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Durability and Long-term Performance Assessment in Eco-efficient Concrete Mixtures Towards a More Sustainable Cement Industry - A Critical Review

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Abstract. *In the current need to fulfill a Net-Zero goal to offset greenhouse gas emissions by 2050, the cement industry deeply needs to renovate its process and products. The amount of concrete, and consequently ordinary Portland cement (OPC) used worldwide, far exceeds other industrial materials, contributing to around 8% of global emissions. A recognized solution would be to partially replace clinker with limestone, an available material in many countries with the potential to improve the particle packing of a concrete mixture. Even though several studies have been carried out on the application of Portland limestone cement (PLC) concrete, there is a need for further investigation regarding durability performance. Problems include alkali-silica reaction (ASR), delayed ettringite formation (DEF), sulphate attack, steel corrosion, and freeze/thawing, whose deteriorating effects tend to be maximized in harsh climate countries such as Canada and should be evaluated when introducing distinct materials into concrete. Therefore, this paper aims to review previous assessment of the durability of PLC concrete mixtures. The use of an advanced mix-design method (particle packing model and mobility parameters - PPM+MPs), will also be discussed on its ability to enhance the durability performance of the mixtures. It is expected that novel guidelines to design more eco-efficient concrete mixtures using PLC can be developed to reduce concrete's cost and carbon footprint, achieving a more sustainable industry.*

Keywords: *Portland-limestone concrete; Clinker substitution; Concrete durability; Particle packing models (PPMs); Mobility parameters (MPs)*

1 Introduction

The cement industry's environmental impact is mainly due to the production of clinker, which accounts for 8% of global greenhouse gas emissions (Petroche & Ramirez, 2022). The clinker comprises over 90% of Portland cement, and its manufacturing process releases a significant amount of CO_2 from fossil fuel combustion and decarbonization of limestone. Reducing the OPC content by incorporating cement replacing materials and ensuring that the new cement maintains the same mechanical properties and durability as pure OPC concrete is challenging. To address this issue, the Cement Association of Canada (CAC) recently partnered with the Federal Government of Canada and launched the roadmap to achieve net-zero carbon concrete by 2050. Research has been conducted in Canada on exploring sustainable materials to replace clinker. Among these research, interground limestone powder, which made the so-called Portland Limestone Cement, appears promising and feasible due to its assessed sustainability (Panesar & Zhang, 2020), wide availability, volumetric stability within the cement paste, and its potential to improve the microstructure of the concrete mixture (Moon et al., 2017). In addition, the durability performance and sustainability of PLC concrete can further be optimized by implementing supplementary cementitious materials (SCMs) (Ramezanianpour & Hooton, 2014; Cost et al., 2013). Two common methods to produce PLC are (1) intergrinding cement clinker and limestone through the cement production process (interground PLC); (2) blending the cement and graded limestone powders through the concrete batching-mixing process (blended PLC). Although both methods of cement replacement seem similar, PLC of higher fineness can be achieved by over-grinding the clinker with the limestone (Kumar et al., 2013), resulting in a higher fineness. Moreover, LFs can be further classified into "replacement filler" and "performance filler", where the former type is of a similar particle size distribution (PSD) of OPC, and the other has a lower PSD value (T. de Grazia et al., 2019).

While some researchers have suggested that higher fineness of LFs can significantly improve the performance of PLC concrete by increasing packing density and improving pore structure (Ramezanianpour & Hooton, 2014; Elgalhud et al., 2016; Moon et al., 2017), it is generally accepted that PLC behaves similarly to OPC only up to 10% - 15% replacement by weight. Beyond this ratio, the performance of PLC concrete can deteriorate significantly compared to OPC (Tsivilis et al., 1999; Cost et al., 2013; Bentz et al., 2015; Garcia et al., 2022). Nevertheless, this conservative conclusion can result from non-standardized PLC characterization procedures and the direct replacement of OPC using a conventional mix-design method (e.g., ACI method). Therefore, a new mix-design guideline for eco-friendly concrete should be developed systematically from material characterization to mix proportioning to ensure comparable or superior engineering properties and durability performance, even at higher LFs replacement ratios (e.g., more than 15%). According to recent literature, the Particle Packing Models (PPMs) combined with the mobility parameters (MPs) method is proven to be an excellent approach to designing low-cement concrete (LCC) with blended LFs up to 50% replacement ratio (T. de Grazia et al., 2019). However, while LFs and OPC are produced, characterized and mix-proportioned individually as fillers, PLC is defined integrally as just one type of cement, which requires further research to modify the PPM-MPs method. Hence, further research is needed to alter the PPMs-MP approach to account for PLC. Additionally, the effect of SCMs and common durability issues in Canada, such as freezing and thawing, sulphate attack, and chloride-induced corrosion, have not been fully investigated. Thus, it is of significance to extend the PPM-MPs method to PLC concrete mix design, with the implementation of SCMs, and a comprehensive assessment of long-term durability behaviour through accelerated lab tests.

2 Particle Packing Models + Mobility Parameters

The concept of packing models stems from the observation that a system of monodispersed spherical particles can only occupy approximately 75% of the available space, regardless of the particle size. Therefore, a highly compacted mixture must consist of particles of various sizes, where the voids formed among coarser particles are filled by finer particles. Then, the smaller voids created by these finer particles are filled by even finer particles and so on. Particle packing models are introduced to achieve a highly compacted concrete mixture with significantly low porosity by quantitatively controlling the aggregate size and proportions. PPMs are generally classified into two categories: discrete PPMs and continuous PPMs. While both paths have been developed and upgraded over the years, some researchers (Yu et al., 2014) suggest

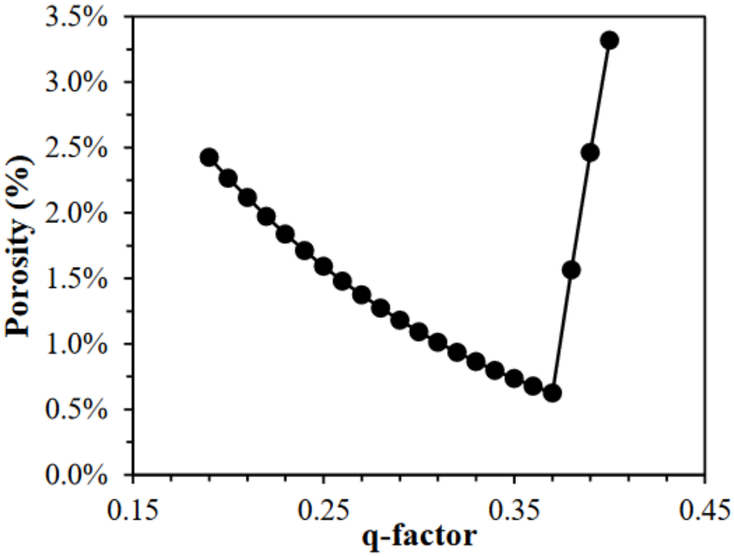

Figure 1. Comparison of porosity with different q-factors (T. de Grazia, 2022)

that real concrete mixture are better represented by continuous models as they consider that concrete contains particles of all sizes. Thus, the most current continuous PPM, also known as the Alfred model or modified Andreasen model (T. de Grazia et al., 2019), is applied in this research.

Through the Alfred (or modifed Andreasen) model, the theoretical porosity of the mixture can be predicted with a coefficient of distribution (q), as shown in Fig. 1. A minimum of approximately 0.5% porosity can be achieved when q factor equals 0.37. However, reducing porosity (increasing packing density) also increases particle friction, resulting in a more viscous mixture. Therefore, smaller q values (0.21 to 0.22) are usually used for applications which require high flowability like self-compacting concretes, while for vibrated and/or pumped concretes, the value is ranged from 0.26 to 0.28 to ensure the shear-thinning property. Equation (1) exhibits the Alfred model which can figure out the optimum particle size distribution for a given set of solid materials (cement,fine and coarse aggregates) to design concrete.

$$CPFT = 100 * \left(\frac{D_P^q - D_S^q}{D_L^q - D_S^q} \right) \tag{1}$$

where CPTF is the cumulative percent finer than D_P (in volume); q is the distribution factor; D_P is the particle diameter; D_L is the largest particle diameter in the system, and D_S is the smallest particle diameter in the system.

However, concrete mixtures designed through PPMs still face challenges in the fresh state due to their highly packed and low porosity system. Thus, two mobility parameters (interparticle spacing and maximum paste thickness) are introduced to supplement the mix-proportioning of the PLC mixtures, ensuring fresh-state behaviours. The interparticle spacing (IPS) measures the average distance between the finer particles, which can also be considered the amount of fluid/water separating the fine particles. Similarly, maximum paste thickness (MPT) measures the maximum thickness of the paste, which indicates the thickness of paste that separates coarser aggregates (T. de Grazia et al., 2019). Generally, the higher the mobility parameters, the higher the flowability of the concrete mixture. As can be seen from equations (2) and (3) (T. de Grazia et al., 2020), both parameters are dependent on the specific surface area (SSA).

$$IPS = \frac{2}{VSA}\left[\frac{1}{V_S} - \frac{1}{\left(1 - P_{of}\right)}\right] \tag{2}$$

where IPS is the interparticle spacing, VSA is the calculated volume surface area per cubic centimetre of powder, V_S is the volume fraction of fine solids (particles smaller than 125 μm), and P_{of} is the pore fraction assuming the densest packing of the fine particles.

$$MPT = \frac{2}{V_{SAC}}\left[\frac{1}{V_{SC}}\right]\frac{1}{\left(1 - P_{ofc}\right)} \tag{3}$$

where MPT is the distance between aggregates, VSA_C is the calculated volume surface area of aggregate (particles greater than 125 μm) fraction, V_{SC} is the volumetric aggregate solid fraction, and P_{ofc} is the porosity of aggregate fraction assuming the densest packing.

3 Long-term Durability Performance of PLC Concrete

3.1 Carbonation

Global warming can increase CO_2 concentration and carbonation penetration in concrete structures, harming the rebars by lowering the pH, and bringing a less alkaline environment within the concrete, thus initiating corrosion. Some research (Elgalhud et al., 2017) reported the implementation of LFs leads to increased carbonation depth. This deterioration was found in the accelerated lab tests and the in-site measurements of 1-5 years of concrete structures made with PLC. However, other studies claim no significant variance was found with a 12% (Thomas et al., 2013) and a 35% replacement ratio (Tsivilis et al., 2002). A proper explanation could be the higher packing density and lower porosity brought by adding LFs. Therefore, PPM-MPs may increase the PLC concrete's resistance to carbonation since the mixture will have a lower porosity.

3.2 Chloride Penetration

Reinforced concrete infrastructures like bridges are prone to be compromised by chloride ingress since the massive amount of de-icing salts is applied in the winter. The chloride ions in the seawater can also trigger chloride ingress. If chloride penetrates concrete, it can cause fast and severe corrosion of the reinforcement, which reduces the cross-section of the reinforcement and thus leads to the loss of its load-carrying capacity. It will also generate a secondary explosive product, causing cracking and spalling of the concrete cover. Chloride-induced corrosion of the reinforcement is one of the leading causes of concrete structural deterioration and, therefore, responsible for a large share of the cost of rehabilitating concrete structures (Loser et al., 2010). The thickness of the concrete cover over the reinforcement and the concrete's permeability is the dominant parameters that influence the resistance of chloride ingress. While a thicker concrete cover is not favoured from a sustainability and design perspective, improving the pore structure (e.g., less permeability) with PPM-MPs seems feasible. Although the previous study (Elgalhud et al., 2018, 2016) reported chloride ingress of concrete accelerates as the LFs content rises, the results are still based on concrete made through the conventional mix-design method, where the pore structure may not be improved through the correct approach to use LFs.

3.3 Alkali-silica Reaction

Alkali-silica reaction (ASR) is a chemical deterioration mechanism worldwide and is especially severe in countries with harsh climates like Canada. The internal reaction between the mineral phases of the aggregates and the alkali hydroxides (high pH) of the concrete pore solution will generate an expansive secondary gel, which causes distress and cracks in the concrete structure. Conversely to carbonation, the lower pH brought by the LFs can significantly mitigate this deleterious issue. The reduced permeability through the PPM-MPs method can further limit the available moisture, which is beneficial as well. Previous research verified the above discussion, where PLC concrete demonstrated less expansion compared to OPC concrete in the Concrete Prisms Test (Thomas et al., 2013).

3.4 Sulphate Attack

Sulphate attack is the chemical/physical attack of cementitious materials by internal sources and/or sulphate ions from solutions external to concrete. Chemically speaking, C-S-H will partially decalcify to provide Ca cations to form ettringite and gypsum with the presence of sufficient sulphate ions, which provides the strength of the concrete, causing expansion and softening. Physically speaking, the accumulation of crystallization of sulphate salts will exert internal stress and damage the concrete as well. This bidirectional attack makes the mechanism more complex. Although most literature does not reach an agreement on the influence of PLC in sulphate condition (Ramezanianpour & Hooton, 2013), there is research indicating the size of limestone fillers contributes significantly to the sulphate damage (Al Shamaa et al., 2016). While it is well established that internal sulphate attack is dominantly dependent on chemical composition of the concretes (e.g., the sulphate and aluminate contents), the external sulphate ingress can be mitigated through optimizing the transport mechanisms (e.g., capillary suction, diffusion, permeation, wick action) through the PPM-MPs method.

3.5 Freezing and Thawing Cycles

Freezing-thawing cycles (F&T) damage occurs when water is absorbed into the concrete, freezes, and expands. The volume of frozen water is 9% greater than liquid water, so when water freezes, pressure is exerted on the surrounding concrete, and distress and cracks will be generated. Although it is widely accepted that improving the pore structure through air-entrained admixture is a reliable solution to diminish the damage, no research has been conducted to investigate the influence of the PPM-MPs modified pore structure combined with the air-entrained admixture against the F&T cycles. Thus, the behaviours of concrete made of high LFs replacement PLC needs further investigation and confirmation.

4 Discussion and Challenges

Holistic life cycle assessment (Celik et al., 2015) has proved that the increasing of LFs percentage in PLC will lower the global warming potential (GWP) of concrete production. Thus, it is indispensable to identify a suitable method for increasing the replacement ratio of PLC to maximize environmental benefits. While LFs addition will introduce a wide range of effects (e.g., nucleation, dilution, filler) that depend on material properties such as specific surface area (SSA), specific gravity (SG), particle size distribution (PSD) Etc., some of which are already at the microscopic scale. It is not reasonable to design PLC concrete the same way as traditional concrete, where many rough assumptions and safety factors are introduced to ensure the engineering properties and durability performance. The durability and long-term performance of the PLC mixtures must be assessed through lab accelerated tests (F&T cycles, concrete prism test, chloride penetration, sulphate attack) in such a way that all the fine (PLC, SCMs) and coarse (aggregates) particle fractions are characterized in terms of the PSD, SSA, SG, absorption and chemical composition to facilitate further analysis of their influence. Thus, advanced methods with comprehensive diagnostic ability, like PPM+MPs, should be refined to suit the PLC applications and propel the PLC standardization to a more sustainable class.

5 Conclusions

- The adoption of PLC containing higher LFs amount can significantly help in carbon footprint reduction, and it appears to be the most attainable material because of its availability worldwide and low cost.
- The disagreement among previous researchers in terms of the PLC concrete on durability performance can be caused by the variance among limestone fines properties (e.g., fineness) and material characterization (e.g., chemical compositions and particle size distribution) since they cannot be revealed by traditional concrete mix-design methods.
- The PPM-MPs method has been proved by previous researchers to have the ability to design low cement concrete with comparative fresh and hardened state behaviour, whereas progress needs to be made when implementing supplementary cementitious materials and considering LFs and clinker fractions integrally as Portland limestone cement.
- Almost all durability deterioration can be mitigated by optimizing the pore structure, where the particle packing model indicates an excellent tool to modify this property. Yet, lab tests will be needed to confirm this conclusion.

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