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Development of concrete mixtures with fine lime-pozzolan binder as supplementary cementitious material

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Abstract: The very high concrete strength and durability achieved in high and ultra-high-performance concrete is associated with a very dense matrix, accomplished through the use of high volumes of very fine mineral additions, such as fly ash, silica-fume, metakaolín and ground quartz sand. The paper reports on a study where a finely ground lime-pozzolan binder (LPB) is used as active mineral addition in concrete. The very fine lime particles, having size between0.1-10µm, can fill the gaps between cement grains, while the larger pozzolan particles, having size between 10-100µm, can fill the gaps between fine aggregate grains; this results in a much denser matrix. The addition of lime during concrete mixing also increases the Ca^{+2} and OH^- ion concentration, which results in a better and faster hydration of both ordinary portland cement (OPC) and pozzolanic reaction products.The use of LPB as an active addition in some ultra-high performance concretes could lowe the cost of the product for equivalent strength and durability performance, through the use of less cement, thus improving the ecologic profile of the material. Results from an initial series of tests are examined in this paper; further testing is required to establish the benefits of the use of LPB in High Performance Concrete. Examples of applications of this work in concretes are also presented.

Key words: mineral addition; lime-pozzolan binder; compression strength; lime; pozzolan; concrete

1 Introduction

Portland cement has contributed substantially to the economic development of modern society; however, its manufacture, which relies on the use of non-renewable resources, has led to the production of large quantities of greenhouse gases. Global production of Portland cement has experienced spectacular growth; in 2000, it stood at 1.75×10^9 tons, with an annual growth rate of 3.5% since 1970. Production is expected to continue increasing in the coming years, potentially reaching increases of between 120% and 180% by 2020.

The measures to be taken to reduce CO_2 emissions during cement production can be classified into two main groups: the first involves increasing the efficiency of the production process by reducing oil consumption, and the second by reducing clinker production through the incorporation of mineral additives during the production process or at the time of concrete manufacturing. (Price et al., 1999; Vanderley 2002) The contribution of any of these measures to reducing energy consumption and emissions is small compared to projected future cement production. However, while the goal is to replace up to 50% of fossil fuels, in practical terms only 15% has been achieved so far. (Schmidt, 2003) Furthermore, the reported use of mineral additives has achieved only a 22% reduction in emissions. Technological improvements depend on the economic level of the country where production takes place. [Schmidt, 2002]

In order to maintain similar levels of energy consumption and emissions in 2014, the cement industry is required to reduce emissions by 50%. This will require a paradigm shift in the production and use of Portland cement to comply with current environmental requirements. One solution to the problem involves reducing the amount of Portland cement used as a binder in concrete production.

Advances in the science of concrete, the use of modern techniques in researching the chemistry and microstructure of concrete, as well as developments in the chemical admixture industry, have opened new avenues for the use of concrete as a modern construction material. These results have revealed new avenues for the development of high-strength, durable concretes using small amounts of Portland cement. The use of small amounts of Portland cement and large amounts of pozzolanic admixtures and other byproducts, combined with high-dispersing-power additives, is seen as an attractive way to improve the environmental profile of concrete.

Currently, there is a wide variety of blended cements. The inorganic materials used to reduce cement consumption can be intimately mixed and/or ground with the clinker and/or blended during its manufacture, or mixed during the production of concrete or mortars. Fly ash, granulated slag, microsilica, and other natural or calcined pozzolans are reported as the most commonly used mineral additives. The nature of the products formed during the pozzolanic reaction in blended cements depends on the properties of the pozzolans and the clinker used to produce the concrete. The main reaction products are calcium silicate hydrate (CSH) and small amounts of ettringite, hydrogarnet, and hydrated aluminates. The relative proportions of the reaction products depend on the chemical characteristics and mineralogy of the pozzolans used. (Day, 1992; Taylor, 1993).

Little is known about the reactions and reaction mechanisms in a supersaturated HC solution that may exist when using either a lime-pozzolan binder or blended cement. Apparently, the excess creates a situation of nucleation of additional undissolved HC, mainly due to its high specific surface area. The early presence of HC appears to accelerate the pozzolanic reaction, likely due to the large amount of available Ca^{2+} ions in the solution. (Williams et al., 2002)

The use of Cal-pozzolan binder as an active mineral admixture in concrete could help reduce production costs without compromising the material's mechanical strength and durability by using less cement, thereby improving the material's environmental profile. Initial results are presented in this study, though further testing is required to establish the benefits of using Cal-pozzolan binder in concrete. An example of the application of this work in concrete is also presented.

2 Discussion and development

2.1 Use of mineral admixtures in concrete

High-fineness mineral admixtures can help improve the properties of concrete. These effects may be physical, such as increased compactness, or physicochemical, such as the new reaction products formed during the pozzolanic reaction. In both cases, the final effect is similar: the porosity of the concrete decreases, and the distribution and size of the pores become smaller. The use of pozzolans can modify the rheological and mechanical properties as well as the durability of concrete.

In normal-strength concretes, pozzolans are added to reduce costs and improve the strength and durability of the hardened mass. In such cases, pozzolans help improve the compactness of the solids, but their primary role is to provide additional hydrated calcium silicate through reaction with water and with the calcium hydroxide resulting from the reaction of Portland cement. This pozzolanic reaction is minor for most pozzolans used in high proportions; thus, the benefits are observed within one week to several weeks after mixing. Some of these highly reactive pozzolans (such as silica fume) are added in small proportions and help improve early-age strength values as well as durability at later ages. (Shannang and Yeginobali, 1995; Singh, 2000).

In high- and ultra-high-performance concretes, the primary goal is to optimize particle size distribution, especially for fine particles. The use of high-dispersing- power plasticizers allows for mixtures with a low water-to-binder ratio. The resulting concrete has high strength, high density, and consequently low porosity. In many of these mixtures, however, the replacement of Portland cement is less than 15% (Malhotra and Mehta, 1996; Zhang et al., 1996; Aitcin, 2000). Conservatively, in concretes with high fly ash content, mineral admixtures are much higher than in ordinary Portland cement concretes, and the water-to-binder ratio is much lower (on the order of 0.35). The 28-day strength is in the range of 60–90 MPa, far below the values obtained in concrete with 100% Portland cement. (Bouzoubaa et al., 1998; Lam et al., 2000; Poon et al., 2000).

Mineral or pozzolanic admixtures perform a dual function in these cases. The fine pozzolanic particles fill the voids between the cement grains and among the remaining pozzolanic grains, improving compactness. Only a small portion of the pozzolanic admixtures, less than 20%, reacts. The compressive strength, however, does not correspond to the low level of hydration achieved; it is attributed to the contribution of the electrostatic interaction between the finer fly ash particles. (Lam et al., 2000; Qualin et al., 2003).

The pozzolanic reaction in many pozzolans is significant after 7 days, when most of the cement reaction products have already formed and the alkaline concentration is high enough to break bonds and facilitate the formation of cement reaction products. In many cases, depending on the pozzolan's reactivity, many of the reactions are completed within the first 60 days. (Shannang and Yeginobali, 1995; Jamal, 1995; Malhotra and Mehta, 1996). However, the use of large volumes of pozzolanic admixtures increases the risk of self-neutralization due to excessive consumption of calcium hydroxide during the pozzolanic reaction. A significant drop in pH can cause the dissolution of other reaction products and the destruction of the cementitious matrix. (Groves and Richardson, 1994).

2.2 Lime-pozzolan binder as an active mineral additive in concrete

The properties of concrete with high replacement ratios can be improved by replacing cement with a lime-pozzolan binder rather than with pozzolans alone. The presence of additional lime reduces the risk of self-neutralization, even for high replacement ratios. The presence of lime increases the concentration of Ca^{2+} ions; this is due to its early contribution to the formation of reaction products. The presence of lime leads to an increase in OH^- ions, whose role is to break the silicate bonds in the pozzolan, thereby accelerating the onset of the pozzolanic reaction.

The effects of lime can be observed in three distinct stages:

1. During mixing, fine lime particles occupy the voids between cement grains and restrict water flow, thereby increasing water retention in fresh concrete, acting as a dispersing agent that prevents flocculation and increases the plasticity of the mixture. (Swamy, 1986; Malhotra and Mehta, 1996).

2. At early ages, lime helps increase the compactness of the concrete, since the lime particles, being so fine, have not been completely dissolved and fill the voids between the cement grains

3. At later ages, the fine hexagonal plates of 2 μm to 5 μm occupy the spaces between the reaction products, as shown in Figure 1, an SEM image of a 28-day-old paste sample in which 60% of the OPC has been replaced by a lime−pozzolan binder. The hexagonal plates are highly visible very close to the CSH phase.

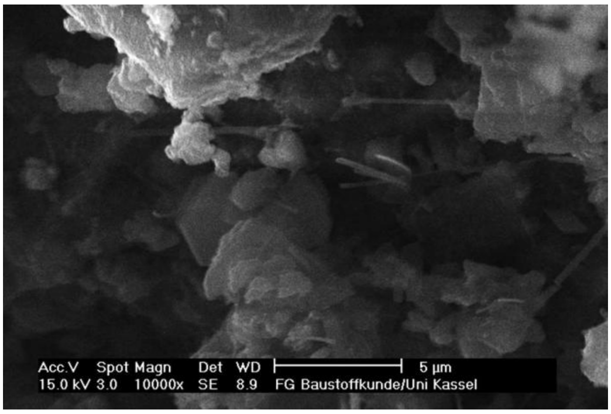

Figure 1. Hexagonal HC lamellae observed under SEM, OPC/LPB paste, 28 days

The lime-pozzolan binder used in this study consists of a mixture of 20% lime and 80% pozzolan by weight, which, when mixed and ground together for one hour into a fine powder, achieves a fineness similar to that of OPC. The pozzolan used was a zeolitic tuff (Zeolite) obtained from abundant deposits in Cuba.

Through the grinding process, the particle size distribution of the pozzolan is improved, compensating for the effect of the irregular shape of the grains. The interaction between the lime and the pozzolan during grinding is also of great interest. Since lime is much softer than pozzolan, it achieves a finer texture. (Martirena, 1994). Figure 2 shows the particle size distribution obtained by laser particle size analysis of the powders used, where the grain sizes can be seen: lime between 1–30µ m, and much larger pozzolana, between 10–100µm.

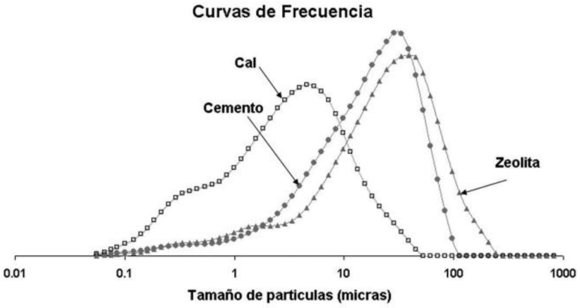

Figure 2. Particle size distribution of OPC, HC, and zeolite

Figure 3 shows the results obtained in the conductometry test, where a loss of conductivity over time is observed, allowing us to infer the reactivity of the studied pozzolan in calcium hydroxide solution.

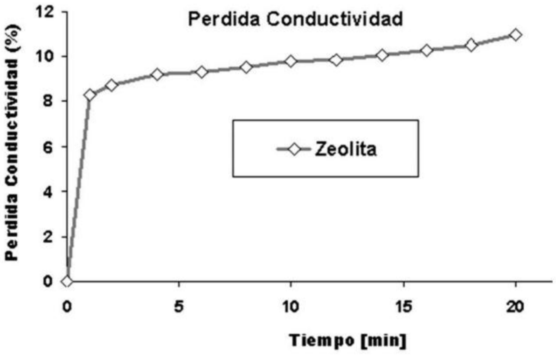

Figure 3. Loss of conductivity in zeolitic tuff

2.3 Concrete with high volumes of Cal-pozzolana admixtures

With the aim of optimizing OPC consumption in concrete mixtures, the relationship between compressive strength and cement content must be properly interpreted. A denser and stronger cementitious matrix will not be achieved simply by increasing the amounts of OPC; higher amounts of cement would lead to increased mixing water demand and, consequently, increased porosity and permeability in the hardened concrete matrix. In contrast, using highly finely ground mineral admixtures as partial substitutes for OPC content, in combination with superplasticizers, would allow for the use of concrete mixtures with a low water-to-binder ratio. This would result in a reduction of microcracks associated with shrinkage and ensure better conditions for the hydration of the OPC used. Similarly, the particles of the additive would complement the particle size distribution of the fines, potentially filling the voids between cement particles and the spaces between the small aggregate particles. This principle can be applied to any type of concrete, as has been done in the production of self-compacting and high-and ultra-high-strength concretes, with the advantage of achieving excellent mechanical properties and durability while significantly reducing OPC consumption.

To assess how much OPC can be substituted without altering the rheological properties of the mixtures, the authors propose a method that allows for the correction of the proportions of traditionally designed mixtures by incorporating high volumes of Cal-pozzolanic mineral admixtures. The proposed correction consists of two main stages.

The first stage involves determining the amount of OPC by weight that can be replaced by adding a lime-pozzolan admixture, in combination with a superplasticizer, without causing significant changes in the rheological properties of the mixture. To this end, experimental mixtures will be prepared by partially replacing the OPC content with an equal weight of lime-pozzolan admixture, for example 10%, 20%, 30%, adjusting the water content for each mix to maintain consistency within the specified range. Early-age compressive strength tests on cylindrical specimens will be conducted for each mix.

The limit for replacing OPC by weight with lime-pozzolan mineral admixture will be defined by the mix in which the compressive strength values show no significant changes compared to those of the control mix and where the water-to-binder ratio does not increase significantly (less than 20%). The paste volume (Vp) defining this mixture is considered adequate to ensure the required consistency and compressive strength values in a mixture with partial substitution of the OPC content.

The use of LPB causes changes in the aggregate-to-paste ratio. Since the mineral additive has a lower density than OPC, it increases the paste volume in the mixture, thereby also increasing the separation between aggregate grains. Figure 4 clearly shows that in a mortar where 40% of the OPC has been replaced, the separation between aggregate grains has increased due to the rise in paste volume.

The second stage involves determining the volume of OPC that can be replaced by adding a lime-pozzolan mineral admixture in combination with a superplasticizer without causing changes in the desired compressive strength and consistency values. In this stage, while keeping the paste volume determined in the previous step constant—so that the water-to-fines volume ratio remains constant, as proposed by Bornemann and Schmidt (Bornemann, 2002; Schmidt, 2003), and with the rest of the constituents also kept constant, the volume proportions of OPC replaced by LPB are varied to determine the mixture in which the compressive strength and consistency meet the expected values. This mixture will allow the minimum amount of cement required to be established.

It stands to reason that the more reactive the pozzolan used, the greater the possibility of reducing OPC consumption within this constant paste volume, due to the contribution of the additive in the form of hydrated products.

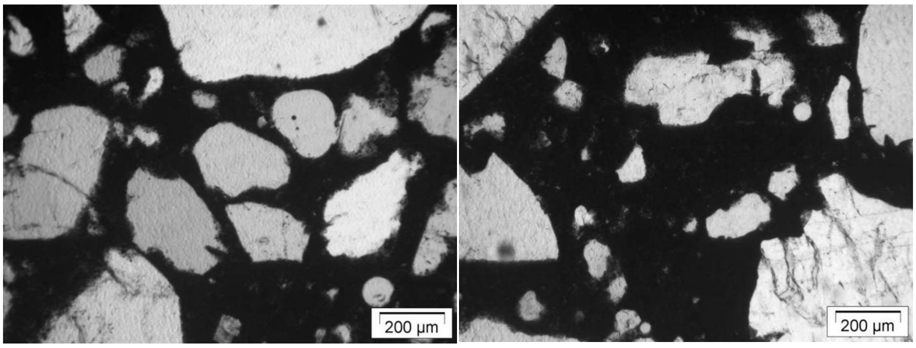

Figure 4. Mortars with 100% OPC (a) and 40% LPB (b)

2.4 Example of the application of the proposal to replace OPC with LPB

As an example of the application of the principles described in this paper, a structural concrete mix was developed to achieve 30 MPa compressive strength at 28 days with 12±1 cm of settlement measured using the Abrams Cone. Crushed aggregates of limestone origin, maximum size 9.52 mm for coarse aggregate, fineness modulus of 2.56 for fine aggregate, Portland cement P-350 according to Cuban standard NC 95, chemical admixture Mapefluid N-200, superplasticizer type F according to ASTM C-494. Mineral admixture consisting of 20% by weight of calcium hydroxide (72% reactive) and 80% by weight of zeolitic tuff, ground together in a ball mill for one hour, achieving a fineness of 12% on a 170-mesh sieve. Table 1 shows the chemical and physical analysis of the binders used.

Table 1. Chemical and physical analysis of binders

	OPC	Cal	Zeolite
Chemical (by mass)			
SiO_2	70.4	0.88	74.68
Fe_2O_3	1.38	0.29	2.87
Al_2O_3	2.42	0.34	12.69
CaO	9.76	97.42	4.89
MgO	2.28	0.59	0.53
K_2O	3.6		1.28
Na_2O	0.23		2.9
SO_3	0.35	0.47	0.03
Physical properties			
Blaine Specific Surface (cm²/g)	2974	7656	3425
Specific Gravity	3.15	2.46	2.29

Stage I: Table 2 lists the components of the mixtures tested with OPC replacement ranging from 0% to 40% by mass through the addition of calcium-zeolite mineral, and Figure 5 shows the results of the compressive strength tests. The mixture in which 10% of the OPC content has been replaced produces an increase in compressive strength values above those of the mixture with 100% OPC. In contrast, above 30% there is a sharp drop in strength. The mixture prepared with 30% OPC substitution by lime-zeolite mineral addition shows no significant changes in compressive strength values at 28 days compared to those of the control series, and is therefore defined as the mixture that establishes the limit for OPC substitution by weight with mineral addition.

Table 2. Proportions of the mixtures prepared in stage I

Materials	UOM	Proportion LPB/OPC (by mass)				
		0%	10%	20%	30%	40%
Cement	kg/m^3	402.12	365.58	317.65	275.66	232.84
Lime-Pozzolan	kg/m^3	0.00	40.62	79.41	118.14	155.22
Water	kg/m^3	159.84	145.22	157.83	158.50	164.44
Fine Aggregate	kg/m^3	834.40	842.86	823.90	817.12	805.23
Coarse Aggregate	kg/m^3	940.96	950.51	929.12	921.48	908.07
Superplasticizer	kg/m^3	4.82	4.87	4.78	4.72	4.66
Paste Volume	L/m^3	287.50	280.47	296.19	301.84	311.70
w/c Ratio (water/cement, by mass)	-	0.40	0.40	0.50	0.57	0.71
w/b Ratio (water/binder, by mass)	-	0.40	0.36	0.40	0.40	0.42
Slump	cm	13	12	11	12	12

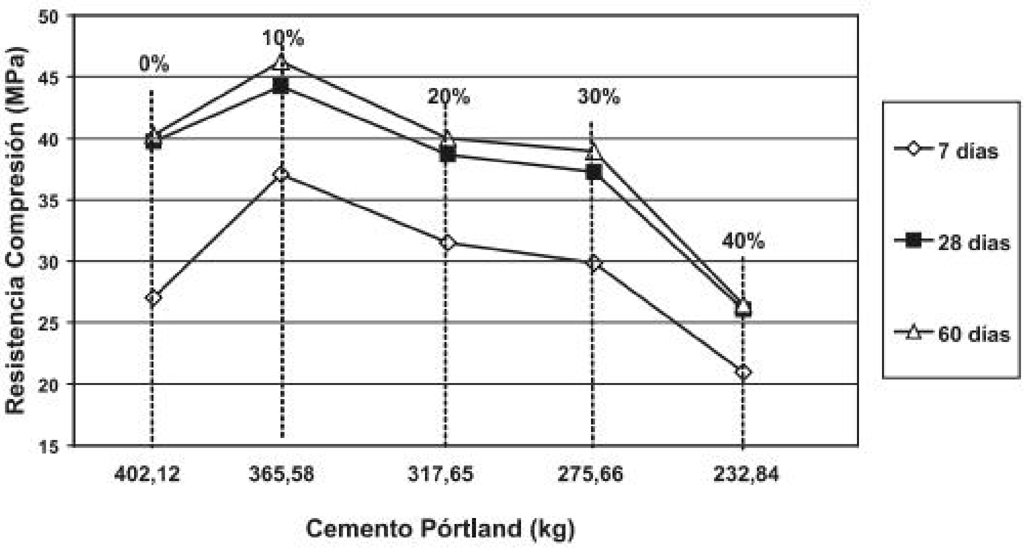

Figure 5. Compressive strength results, stage I

Stage II: The 30% weight substitution mixture selected in the previous step represents, in turn, a 39% volume substitution, hereafter 40%. Keeping the paste volume constant at 301.84 liters, so that the volumetric water-to-fines ratio is also constant (1.07), the proportions between OPC and LPB were varied in volume for points above and below 40%. Table 3 shows the dosages of the mixtures produced, and Figure 6 shows the compressive strength results obtained

Table 3. Proportions of the mixtures prepared in stage II

Materials	UOM	Proportion LPB/OPC (by volume)				
		20%	30%	40%	50%	60%
Cement	kg/m^3	361.22	316.06	275.66	225.76	180.61
Lime-Pozzolan	kg/m^3	60.93	91.42	118.14	152.37	182.84
Water	kg/m^3	158.50	158.50	158.50	158.50	158.50
Fine Aggregate	kg/m^3	817.12	817.12	817.12	817.12	817.12
Coarse Aggregate	kg/m^3	921.48	921.48	921.48	921.48	921.48
Superplasticizer	kg/m^3	4.72	4.72	4.72	4.72	4.72
Paste Volume	L	301.84	301.84	301.84	301.84	301.84
Water/fines Ratio	-	1.07	1.07	1.07	1.07	1.07
w/c Ratio (water/cement, by mass)	-	0.44	0.50	0.57	0.70	0.88
w/b Ratio (water/binder, by mass)	-	0.38	0.39	0.40	0.42	0.44
Slump	cm	12.00	13.00	12.00	12.00	11.00

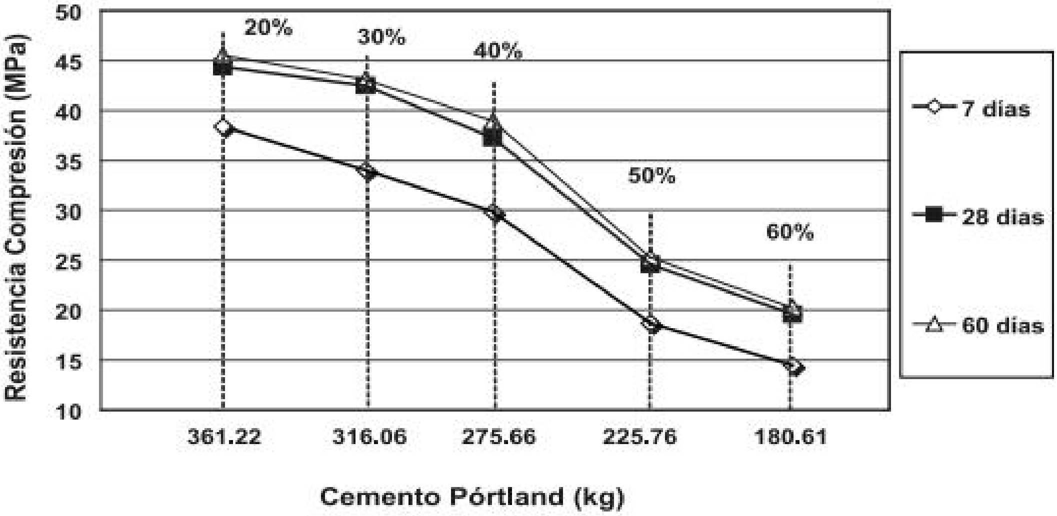

Figure 6. Compressive strength results, stage II

The results obtained identify the 40% mixture as the mixture representing the limit for replacing OPC by volume with lime-zeolite; higher replacement values cause compressive strength values to drop sharply, resulting in concrete with normal strength but with significant reductions in cement consumption. conversely, lower replacements of OPC content result in high-performance concrete according to Cuban standard NC-120, even for OPC contents below those required by the control mix. All mixes maintain their consistency values. The results of the tests confirm that large amounts of OPC can be replaced by LPB without affecting the 28-day strength requirements or the consistency of the concrete.

The specimens tested in Stage II show better performance in compressive strength values between 20% and 30% when compared to the results of Stage I for similar mass contents of OPC per cubic meter. Increases in compressive strength at 60 days are not appreciable when compared to the results at 28 days.

2.5 Influence of the lime-pozzolan addition

One might think that adding lime beyond what is produced by OPC during hydration could be detrimental to the mixture, as the presence of free lime is a weakness for matrices in aggressive environments. For this reason, it was decided to evaluate the amount of free lime in pastes where OPC is partially replaced by LPB.

Figure 7 shows the X-ray diffractograms of pastes manufactured with different OPC/LPB ratios. It can be observed that for higher values of OPC substitution by the addition of lime-pozzolan, the intensity of the calcium hydroxide peaks is lower, suggesting considerable pozzolanic reactivity during the first 28 days of reaction.

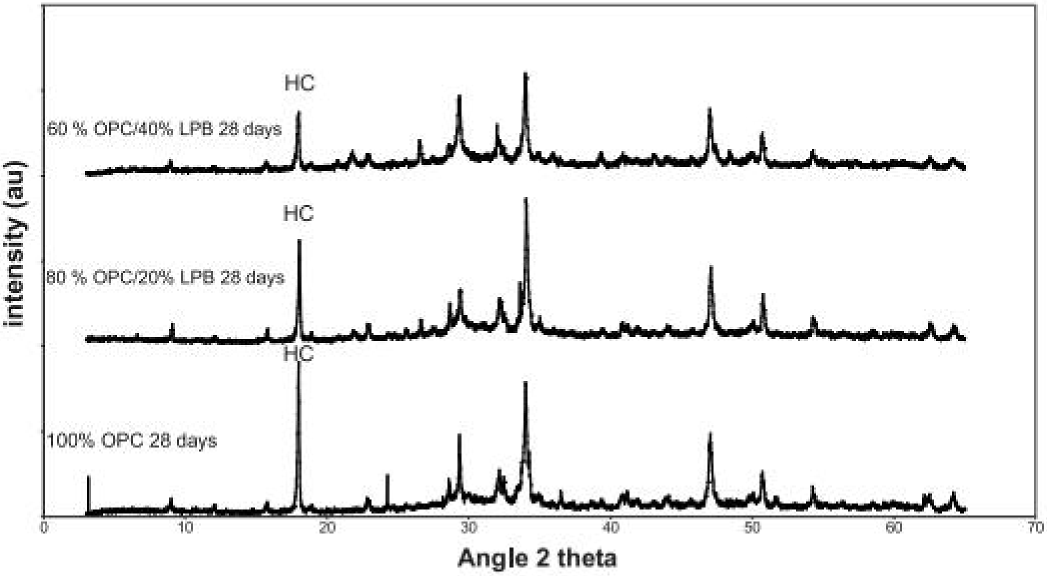

Figure 7. X-ray diffractograms of hydrated OPC and OPC/LPB pastes

It was decided to verify this result with TG analysis of pastes with 0%, 20%, and 40% OPC substitution by LPB, cured for 28 days without exposure to the environment to prevent carbonation. The mass loss versus temperature curves allow for the determination of water loss during the dehydroxylation process in the temperature range from 470°C to 550°C. Given the proportions of the pastes, the theoretical calcium hydroxide contents in the pastes can be calculated, and with that, the degree of pozzolanic activity. Table 4 shows the calculations performed.

Table 4. Determination of calcium hydroxide consumption

OPC (g)	LPB (g)	Theoretical Mass of HC Present	Calculated Mass of HC Present	HC Consumed	HC Consumed per (g) of Pozzolan
100	0	8.34	8.34	0	0
80	20	10.67	8.75	1.92	0.12
60	40	13	5.34	7.66	0.24

Analysis of the results in Table 4 confirms the presence of pozzolanic activity, also indicating that this activity is much higher as the proportion of LPB used increases; the paste with a 60 OPC/40 LPB ratio consumes 0.24 g of HC per gram of pozzolan, whereas in the 80:20 ratio, only 0.12 g is consumed. This indicates that the addition of lime has beneficial effects on the matrix, as it apparently increases the volume of stable products resulting from the pozzolanic reaction.

3 Conclusion

The use of the lime-pozzolan admixture allows for a reduction in the amounts of Portland cement used in high-performance concretes, constituting an attractive proposition from an environmental standpoint. The test results reported in this study correspond to normal-strength concretes, but the principles are also applicable to high-strength concretes.

The results suggest that the amount of cement can be reduced without affecting the expected strength values and consistency by using the described procedure, which allows determining the substitution limit of OPC by LPB in combination with a superplasticizer while maintaining a constant water-to-fine aggregate ratio within the mixture.

From a durability standpoint, the use of high levels of calcium hydroxide in the fresh mix is of interest. Preliminary tests indicate that the added lime combines with the pozzolan to produce new stable reaction products, and that the amount of free lime in the matrix does not increase. Implementation on a larger technological scale suggests conducting additional tests to evaluate the transport behavior of aggressive substances within the specimens under various exposure conditions, in order to establish the limits for replacing OPC with LPB that are appropriate for durability requirements.

Conflicts of interest

The author declares no conflicts of interest regarding the publication of this paper.

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