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EFFECT OF LIMESTONE AGGREGATE ON THE PROPERTIES OF NATURAL HYDRAULIC LIME MORTAR (NHL 5).
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KEYWORDS: Portland limestone aggregate; NHL 5; mechanical and hygric properties

ABSTRACT

Although the properties of mortars depend mainly on the binder nature, the aggregate plays a role. There are contradictory reports on the effect of limestone aggregate on lime mortars, probably due to the use of different lime binders and the wide range of limestones used to manufacture aggregate. This paper studies the influence of Portland limestone aggregate on NHL5 mortars and compares them with siliceous aggregate mixes.
The aggregate’s mineral composition (calcitic vs siliceous) does not affect the properties of NHL5 mortars to a great extent. The results suggest that the limestone aggregate mortars have a greater number of larger, active open pores, while the silica sand mortars include a greater number of fine, suction-active pores probably associated with the NHL5 binder. These would agree with the greater porosity and suction of the limestone aggregates, displaying porous surfaces and the smooth, impermeable surfaces of brittle fracture in the silica sand determined with SEM.
At low aggregate contents, the silica sand mortars are stronger in compression and flexion than the limestone sand mortars however, at greater aggregate contents, the limestone sand mortars are stronger. Some authors found a strength increase in lime mortar using limestone aggregate however, results are not comparable due to the use of different lime binders, limestone aggregate types and water/binder ratios. Rather, as the NHL5 includes significant clinkers, the results here are linked to PC research. As a result, the fine lime aggregate powder probably provides a favourable surface for the nucleation and early growth of calcium silicate hydrate, enhancing hydration and strength development in the NHL5 paste. In addition, the coarser limestone aggregate, may enhance strength, by reaching a higher bond at the interfacial transition zone, due to the auspicious physical (texture), reactive nature and high suction of the limestone surfaces.
Both the siliceous and limestone aggregate mortars follow similar trends when increasing aggregate content: As the aggregate content increases, the compressive and flexural strengths drop however, porosity, water absorption and capillary suction increase. These effects are probably on account of the multiplication of interfaces or transition zones which are discontinuities in the structure of the composites.

INTRODUCTION

This is part of a wider research that aims at designing a compatible and durable mortar to repair Portland stone, in an effort to apply compatible and durable repairs to Portland stone in a number of monuments in Dublin. The Portland stone is a limestone quarried in the Island of Portland, Dorset, UK. The rock was used in many historic buildings throughout Ireland and the UK which are now weathered and need repair.

Although the properties and characteristic of the mortars depend mainly on the nature of the binder, the aggregate quality plays an important role. This fact has been known since early civilizations. The grading and shape of the aggregate can affect workability, bulk density and mechanical strength of
mortars while the mineralogical composition and reactivity can induce chemical reactions that may lead to changes in hygienic properties and inter-particle and transition (aggregate–binder interface) zones increasing bond and strength. Pavía and Toomey [1] include a comprehensive review (ranging from Roman to current accounts) on the effect of the type aggregate in lime mortars. According to these authors, it seems generally accepted that the strongest mixes are produced from well graded, clean and sharp aggregate; and that increasing aggregate angularity and fineness enhances compressive strength while rounded aggregates increase porosity reducing strength. It also seems generally accepted that grain size distribution influences the porosity and pore size distribution and thus permeability and strength however, there are discrepancies on whether limestone aggregate produces stronger mortars than siliceous aggregate and whether a coarser or a finer average particle size would enhance mortar properties.

There are contradictions on whether calcitic (limestone) aggregate produces stronger mortars than siliceous aggregate. Vicat [2] concluded that calcitic aggregates produced slightly stronger mortars than non-calcitic aggregates. Lanas and Alvarez [3], using hydrated lime, report strength increase with limestone aggregate. They attribute this to homogeneity between aggregate and binder and the possibility of a syntaxial growth during portlandite carbonation favoured by the calcite in the aggregate providing nucleating sites for crystal growth. However, Holmes and Windgate [4] disagree, claiming that limestone aggregate produces weaker lime mortars. Pavía and Toomey [1] found that NHL2-limestone aggregate mortars were slightly weaker than those produced with sandstone and quartz aggregate.

The studies above used different types of lime (hydrated and hydraulic) and different types of limestone aggregate. Therefore, some of the discrepancies on whether or not limestone aggregate produces stronger mortars are probably due to the use of different limes and ‘parent’ limestones to produce the aggregate. A wide range of limestones are used for manufacturing aggregate in construction. Most limestone aggregates are manufactured from natural limestones which, although mostly uniform in composition (they are carbonate rocks consisting mainly of CaCO₃), greatly vary in porosity, suction and strength: from the nonporous, dense rocks as strong as some granites to much weaker, permeable limestones. This wide property variation can impact mortar in different ways. Former studies on the effect of limestone aggregate on lime mortars do not discuss how the reactivity or the suction ability of the limestone aggregate can affect the physical properties of the mortars.

This paper studies the influence of Portland limestone aggregate on the flexural and compressive strength, porosity, water absorption, water demand and capillary suction of natural hydraulic lime (NHL 5) mortars and compares the results with mortars made with a pure silica sand (mostly quartz). It considers the properties of the limestone used to manufacture the aggregate. The limestone aggregate is manufactured from an oolitic rock including abundant microcrystalline calcite (micrite) and significant active pores while the siliceous aggregate consists mainly of inert crystalline quartz of low porosity. The NHL5 binder contains some free lime (≥3% by weight) and a significant amount of clinkers (calcium silicates) similar to those found in Portland cement (mainly belite C₃S).

The role of limestone aggregate and limestone fillers has been investigated in depth in Portland cement (PC) technology. As the NHL5 contains significant clinkers identical to those in PC, the role of limestone aggregate (both filler-graded and coarser) in PC concrete is reviewed below. Soroka and Setter [5] showed that limestone fillers in concrete can act as nuclei for precipitation of CSH enhancing clinker hydration and early strength development. Many authors have later agreed to this, observing an acceleration of cement clinker hydration using limestone fillers. Currently, in a multi-scale investigation of the performance of limestone in concrete, Bentz et al. [6] re-state the above, claiming that fine limestone powder in the form of calcite provides a favourable surface for the nucelation and growth of calcium silicate hydrate gel at early ages, enhancing hydration. With respect to coarser limestone aggregate, the authors state that limestone-aggregate based, PC concrete often reach higher compressive strengths at equivalent levels of hydration, as a result of a higher bond at the interfacial transition zone, likely due to the auspicious physical (texture) and chemical nature of the limestone surfaces.
1. MATERIALS AND METHODS

1.1 Materials

Natural hydraulic lime NHL 5 complying with EN 459-1 [7] was used as a binder. The NHL5 binder contains some free lime (≥3% by weight) and a significant amount of clinkers (calcium silicates) similar to those found in Portland cement (mainly belite C$_2$S). According to the producer (Saint Astier) [8], the NHL5’s principal components are calcium dihydroxide (hydrated lime = Ca(OH)$_2$) in concentration 15–65% (m/m) – 30% (m/m); calcium silicate (C$_2$S/C$_3$S) in concentration 10–45% (m/m) – 30% (m/m) and calcium carbonate (Ca CO$_3$) in concentration 10–40% (m/m) – 25% (m/m); with no impurities relevant to classification and labelling.

The limestone aggregate (Figure 1) is manufactured from Portland Witbed limestone, an oolitic limestone from the Jurassic period. According to Dubelaar et al. [9], macropores ranging from 10-100 µm dominate the pore system of this limestone and approximately 20% range between 0.01 and 0.2 µm therefore under the micropore range (<1µm). Further physical properties of the Witbed Portland limestone were analysed in the laboratory and are included in table 1. The standards used to measure these properties can be found in section methods below. This aggregate was supplied by Suttle quarries, it was sieved in the laboratory and the fraction under 5 mm used.

Table 1. Physical properties of Portland Witbed used to manufacture the aggregate – in brackets coefficient of variation (COV in %).

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
<th>COV (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Compressive strength</td>
<td>54.23</td>
<td>2.1</td>
</tr>
<tr>
<td>Flexural strength</td>
<td>6.23</td>
<td>1.12</td>
</tr>
<tr>
<td>Bulk density</td>
<td>2159.15</td>
<td>0.6</td>
</tr>
<tr>
<td>Porosity %</td>
<td>15.4</td>
<td>0.6</td>
</tr>
<tr>
<td>Water absorption (%)</td>
<td>7.19</td>
<td>0.79</td>
</tr>
<tr>
<td>Capillary Suction</td>
<td>0.638</td>
<td>2.54</td>
</tr>
</tbody>
</table>

The siliceous aggregate (Fig. 1) is a pure quartz sand supplied by St. Astier/CESA in France, with grading similar to that of the CEN normal sand. The bulk density and porosity could not be measured however, it consists of monocrystalline and occasionally polycrystalline quartz (SiO$_2$) – of high bulk density and virtually no porosity.

Table 2 includes the chemical composition, petrography, mineralogy and morphology of the aggregates determined by X-Ray Fluorescence, Energy Dispersive X-Ray analysis, petrographic microscopy and Scanning Electron Microscopy respectively. As it can be seen from Table 2, both aggregates are quite pure in composition (calcium carbonate vs silica). Their pore system, hygric properties and reactivity greatly differ: while the silica sand is made up of monocrystalline and occasionally polycrystalline quartz (SiO$_2$) - a non-porous, highly-crystalline phase of low reactivity (Table 2)-; the Portland limestone aggregate comprises abundant microcrystalline calcite (micrite), a high specific surface phase of greater reactivity.

The limestone aggregate consist of microcrystalline calcite ooids linked to each other with a significant amount of inter-ooid space, sometimes filled with larger crystals of calcite (sparite cement). This grain supported fabric typically leads to significant open porosity and permeability. Therefore, the limestone
sand is more porous and permeable and more reactive than the highly crystalline quartz in the siliceous sand. The particle size distribution for both types are presented in Fig. 3. As it can be seen from this figure, the limestone sand is coarser and has a wide range of particle sizes varying from filler grade (0-100µm) up to 5 mm.

Table 2: Aggregate properties. (*) SEM micrograph of a similar, finer grained, manufactured limestone.

<table>
<thead>
<tr>
<th>Aggregate type</th>
<th>Chemical composition %</th>
<th>Petrography and Mineralogy</th>
<th>Morphology</th>
</tr>
</thead>
<tbody>
<tr>
<td>Portland limestone</td>
<td>- calcium carbonate 95.8</td>
<td>calcite (micrite and sparite) traces of quartz</td>
<td>angular, low/md sphericity</td>
</tr>
<tr>
<td></td>
<td>- magnesium carbonate 1.2</td>
<td></td>
<td>porous surfaces evenly rough (*)</td>
</tr>
<tr>
<td></td>
<td>- aluminium oxides 0.3</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>- iron oxides 0.3</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>- silica 1.3</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>- water (and other) 1.4</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Siliceous</td>
<td>- silica / trazes of Al</td>
<td>monocristalline &amp; polycristalline</td>
<td>Angular, md sphericity, smooth surfaces with brittle fracture</td>
</tr>
<tr>
<td></td>
<td>- quartz occasional feldspar</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Fig. 3: Accumulative curves of aggregate particle size.

The binder/aggregate ratios (B/A) are 1:1, 1:2 and 1:3 by volume. Table 3 shows the mix proportions and water demand to achieve a flow diameter of 170±5 mm for each mortar. As it can be seen from this table, the limestone sand mortars show a higher water demand to achieve a given consistency at low aggregate contents however, as the aggregate content increases the difference is not significant.

Table 3: Mix proportions and water content of the mortars studied.

<table>
<thead>
<tr>
<th>Mix</th>
<th>Aggregate type</th>
<th>Binder/Agg.</th>
<th>Water/binder</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Portland limestone</td>
<td>1:1</td>
<td>0.72</td>
</tr>
<tr>
<td>2</td>
<td>Portland limestone</td>
<td>1:2</td>
<td>0.89</td>
</tr>
</tbody>
</table>
1.2 Sample preparation and curing

The mortars were mixed with the amount of water required to achieve a flow diameter of 170±5 mm (measured with the flow table in accordance with EN 459-2 [11]). Lime and aggregate were dry mixed for 2 min. Water was then added and mixed for 2 min at low speed, and finally for 1 min at high speed. The mortars were moulded, compacted on a vibration table for 5 seconds, according to EN 459-2, and removed from the moulds after 3 days. They were initially covered with damp towels to prevent shrinkage cracking and cured for 25 days at approximately 90% humidity and 20 ± 2°C temperature. Each property reported is the arithmetic mean of three samples.

1.3 Methods

The mechanical properties of the mortars were characterized by measuring the compressive strength (Fc) and the flexural strength (Ff) according to EN 1015-11[12]. The flexural test was performed on the mortar samples (40x40x160 mm) using Zwick testing machine at rates of loading (1 mm/min). The compression strength test was carried out on the half prims using a loading rate of 1 mm/min.

The porosity and bulk density was tested according to RILEM recommendations [13]. The water absorption was measured according to UNE [14]. Capillary suction was measured according to EN 1925 [15]. In this test, the dry samples were immersed to a depth of 3 ± 1 mm. They were removed and weighed at specific time intervals of 1, 3, 5, 15, 30 and 60 min. Then, the coefficient of water absorption by capillarity (CS) was calculated according to the formula in [15].

2. RESULTS AND DISCUSSION

2.1 Hygric properties

The hygric properties of mortars are of great importance as they significantly impact their performance in relation to water, frost, salt and chemical weathering which determine durability.

According to the results in Fig 4, the open porosity of the limestone aggregate mortars (with values between 14.74 and 19.75%) is slightly higher than that of the silica sand mixes (11.06-18.43%). This can be partially attributed to the greater open porosity of the limestone aggregate. In addition, the greater difference in porosity at low aggregate contents (1:1) (14.74 vs 11.06% for limestone and silica sand mortars respectively) may be due to the higher water demand of the limestone mortar at low aggregate content (Table 3). It is well known in mortar and concrete technology that some binder pores with active moisture transport are remnants of space once filled with water therefore, increasing water/binder ratio increases porosity. Papayianni and Stefanidou [16] stated that, in lime mortars, water/binder ratio is the most important factor influencing porosity.
As expected, the water absorption shows a similar trend (Fig. 5), where the limestone mortars possess the highest water absorption with values between 6.5 and 10.42%.

On the contrary, capillary suction shows the opposite trend (Fig. 6): the silica mortars have a greater suction than the limestone sand mortars however, the values are close.
The Portland stone aggregate produced the mortars with slightly higher bulk density at all B/A ratios (Fig 7) however, the influence of limestone vs siliceous aggregate on the mortar’s bulk density is not significant. Low coefficients of variation (0.18-4.04) indicate that the results are consistent.

![Bulk density of NHL5 mortars at 28 days.](image)

### 2.2 Mechanical properties

The compressive and flexural strength of the mortars are shown in Fig. 8 and 9 respectively. As expected, strength lowers with increasing aggregate content, this is probably due to the multiplication of interfaces or transition zones which are discontinuities in the structure of the composite.

At 1:1 (B/A), the siliceous sand mortar is stronger than the limestone one (despite the higher water demand of the latter), with substantial difference after 90 days of curing. However, at higher aggregate contents (1:2 and 1:3 B/A ratios) the compressive strength of the limestone aggregate mortars is higher increasing by about 9 and 18 % (at 28 days) when compared to the siliceous sand.

![Compressive strength of lime mortars at 28 days.](image)

The flexural strength behaves in a similar manner (Fig. 9), at a 1:1 ratio, the siliceous sand mortars are stronger but as the amount of aggregate increases (1:2 and 1:3) the limestone aggregate produces mortars of higher flexural strength (36% and 12% strength increase for 1:2 and 1:3 ratios respectively).

Therefore, generally, the limestone aggregate at 1:2 and 1:3 B/A ratios increased mortar strength. Other authors [20, 21, 22] have found a strength increase in lime mortar using limestone aggregate however, these results are not comparable due to the use of different lime binders and limestone aggregate types and water/binder ratios. For example, using hydrated limes, the greater strength of limestone mortars has been attributed by Lanas and Alvarez [3] to homogeneity between aggregate and binder and the
possibility of a syntaxial growth during portlandite carbonation favored by the calcite aggregate providing nucleating sites for crystal growth. However, the hydraulic lime binder in this study is substantially different to hydrated lime.

The NHL5 used in this study contains clinkers identical to those in PC and furthermore, approximately 4% of the limestone aggregate is sized under 300μm. This fine limestone powder (in the form of calcite) can provide a favourable surface for the nucleation and growth of calcium silicate hydrate gel at early ages, enhancing hydration and strength development in the NHL5 paste. A similar mechanism was reported by Soroka and Setter [5] and many other authors after them in PC concrete.

The coarser limestone aggregate, may also have enhanced strength, as a result of reaching a higher bond at the interfacial transition zone, due to the auspicious physical (texture), reactive nature and high suction of the limestone surfaces.

![Flexural strength of lime mortars at 28 days. COVs 1.3-7.6 for both mortars.](image)

**CONCLUSIONS**

The aggregate’s mineral composition (calcitic vs siliceous) does not seem to affect the properties of NHL5 mortars to a great extent.

The limestone aggregate mortars reached greater porosities at all aggregate contents than the silica mixes but, as the aggregate content increases, the difference becomes less significant. The water absorption shows a similar trend while the capillary suction shows an opposite trend: the silica mortars have a greater suction than the limestone sand mortars however, the values are close.

This suggests that the limestone aggregate mortars have a greater number of larger, active open pores while the silica sand mortars include a greater number of fine, suction-active pores probably associated with the NHL5 binder. These would agree with the greater porosity and suction of the limestone aggregates (15.4% porosity; 7.19% water absorption and 0.64 kg/m² min⁻⁰.⁵ capillary suction) displaying porous surfaces and the smooth, impermeable surfaces of brittle fracture in the silica sand determined with SEM.

At low aggregate contents, the silica sand mortars are stronger (in compression and flexion) than the limestone sand mortars however, at greater aggregate contents, the limestone sand mortars are stronger. Some authors have found a strength increase in lime mortar using limestone aggregate however, results are not comparable due to the use of different lime binders, limestone aggregate types and water/binder ratios. Instead, here, as in PC concrete, the fine limestone powder (calcite) probably provides a favourable surface for the nucleation and growth of calcium silicate hydrate gel at early ages, enhancing hydration and strength development in the NHL5 paste. In addition, the coarser limestone aggregate, is probably enhancing strength, as a result of reaching a higher bond at the interfacial transition zone, due to the auspicious physical (texture), reactive nature and high suction of the limestone surfaces.

Both the siliceous and limestone aggregate mortars follow similar trends in relation to increasing aggregate content: As the amount of aggregate increases, the compressive and flexural strength drop however, the open porosity, water absorption and capillary suction increase. These effects are probably
on account of the multiplication of interfaces or transition zones which are discontinuities in the structure of the composite.

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