 seconds, reducing the initial CO<sub>2</sub> concentration to approximately 0.08%. The sealed cell was then placed in a carbonation chamber maintained at 20% CO<sub>2</sub>. CO<sub>2</sub> subsequently accumulated inside the cell solely through diffusion across the specimen. Concrete samples had LS replacement levels of 20%, 40%, 50%, and 60% by weight. The results revealed that increasing LS replacement significantly increases CO<sub>2</sub> diffusivity and carbonation depth, indicating reduced carbonation resistance as LS content increases.

Méndez-Páramo (2024) included in his experimental program the carbonation resistance of mortar mixtures fabricated with PLC containing limestone filler contents between 4% and 35%, exposed for one year to outdoor conditions in Querétaro, Mexico. The results showed that a limestone filler content of 15% produced approximately 50% more carbonation than an OPC mortar, while contents of 20% and 35% doubled and quadrupled the carbonation depth of the OPC mortar, respectively.

Beyond the pH-reduction effects discussed above, the incorporation of LS filler in Portland-cement-based materials induces additional carbonation-related mechanisms that extend well beyond alkalinity loss (Torres-Acosta, 2024a, 2024b). In the early stages of exposure, CO<sub>2</sub> reacts preferentially with portlandite (CH), forming calcium carbonate and reducing the pore solution pH; however, once CH is depleted—an effect that occurs earlier in systems with high LS contents due to reduced clinker fraction and higher capillary porosity—the carbonation front reaches the calcium silicate hydrate (CSH) gel. The carbonation of CSH causes progressive decalcification, lowering its Ca/Si ratio and generating silica-rich, poorly binding gels (Thiery et al., 2007; Chen et al., 2021). This transformation is associated with structural densification of the newly formed carbonates and a collapse of the CSH nanostructure, producing measurable carbonation-induced shrinkage (Hoult & Wittmann, 2002; Saetta et al., 1995). Unlike the volumetric stability observed when only CH is carbonated, the carbonation of CSH generates internal tensile stresses sufficient to initiate microcracking in the cement matrix even without external loading. These microcracks, in turn, accelerate the ingress of CO<sub>2</sub> and water vapor, creating a feedback loop that accelerates both chemical alteration and mechanical deterioration (Drouet et al., 2019; Suda et al., 2021).

In LS-modified systems, this phenomenon may become more pronounced due to a combination of faster CH consumption, enhanced nucleation of calcium carbonate on calcite surfaces, and a modified pore structure that facilitates CO<sub>2</sub> transport. At high LS contents, the earlier onset of CSH carbonation means that cracking can occur sooner and at shallower carbonation depths compared to ordinary Portland cement (OPC) matrices (De Weerdt et al., 2011). Consequently, while reduced pH and steel depassivation are traditionally highlighted as the primary risks associated with carbonation, the mechanical consequences linked to carbonation shrinkage and CSH decalcification represent a more severe deterioration pathway. This dual impact, chemical (loss of alkalinity) and mechanical (shrinkage-induced cracking), is vital for evaluating the long-term durability of cementitious systems with LS filler, especially under high CO<sub>2</sub> exposure or in structures with limited moisture buffering capacity (Torres-Acosta, 2024a, 2024b).

## 8. RESEARCH GAPS

PLC has been extensively studied for almost 50 years, with its potential to reduce CO<sub>2</sub> emissions while maintaining mechanical and durability properties comparable to those of traditional Portland cement. However, existing research presents several key limitations:

- **Limited direct correlation between porosity and performance indicators** – While numerous studies have assessed mechanical properties (e.g., compressive, flexural, and tensile strength) and transport-related properties (e.g., resistivity, ultrasonic pulse velocity, rapid chloride permeability, diffusion, and carbonation), the relationship between porosity and these performance metrics remains insufficiently explored. Understanding this link is crucial for evaluating long-term durability and optimizing mix designs.
- **Inconsistent research timelines and experimental frameworks** – The evolution of PLC's mechanical and transport properties is often evaluated over varying timeframes, influenced by differences in test standards and experimental methodologies. This lack of consistency hinders the ability to draw reliable, long-term conclusions and to reach consensus about PLC's performance under different conditions.
- **Long-term performance of PLC with different LS replacement levels** – Many studies focus on early-age strength development, but fewer systematically examine the long-term effects (beyond 28 days) of varying LS contents in PLC. There is a need for a structured, year-long assessment that quantifies the impact of different replacement levels on compressive strength, transport properties, and porosity over time.

- **Sustainability validation of PLC as an eco-friendly alternative** – Although PLC is widely promoted as a sustainable cement alternative due to its lower clinker content and reduced CO<sub>2</sub> footprint, its actual long-term performance relative to its environmental impact remains an open question. A more comprehensive, integrated evaluation is needed to assess whether PLC truly delivers an optimal balance among sustainability, strength, and durability.
- **Transition from prescriptive to performance-based standards** – Current cement and concrete standards are primarily prescriptive, specifying material compositions rather than performance-based criteria. However, there is a gradual shift in the industry toward performance-based standards, which require validated data to support their implementation. The challenge lies in how PLC can meet these performance standards when fundamental prescriptive data on its behavior is still incomplete. Without a robust understanding of its mechanical, durability, and transport properties, it is unclear how performance-based criteria can be effectively calibrated for PLC.
- **Lack of systematic calibration of PLC in conventional testing methods** – The calibration of PLC within conventional test methods remains largely empirical, relying on batch or trial-based adjustments rather than a structured, data-driven approach. This trial-and-error methodology introduces inconsistencies in assessing PLC's performance relative to traditional Portland cement. A systematic evaluation of PLC's behavior is necessary to develop more reliable predictive models and calibration frameworks for standard testing methods.

## 9. CONCLUSIONS

### Compressive Strength

Research shows that when LS is used as a cementitious replacement of clinker or Portland cement, LS-Portland cement can achieve strength levels similar to OPC while providing sustainability benefits. PLC generally maintains compressive strength comparable to OPC at early ages (28 days), particularly for moderate LS replacements (<15%). However, strength gain or comparison may diminish at longer ages if mix designs are not calibrated to maintain performance, especially in systems with higher LS content, high w/c ratios, or without supplementary cementitious materials. When LS is not used as a clinker replacement—acting instead as an ultra-fine aggregate—no sustainability benefit is achieved, though compressive strength may remain comparable or even slightly higher.

### Porosity

Total and effective porosity of PLC increases with higher LS replacement. Both thermodynamic models and experimental measurements indicate that this trend is influenced by cement composition (alumina, gypsum) and w/c ratio. Increased porosity may affect long-term durability, highlighting the importance of microstructure control through proper curing and SCM incorporation and LS replacement limits.

### Chloride Ingress

Chloride diffusion increases with LS content, particularly in concrete with higher w/c ratios. Experimental studies using accelerated and natural methods show that incorporating SCMs and optimizing mix design can mitigate this effect, allowing PLC to meet durability criteria against steel corrosion in chloride-exposed environments.

## Carbonation

PLC exhibits greater carbonation depths compared to OPC, especially in mixtures with high LS content and insufficient curing. Carbonation resistance strongly depends on curing time, w/c ratio, and SCM presence, with slag-based systems showing greater resistance and fly-ash-based systems being more susceptible. Unlike the relatively stable volumetric response observed when only CH carbonates, the carbonation of CSH induces internal tensile stresses that can initiate microcracking within the cement matrix even in the absence of external loading. The formation of these microcracks subsequently enhances the ingress of CO<sub>2</sub> and water vapor, establishing a self-reinforcing feedback mechanism that accelerates both the chemical transformation of the binder and the progressive mechanical deterioration of the material.

Overall, PLC represents a sustainable alternative to OPC, reducing CO<sub>2</sub> emissions while providing acceptable structural performance. However, durability-related properties—porosity, chloride transport, and carbonation—are influenced by composition and curing conditions. These findings highlight the need for a continuous, integrated approach linking microstructure, ion transport, and mechanical behavior to the design of reliable and durable PLC mixes, supporting the transition toward performance-based cement standards.

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