

**Heft 3**  
**No. 3**

**Schriftenreihe Baustoffe und Massivbau**  
**Structural Materials and Engineering Series**

**U N I K A S S E L**  
**V E R S I T Ä T**

# **Ultra High Performance Concrete (UHPC)**

**International Symposium on  
Ultra High Performance Concrete  
September 13-15, 2004**

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## **Lime-pozzolan binder as a very fine mineral admixture in concrete.**

### **Summary**

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, metakaolin 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 between 0.1-10  $\mu\text{m}$ , can fill the gaps between cement grains, while the larger pozzolan particles, having size between 10-100  $\mu\text{m}$ , 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  $\text{Ca}^{+2}$  and  $\text{OH}^-$  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 contribute to lowering 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 LPC in UHP concrete. Examples of applications of this work in normal and high-performance concrete are presented.

### **Introduction**

Although portland cement has contributed substantially to the development of our modern society and economy, its manufacture is based on the intensive use of non-renewable resources, and the production process produces large quantities of greenhouse gases. World cement production grows at spectacular rates. In 2002 it was  $1.75 \times 10^9$  tons, with an average yearly growth of 3.5%, measured since 1970. These production figures will continue to increase in the coming years as large segments of the world's population move to a higher level of industrialization. Cement production growth is estimated to range between 120-180% in 2020 [1].

Measures to decrease the rate of  $\text{CO}_2$  emissions during cement manufacture can be classified in two main groups: a) attaining a higher efficiency in the production process, by decreasing fuel consumption; b) reducing the rate of clinker production by using mineral re-

placements, added during manufacture or directly through cement replacement at the building site [1,2]. The contribution of any of these measures to the reduction of energy consumption and emissions are small in comparison to the projected production increase. Although the goal is to replace 50% of fossil fuel by alternative fuels, in practical terms only 15% has so far been achieved [3]. Moreover, the reported use of mineral additions has produced only a 22% emissions reduction. The technological improvements depend on the economy of the country where the production takes place [3].

In order to have the same levels of energy consumption and emissions in 2014 than those existing in 2004, the cement industry is challenged to reduce emissions by more than 50%. This implies a change of paradigms in production and use of portland cement, to make it comply with environmental requirements. Any solution of the problem includes the reduction of the amount of portland cement in the binding materials used to make concrete.

Progress in our knowledge of the science of concrete, the use of modern techniques to investigate the chemistry and microstructure, as well as the dynamic developments in the chemical-admixture industry, has opened new perspectives for the use of concrete as a modern construction material. These developments have revealed new routes towards the development of high strength, high durability, concretes that contain reduced quantities of portland cement. The use of less cement and larger amounts of pozzolanic minerals and other wastes, combined with highly active dispersing agents seems to be an attractive way to improve the environmental profile of concrete.

There are now a wide variety of blended cements available [4-7]. The inorganic materials that are used to reduce cement quantities can be blended and/or ground intimately with clinker and/or cement during manufacture, or blended while preparing the concrete or mortar. The most commonly used minerals are fly ash, granulated slag, micro silica (silica fume) and various natural and calcined pozzolans [4-7]. The nature of the product formed during the pozzolanic reaction in blended cements depends on the properties of the pozzolan and the clinker used to manufacture concrete. The main reaction products are calcium silicate hydrate (CSH), and smaller amounts of ettringite, hydrogarnet and hydrated aluminates; the relative proportions of the product depends upon details of the chemistry and mineralogy of the pozzolan used [4, 7].

Little is known about reactions and reaction rates in CH supersaturated solutions that may exist when using either lime-pozzolan binders or in blended cements. Apparently, the excess, undissolved, CH creates additional nucleation sites mainly because of its high specific surface. The early presence of CH in the mix seems to accelerate the pozzolanic reaction, probably because of the high amount of  $\text{Ca}^{2+}$  ions immediately available in the solution [8].

#### **The use of mineral admixtures in concrete**

Fine-grained mineral additions can help improve many qualities of concrete. There can be physical effects such as increased packing, or chemical-physical effects such as new reaction products formed during the pozzolanic reaction. In both cases the end effect is similar: the concrete porosity decreases and pore size distribution shifts towards smaller size fractions. The use of pozzolans can modify any or all rheological, mechanical and durability properties of concrete.

In regular-strength concrete, pozzolans are added to reduce cost and to improve the longer-term strength and durability of the hardened mass. In such cases the pozzolan helps to improve the packing density of the solids, but the primary role of the pozzolan is to provide additional calcium-silicate-hydrate through reaction with water and the calcium hydroxide contributed by the reaction of portland cement. This pozzolanic reaction is slow for most pozzolans that are used in high proportion; thus, the benefits of the pozzolan are seen within the time-frame of a week to several weeks after casting. Some of the finer, more highly reactive

pozzolans (such as silica fume) are added in smaller proportion and help to improve early strengths as well as durability at later ages [9, 10].

In high and ultra-high performance concrete, the primary aim is to optimize the grain size distribution, especially of the fine particles. The use of superplasticizers and high-quality aggregates permits fluid mixes with very low water to cementing-materials ratio. The resulting concrete has a very high strength and density and very low porosity. In many of these mixes, however, the replacement by mass of portland cement is less than 15% [5, 11-13]. Conversely, in "high volume fly ash concrete" (HVFA) the mass of mineral addition is higher than that of ordinary portland cement (OPC), and the water/binder ratio is very low (under 0.35). Twenty-eight days strength is in the range of 60-90 MPa, far higher than the control series manufactured with 100% OPC [14,15,16].

The pozzolan or mineral admixture plays a double role in these cases. The pozzolanic particles fill the empty spaces between cement grains and between other pozzolanic particles and improve packing. Only a small part of the added pozzolan, less than 20%, reacts. The compressive strength, however, does not correspond to the low level of hydration achieved. Some have attributed the strength increase to the contribution of electrical interaction between the smallest fly ash particles [15-17]

The pozzolanic reaction in most pozzolans becomes significant after the seventh day, when enough cement reaction products already exist and the alkali concentration is high enough to break chemical bonds and enable the formation of cement reaction products. In most cases, and depending on the reactivity of the pozzolan, most of the reaction is finished in the first 60 days [5, 9, 18]. However, the use of large volumes of pozzolanic additions increases the risk of self-neutralization because of excessive consumption of CH during the pozzolanic reaction. A significant drop in pH can result in the dissolution of the other reaction products and destruction of the cementing matrix [19].

#### **Lime-pozzolan blends as fine mineral admixture in concrete**

The properties of concrete with large volumes of pozzolan can be improved by replacing cement with lime-pozzolan blends (LPBs) rather than with pozzolan alone. The excess lime available immediately upon mixing reduces the risk of self-neutralization, even for large replacement volumes. The early presence of lime increases the  $\text{Ca}^{2+}$  concentration; it is hypothesized that this contributes to a faster formation of the reaction products. The presence of lime brings about an increase in  $\text{OH}^-$  ions, whose task is to break the silica bonds in the pozzolan; this can accelerate the start of the pozzolanic reaction.

The effect of lime can be seen in three different stages:

- a) During mixing the very fine particles of lime occupy the empty spaces between the cement grains and limit the flow of water, thus helping to increase water retention in the fresh mix. The smallest particles with high specific surface can adsorb on the surface of cement grains, thus acting as a dispersing agent that prevents flocculation and increases mix plasticity [5, 6].
- b) At early age the lime helps increase concrete packing, as the small lime grains with size between 1-30  $\mu\text{m}$ , have not yet been completely dissolved and fill the gaps between cement grains.
- c) At later ages the 2-5  $\mu\text{m}$  hexagonal plates occupy spaces between the reaction products. Fig 1 presents an SEM picture of a sample of a 28 days paste where 60% of the OPC has been replaced by an LPB. The hexagonal plates are quite visible, very close to the CSH phases.

A pozzolan for use in an LPB must be highly reactive and finely ground. Many authors state that using natural pozzolan and agriculture ashes brings about an increase in water demand in the mix, because of the irregular shape of the particles and the high internal porosity.

However, when these particles are ground to fine powder, the internal structure is destroyed and the grains become more regular in shape and the internal porosity is thus reduced [20].

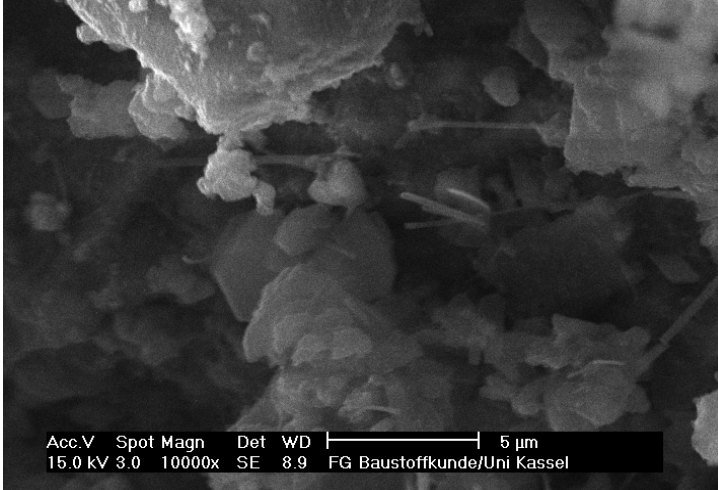


Fig. 1: Lime hexagonal plate crystals observed on the SEM on a 28 days OPC-LPB paste

Figure 2 shows a sugar-cane straw ash (SCSA) grain ground to an average particle size of approximately 10- $\mu$ m. It has a cubic to spherical morphology with fewer visible internal pores, presumably because they were destroyed at grinding. The LPBs used in the present study consist of an interground blend of 20% lime and 80% pozzolan. The pozzolan used was either sugar cane straw ash or a natural pozzolan (zeolite) found in Cuba. There was no significant difference in engineering performance between equivalent mortars and concretes made with these two pozzolans.

Mixing and grinding the pozzolan with lime should be done until the fineness of the powder equals that of OPC. This fineness level can be accomplished after only one-hour grinding in a batch ball mill. Through grinding, the size distribution of the pozzolan is improved, thus compensating for the effect of the irregular shape of the grains. The interaction between pozzolan and lime during grinding is also of great interest. Being softer, lime is more finely ground than the pozzolan. Figure 3 presents a typical grain size distribution for the pozzolan the CH and a portland cement. The resulting grain size of the lime particles lies between 1 - 30  $\mu$ m. The pozzolan fraction has a larger grain size -- between 10-100  $\mu$ m [21].

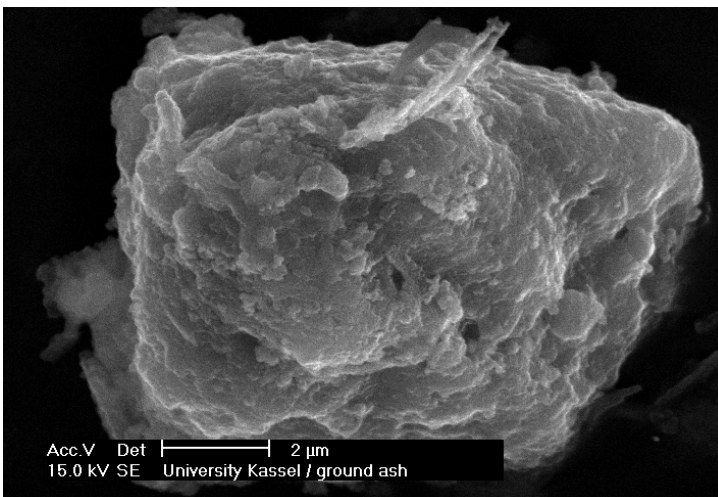


Fig. 2: Sugar Cane Straw Ash grain after grinding

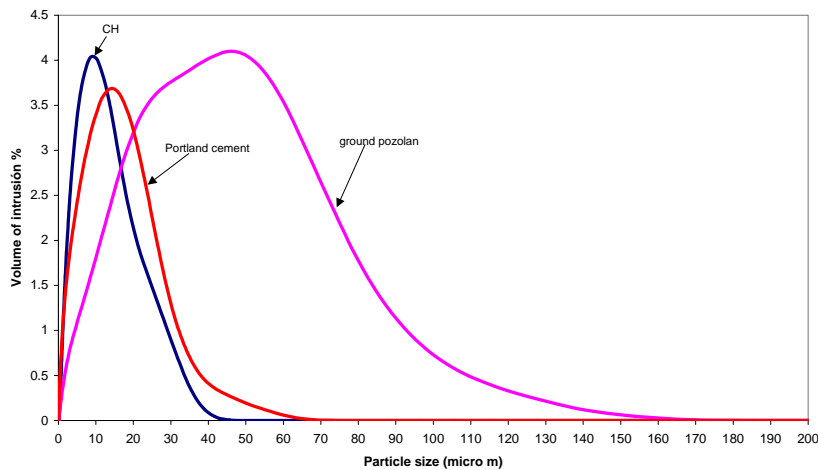


Fig. 3: Particle size distribution for CH, OPC and ground pozzolan

### Concrete with high volumes of lime-pozzolan blend (LPB)

In order to optimize consumption of OPC in concrete, the relationship between compressive strength and cement content and fines in concrete must be thoroughly understood. A denser and stronger matrix cannot be achieved by only increasing the amount of OPC, since this implies an increase of the water demand in the mix -- which implies an increase in the porosity and microcracking in the hardened matrix. Instead, one can use very fine, low water-demand mineral additions that partially replace OPC and combine these with superplasticizers in order to manufacture concrete mixes with a lower water to fines ratio. This should produce a decrease in shrinkage and thermal microcracking, and provide a better hydrating condition for the OPC used. Also, the fine grains of the LPB complement the particle size distribution of the cement by filling the voids between cement grains, and potentially those existing between the smaller grains of aggregate. This principle can be applied to any type of concrete, just as it has been applied to SCC and UHPC with the advantage of achieving excellent mechanical properties and durability with a very low OPC consumption.

To assess how much OPC can be replaced without varying the rheological properties of the mix, the authors propose a method that allows a correction to the traditional mix proportion to incorporate large volumes of LPB. The process has two stages. The first stage is to determine the maximum volume of cementing fines (volume of OPC + LPB, hereafter termed "fines") that a mix can assimilate without changing rheological properties significantly. To do this, a series of experimental trials is performed, where a given mass of OPC is replaced by the same mass of LPB at different replacement levels – for example, starting with 20% OPC replacement and increasing. The water is adjusted for each mix in order to remain within a target slump range. For each mix, strength of test cylinders at early age is measured. The optimum mix is the one with the highest replacement level but in which the water/fines ratio has not increased significantly (less than 20%) and there is an acceptable fall in the early compressive strength (less than 40%). The volume of fines at this point is considered to be the maximum volume of fines that the mix can assimilate for a given consistency.

Once the maximum volume of fines is determined, Stage 2 adjustments are made. The total volume of cementing materials (OPC + LPB) and the amount of water used is kept constant – thus, the volumetric ratio of water/fines is constant – and the optimum volume ratio between OPC and LPB is determined, as proposed by Bornemann and Schmidt (23). Usually, the

proportion of LPB to OPC is increased until the target property, say 28-day strength, and a given consistency starts to show a significant change. The minimum amount of OPC to use is established in this way.

The use of a large volume of LPB results in a change in the aggregate/paste ratio, since the LPB has lower density than OPC. As the proportion of LPB increases, the volume of paste increases and the average spacing between aggregate grains increases. A thin section (Figure 4), clearly shows that in mortars where 40% of OPC is replaced, the aggregate grains separate from each other because of the increase of the volume of paste.

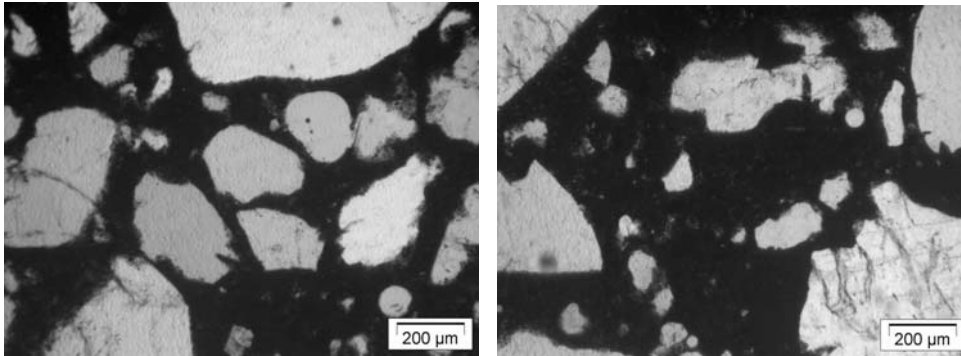


Figure 4: Mortars made with OPC (left) and with 40% lime-pozzolan addition (right)

#### Example of Optimum OPC-LPB Determination

To demonstrate the principles outlined here, a concrete was designed for the Cuban construction industry. The concrete has a 30 MPa target strength at 28 days. A traditional mix design requires 515 kg/m<sup>3</sup> of OPC because of the very poor quality limestone aggregate common in Cuba. The mix was designed for a target slump between 100-150 mm.

Stage 1: from 0 – 80% of the cement was replaced by the same mass of LPB. Table 1 tabulates the components of the mixes prepared and Fig. 5 shows the results of the compressive strength tests. OPC replacement always produces a lower compressive strength than the control at all ages. The mix prepared with 40% LPB (mass) proved to be able to assimilate the maximum volume of fines, while still maintaining an acceptable consistency. Above 40% replacement, 3d and 7 d compressive strength fell significantly in comparison with the control series, and the water to fines ratio increased approximately 20% compared to that of the control series.

Table 1: Components of the mixes prepared in stage 1 (LPB Density = 2170 kg/m<sup>3</sup>)

| Material                  | unit              | Proportion between LPB/OPC (mass) |      |      |      |      |
|---------------------------|-------------------|-----------------------------------|------|------|------|------|
|                           |                   | 0%                                | 20%  | 40%  | 60%  | 80%  |
| cement                    | kg/m <sup>3</sup> | 515                               | 412  | 309  | 206  | 103  |
| LPB                       | kg/m <sup>3</sup> | 0                                 | 103  | 206  | 309  | 412  |
| Volume of "fines"         | L/m <sup>3</sup>  | 166                               | 178  | 193  | 209  | 223  |
| Fine aggregate            | kg/m <sup>3</sup> | 737                               | 737  | 737  | 737  | 737  |
| Coarse aggregate          | kg/m <sup>3</sup> | 737                               | 737  | 737  | 737  | 737  |
| superplasticizer          | L/m <sup>3</sup>  | 6.2                               | 6.2  | 6.2  | 6.2  | 6.2  |
| Water                     | kg/m <sup>3</sup> | 175                               | 183  | 210  | 213  | 275  |
| Water to OPC ratio (mass) | -                 | 0.34                              | 0.44 | 0.68 | 1.03 | 2.67 |
| Water to fines ratio      | -                 | 0.34                              | 0.36 | 0.41 | 0.41 | 0.53 |
| Slump                     | mm                | 156                               | 151  | 128  | 152  | 120  |

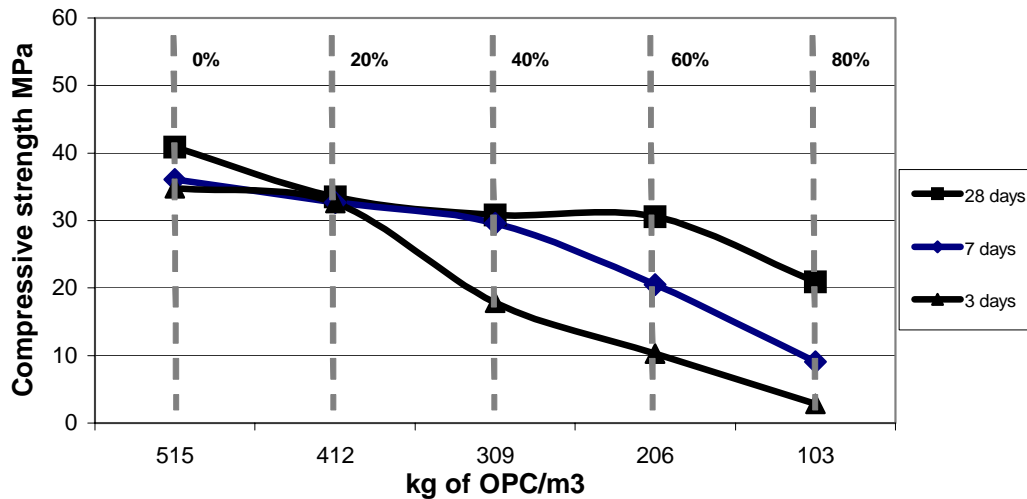


Fig. 5: Results of Stage I Strength Tests

**Stage 2:** the volumetric proportion between OPC and LPB was varied while keeping the total volume of fines and the water content constant (thus keeping the volumetric water to fines ratio constant), in order to assess the limits of OPC replacement. Table 2 presents the components of the mixes and Fig. 6 shows the results of compressive strength. The differences in compressive strength between the control series and that with 60% OPC volume replacement for mixes prepared with the maximum volume of fines are under 15%, while a 20% replacement of OPC with LPB appears to give optimum strength at 7 and 28 days.

The testing confirms that large amounts of OPC in a given mix can be replaced by LPC without serious reduction in the 28 days compressive strength and without significant variations in consistency, provided the volumetric ratio water to fines is kept constant and the mix is prepared with the maximum volume of fines that it can assimilate. The samples tested in stage 2 show improvement in the compressive strength between 15-20% compared with samples prepared in stage 1 having similar mass of OPC per cubic meter.

#### Influence of lime-pozzolan admixture on pore structure

As concrete is a porous material, many aspects of its durability are associated with liquids transport within the pore structure of the concrete. In cold climates the water retained in the capillary pores can expand at temperatures below freezing and destroy the matrix. During chemical attack, pore water becomes a medium for reaction of various expansive compounds.

In the absence of cracking, impermeability is generally related to the volume of the capillary pore fraction. The definition of the boundary between capillary and the smaller pore fraction is a matter of some controversy. Here, we adopt the definition of capillary pores as those between 0.03 and 10 microns. Figure 7 shows the cumulative volume of mercury intruded for one OPC paste at 2 ages, and one 60:40 OPC:LPB paste at the same two ages. These pastes were water cured for 28 days and 60 days.



Table 2: Components of the mixes prepared for stage 2

|                                 | unit              | LPB/OPC proportion (vol.) |      |      |      |      |
|---------------------------------|-------------------|---------------------------|------|------|------|------|
|                                 |                   | 0%                        | 20%  | 40%  | 50%  | 60%  |
| cement                          | kg/m <sup>3</sup> | 626                       | 501  | 376  | 313  | 250  |
| LPB                             | kg/m <sup>3</sup> | 0                         | 81   | 163  | 203  | 244  |
| Total LPB + OPC                 | kg/m <sup>3</sup> | 626                       | 582  | 538  | 516  | 494  |
| Water                           | L/m <sup>3</sup>  | 210                       | 210  | 210  | 210  | 210  |
| Volume of "fines"               | L/m <sup>3</sup>  | 202                       | 199  | 196  | 195  | 193  |
| Fine aggregate                  | kg/m <sup>3</sup> | 737                       | 737  | 737  | 737  | 737  |
| Coarse aggregate                | kg/m <sup>3</sup> | 737                       | 737  | 737  | 737  | 737  |
| superplasticizer                | L/m <sup>3</sup>  | 7.5                       | 7.0  | 6.5  | 6.2  | 5.9  |
| Slump                           | mm                | 160                       | 130  | 115  | 110  | 92   |
| Water to OPC ratio (mass)       | -                 | 0.34                      | 0.42 | 0.56 | 0.67 | 0.84 |
| Water to fines ratio (mass)     | -                 | 0.34                      | 0.36 | 0.39 | 0.41 | 0.42 |
| Volumetric water to fines ratio |                   | 0.40                      | 0.40 | 0.40 | 0.40 | 0.40 |

Kommentar: Corregir valor en función del uso de densidad en lugar de peso unitario

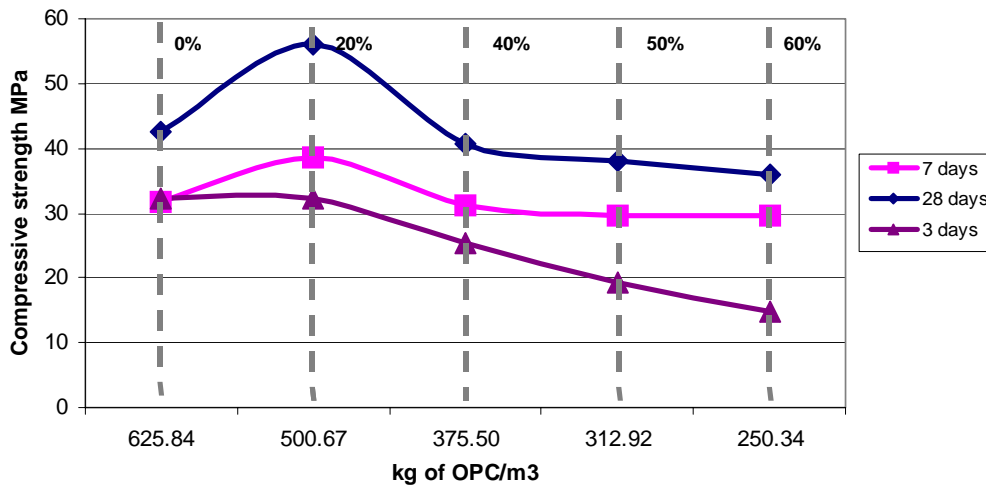


Fig. 6: Results of the experimental test carried out in stage 2

The refinement of pore structure in the OPC:LPB blend with age is a result of OPC hydration and continuing pozzolanic activity – additional confirmation of this is given below. However, Figure 7 clearly shows that the OPC:LPB paste has a significantly larger volume of capillary pores when compared to the OPC paste. One might expect a concrete made from the OPC:LPB blends to have a higher permeability than its OPC equivalent. This is not certain, however, since the effect on mass transport properties of optimizing the mix design – as described above – has yet to be determined.

The larger capillary porosity of pozzolan blended pastes that refines more quickly with time than OPC pastes is a common phenomenon for many types of pozzolans. In all cases additional testing of concretes made with these pastes under a wide range of exposure conditions is necessary.

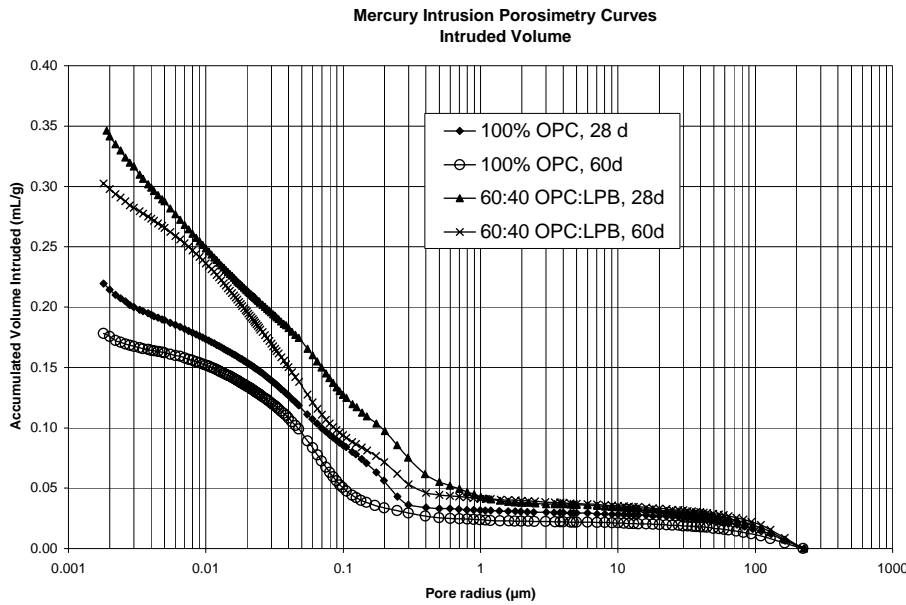


Figure 7: Pore-size distribution determined using mercury porosimetry. All pastes water cured for the time shown.

Fig. 8 shows the XRD traces of pastes made with different OPC/LPB blends. It can be observed that the higher the amount of LPB admixture, the lower the intensity of the CH peak, which suggests a considerable amount of pozzolanic activity in the OPC/LPB blends during the first 28 days of hydration.

The pozzolanic activity suggested by the XRD traces was confirmed by performing TGA on an OPC paste, and on 80:20 and 60:40 OPC/LPB pastes. Pastes were manufactured and then cured in sealed glass vials for 28 days, at which point TGA analysis was performed. The weight-loss vs. temperature curve allows one to determine the water lost from CH dehydroxylation during heating in the range from 470 to 550 °C. Since the material proportions are known, one can calculate the theoretical CH content of the pastes assuming no pozzolanic activity. This value can then be compared to the estimated CH content from the measured weight loss. Thus, the following table was calculated:

| OPC (g) | LPB (g) | Theoretical mass of CH present | Calculated mass of CH present, determined from measured water loss | CH consumed | CH consumed-per g pozzolan |
|---------|---------|--------------------------------|--|-------------|----------------------------|
| 100     | 0       | 8.34                           | 8.34   | 0           | 0                          |
| 80      | 20      | 10.67                          | 8.75   | 1.92        | 0.12                       |
| 60      | 40      | 13.00                          | 5.34   | 7.66        | 0.24                       |

The TG analysis confirms the presence of pozzolanic activity. It also indicates that pozzolanic activity is higher when higher proportions are used – the 60:40 OPC:LPB blend consumes 0.24 g of CH per g of pozzolan, which the value is 0.12 for the paste containing only 20% LPB.

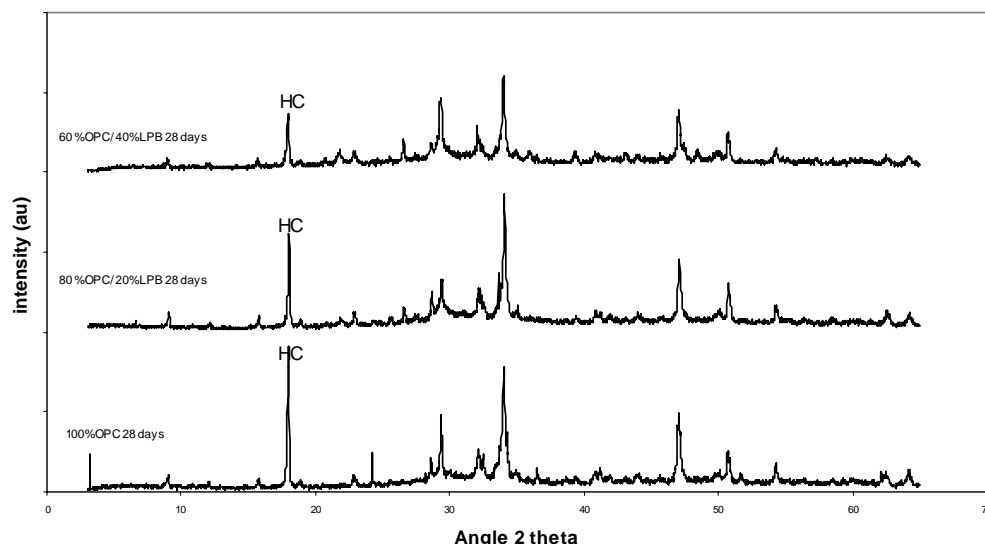


Figure 8. X-ray diffraction traces of hydrated cement paste and OPC:LPB pastes.

## Conclusions

The use of lime-pozzolan binder (LPB) to reduce the amount of cement used in high and ultra-high performance concrete is an attractive proposition from an ecological viewpoint. Tests reported here have concentrated on regular-strength concrete, but the principles are expected to also apply to higher strength mixes. Results suggest that the quantity of cement can be reduced while maintaining equivalent strength by using a systematic procedure that optimizes the OPC/LPB ratio while maintaining a constant water/fines ratio and constant consistency.

From the standpoint of durability, the use of high levels of free-lime in a fresh concrete mix is a concern. Preliminary tests on pore structure and mineralogy indicate that this concern may not be founded, since chemical reaction between the pozzolan and lime reduces the amount of lime and also refines the pore structure of the concrete. However, for full-scale implementation of the proposed technology, additional engineering testing to establish mass transport properties and volume stability of OPC/LPB concretes under a wide range of exposure conditions is required.

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