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Potential use of binary and composite limestone cements in concrete production

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HIGHLIGHTS

- The use of LS in binary, ternary and quaternary cementitious systems was investigated.
- Using LS permits the design of concrete with low environmental impact.
- Composite cement with LS seems to perform better than binary LS-cement.
- An optimal replacement level of 15% LS is recommended.
- LS composite cements provide a durability improvement compared to binary LS-cement.

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ABSTRACT

Over the last decades, the use of various by-products and pozzolanic materials in concrete production has become a common practice, not only to reduce the environmental impact of Portland cement (PC) manufacturing and to save natural resources but also to enhance the mechanical and durability performance of concrete.

The present study highlights the main performance properties of 50 concrete mixes designed with binary, ternary and quaternary cementitious systems, including the use of various proportions of slag (S), fly ash (FA), limestone (LS), silica fume (SF) and metakaolin (MK) as a partial replacement by weight of PC. The binary cements were designed with various LS proportions ranging from 10% to 45%, while the ternary system consisted of 29% slag and 21% FA as a partial substitute of PC. The three quaternary systems were designed with 25% FA and slag (50.1% or 47.5%) combined with either 4.9% SF, 4.9% MK or 2.5% LS. The concrete mixes were designed with a wide range of water-to-cementitious ratios (w/c) ranging from 0.45 to 0.79.

The main objective of this paper is to design a concrete with low environmental impact using various types and proportions of cementitious materials.

It has been observed that the use of composite cements improves concrete workability and reduces the amount of superplasticizer required to reach the same slump value compared with LS and PC cements, while the setting time is decreased for both LS-cement and composite cements. The strength results indicate that LS could lead to significant strength loss compared with PC and composite cement concretes. In addition, the quaternary PCSFALS mix appears to perform better than the binary LS-cement in terms of strength development and durability performance.

The results indicate that PCLS15 is freeze–thaw durable (durability factor over 80%); however, with replacement levels higher than 15%, the durability factor decreased. However, the composite cements generally exhibited a satisfactory durability factor of approximately 80% or a slightly lower DF. Moreover, the composite cements exhibited improved resistance to chloride ingress, while a negative effect on carbonation depth was observed.

Overall, the results indicate that the mechanical and durability performance of both binary and composite cement concretes are strongly linked to the chemical composition, fineness, particle size distribution and potential reactivity of the cementing materials used.

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1. Introduction

Since new environmental regulations have been implemented, the pressure to reduce CO_2 emissions generated by the concrete industry has increased. Over the last decades, extensive research has been undertaken to minimize the use of Portland cement by increasing the amount of various supplementary cementing materials (SCM) or fillers embedded in concrete as a partial replacement of PC. Indeed, it is well recognized that Portland cement manufacturing, with an annual production of 3.7 billion tons in 2012, accounts for approximately 7% of the global CO_2 emissions [1–3]. Various supplementary cementing materials, whether natural pozzolans or those derived from industrial by-product waste materials such as silica fume, fly ash and ground granulated blast furnace slag, have now been used for many years to develop composite cements not only to reduce the environmental load but also to improve concrete durability.

The use of limestone as a construction material dates back to ancient times when calcined limestone or gypsum was used to make mortar [4]. Limestone consists mainly of calcium carbonate ($CaCO_3$), which reacts with the tricalcium aluminate (C_3A) of Portland cement to form calcium carboaluminate (CCA). The effects of LS, as well as other mineral admixtures such as slag, FA, SF and MK used as partial replacements for Portland cement, have been widely discussed and are now well established and documented [5–14]. Natural pozzolana, fly ash, slag, silica fume and limestone are the main cementing materials that are permitted by the EN 197-1 [15]. According to BS EN 197-1:2000 [15], type II cements (CEMII/A-LL 32,5/42,5) may contain various materials as their main constituents in percentages ranging from 6% to 35%.

Although limestone has been widely used as a filler material, it is also used in blended cement as a partial substitution for Portland cement [15], with a recommended amount ranging between 6% and 20% [16]. It has been reported that fine limestone powder could promote the early age hydration of Portland cement [17,18] and may reduce the total porosity and delay the initial and final setting time as well [9]. While the setting time might be slightly shortened and the tendency to bleeding significantly reduced, the fresh properties, such as plasticity and water retention, in Portland-Limestone mortar and concrete are slightly improved or similar to those of a bulk Portland CEM I concrete [19].

In addition to its filler effect, LS also has a chemical effect: the calcium carbonate of the limestone powder can interact with the aluminate hydrates formed by the hydration reactions of Portland cement [18,20]. This interaction leads to the stabilization of the ettringite and could increase the total volume of the hydration products, decrease the porosity of the concrete and consequently increase its strength. Limestone powder could also interact with the AFm and AFt hydration phases, leading to the formation of carboaluminates at the expense of monosulfate, thereby stabilizing the ettringite, as reported by De Weerdt et al. [21].

It has been reported that the addition of LS to Portland cement increases the rate of hydration at early ages and consequently enhances the early strength; however, LS can reduce the 28-day and long-term strength compared with concrete prepared with CEM I due to the dilution effect [11,22]. Meanwhile, at the same concrete strength, Portland-LS cement concrete exhibits similar performance as CEM I concrete with respect to the carbonation rate, chloride ingress and resistance to freezing and thawing (both air-entrained and non-air-entrained concrete). It has been observed that depending on the amount used, limestone in concrete increases chloride ion diffusion [14] while significantly reducing the peak rate of heat evolution [16]. Moreover, Ghrici et al. [11] reported that a ternary cementitious system containing 20% LS filler and 30% natural pozzolans exhibited improved early and long-term compressive and flexural strengths and enhanced durability against sulfate, acid and chloride ion ingress.

However, some studies have focused on the "thaumasite problem" linked with the use of limestone-cement concrete and calcareous aggregates. The risk of thaumasite formation ($CaSiO_3 \cdot CaCO_3 \cdot CaSO_4 \cdot 15H_2O$) is a serious problem associated with the use of limestone in cement and concrete exposed for a few months to sulfate solutions at low temperature (approximately 5 °C) [19,23–25]. It is believed that this form of sulfate attack completely destroys the binding ability of the cement by transforming the C–S–H gel into a mush, weakening the C–S–H and leading to lower strength. Thaumasite formation requires a source of calcium silicate, sulfate and carbonate ions, excess humidity and preferably low temperature [24,26].

In this paper, the effect of various limestone contents (15–45%) in binary cement and various amounts of fly ash, slag, silica fume, metakaolin and limestone in composite cements (ternary and quaternary systems) on the resulting concrete performance is studied. The results presented herein are part of an extensive research project aiming to develop concrete made with various Portland-composite cements to reduce the environmental load and design more sustainable and durable concrete.

2. Experimental work

2.1. Materials

General use Portland cement CEM I 42.5 N meeting the requirements of EN 197-1:2000 was used in all the mixes with and without SCMs. Five different SCMs, namely, slag, fly ash, silica fume, metakaolin and limestone, were used in various proportions as substitutes for Portland cement to produce binary, ternary and quaternary binders. The chemical and mineralogical compositions and physical properties of Portland cement and the five cementing admixtures (S, FA, SF, MK and LS) used are listed in Table 1, and their particle size distribution is shown in Fig. 1.

Natural siliceous sand and crushed granite with maximum sizes of 5 mm and 20 mm, respectively, were used as fine and coarse aggregates. A superplasticizer (SP) was used to obtain a nominal target slump value of 75 ± 5 mm, while an air-entraining agent was employed to investigate the freeze–thaw resistance of the air-entrained concrete mixes.

2.2. Details of mixtures, concrete mixing and specimens

Both the Portland cement and blended cement concretes were designed with a wide range of w/c ratios of 0.79, 0.65, 0.60, 0.52 and 0.45. Six types of mixes designed with five different SCMs were investigated in this study. A single content of 4.9% (by weight) of SF and MK was introduced in the quaternary systems, while FA was used in proportions varying from 20% to 25% and slag was incorporated at two different proportions of 29% and 25%. In addition, LS was introduced at 2.5% in the quaternary system, while the LS-binary cements were formulated by varying the replacement (by weight) of PC by LS from 15% to 45%. In all the mixes, the free water (185 kg/m^3) and coarse aggregate (1200 kg/m^3) contents were kept constant, while the fine aggregate content was adjusted depending on the type and content of SCM incorporated. Tables 2 and 3 provide the mix proportions of the PCLS concretes and the various composite cement concretes investigated, respectively.

All the concrete mixtures were produced using a horizontal forced-action pan mixer with a 0.045 m^3 capacity, and each mix was appropriately labeled. The concrete mixes were referred to as control Portland cement (PC), binary cement-limestone (PCLS), ternary cement–slag–fly ash (PCSFA) and quaternary systems cement–slag–fly ash–silica fume (PCSFASF), cement–slag–fly ash–metakaolin (PCSFAMK) and cement–slag–fly ash–limestone (PCSFALS).

After mixing, a slump test was performed, and then, the concrete was cast into steel molds (cubes, cylinders and prisms) in three layers and compacted using a plate vibrator, as specified by BS 1881: Part 108: 1983. All the concrete specimens were stored for the first 24 h under a plastic sheet in a laboratory environment. The specimens were then demolded and wet-cured at 20 ± 2 °C for the first 28 days, and afterwards, specific curing conditions were applied depending on the durability test. For all the concrete types, three samples of Portland and blended cement concretes (binary, ternary and quaternary) were tested.

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