Mechanical and hygric properties of natural hydraulic lime (NHL) mortars with additives

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ABSTRACT: Currently, hydraulic limes are used as binders to strengthen and repair traditional and historic masonry. For hundreds of years, additives such as pozzolans and stone dust have been used in combination with lime binders to improve the properties and durability of masonry mortars and concrete. This paper studies flexural and compressive strength, porosity, water absorption and capillary suction of natural hydraulic lime (NHL5) mortars prepared with two additions (ground granulated blastfurnace slag –GGBS- and rice husk ash-RHA). Portland limestone dust was used as a filler. The mixing, curing, binder/aggregate ratio and flow were kept constant in order to attribute variation of mortar properties to the type of additive. Two additive/lime percentages were used (10% and 20%). The replacement of NHL5 with GGBS, RHA and stone dust enhances compressive and flexural strength of NHL5 mortars at 28 days. Replacement of NHL 5 by 20%GGBS increases NHL5 mortar compressive strength by 125% and either doubles or increases by 50% its flexural strength. Replacing 10-20% NHL5 with RHA enhances compressive strength of NHL5 mortars to a smaller extent (by 26 -47%). The strength increase of the NHL5-additive mortars is due to the presence of additional cementitious hydration products, and the filler effect of the additives increasing packing density. GGBS, RHA and stone dust lower capillary suction of NHL5 mortars. Replacing NHL5 with GGBS significantly lowers the porosity of NHL5 mortars (by 46-33%); their capillary suction (49-64% drop) and water absorption (c.49% drop for the 20%GGBS). Replacing NHL5 with RHA does not lower porosity or water absorption but reduces capillary suction (41% decrease for the 20%RHA mix). The higher porosity/water absorption of the NHL5/RHA mortars are probably due to the much greater specific surface and higher water demand of the RHA increasing the amount of pores.

1 INTRODUCTION

Natural hydraulic limes (NHLs) have been used as a binder for building since antiquity. Nowadays, they are used for new building and repairs to existing masonry; often chosen as compatible materials to repair traditional and historic masonry. They are sometimes preferred to air lime because their hydraulic set leads to an earlier strength development, a lower shrinkage and greater durability (Grilo et al. 2014 b). In the past, NHLs were locally produced and used wherever impure limestone was available. Their use decreased with the development of cements in the early 19th century, however, NHLs are regaining popularity as a sustainable alternative to Portland cement (PC). Their environmental credentials are based on their lower production energy consumption and the reabsorption of the CO₂ emitted during burning (Grist et al. 2013).

Pozzolans are materials with an amorphous siliceous or siliceous and aluminous content that react with calcium hydroxide in the presence of water to form cementitious hydrates. In many ancient civilizations, pozzolans were used to enhance the properties of lime mortars and concrete and many structures still remain as a testament to the durability of lime–pozzolan mortars and concrete. Today, pozzolans are often used to enhance the properties and increase
the durability of PC concrete and other PC composites. Most pozzolans commercially used today are often industrial or agricultural byproducts, therefore, as wastes, their use in construction drops energy consumption and CO₂ emissions and is a better alternative to landfill disposal.

Evidence of the use of pozzolans has been found in the Neolithic period (7000 BC) in Galilee (Caijun 2001 referring to Malinowski et al.), the Minoan civilization (2700 to 1450 BC) (Carr 1995) and Ancient Greece (1500 BC) (Moropoulou et al. 2004 referring to Jiang and Roy). The Roman Empire is however most associated with the use of pozzolans. According to historic records and current research (Hicky Morgan (1914) Hooper and Ash (1939) Davey (1961) Plommer (1973) Boynton (1980) Pavia and Caro (2008)) the Romans often used both natural (volcanic ash) and artificial pozzolans (brick and tile dust) to render pure lime hydraulic.

According to the European lime standard EN 459-1 (Schiffner 2011) there are three strength classes of natural hydraulic lime: NHL2, NHL3.5 and NHL5; where the numbers refer to the minimum compressive strength (MPa) at 28 days. According to the standard, when pozzolanic or hydraulic materials such as PC are incorporated (up to 20%) the letter Z follows thelime designation e.g. NHL5-Z. NHL is obtained by burning limestones with a high content of clay (6.5–20%) below the sintering temperature (1250°C). This results in the formation of mainly (dicalcium silicate-belite-C₂S) clinkers that hydrate and raise strength slower than some PC clinkers and a significant amount of available lime (≥15% by mass for NHL5). Tetracalcium aluminoferrite (C₄AF), C₃S, and tricalcium aluminate (C₃A) can also form in small amounts, due to a local overheating in the limekiln (Silva et al. 2014). As a result, NHL mortars harden through a dual mechanism (a more or less fast hydration and slow carbonation).

In the last decades, practical research in historic building conservation has linked the use of NHL mortars to an ease of application, compatibility with substrates and durability. For example, Maravelaki-Kalaitzaki et al. (2005) used NHL3.5-Z lime mortar to restore historic bioclastic limestone masonry in Chania Crete, Greece. The authors recorded no failure, cracks or release of soluble salts after three years. Proprietary hydraulic lime mortars were used in the restoration of the renaissance façade of the Courts Office in Bruges (Naeyer 2000). These were reported to retain workability being easy to apply and remaining plastic for longer periods so that masons could rework and reset their repairs. In the restoration of the Cathedral at Kirkjubøur in the Faroe Islands (Larsen et al. 2007) different NHL mortars were tested to determine resistance against weathering. The authors concluded that only NHL5-Z and NHL3.5 would be durable in the exposed environment of the islands. Allanbrook and Normandin (2007) used NHL mortars to repair the limestone of the Fifth Avenue facade of the Metropolitan Museum of Art in New York City. They reported that the mortars were inherently compatible with limestone because of their composition and found that they can reach the strength typically achieved by type N mortar (an ASTM C270-07 mix widely used by craftsmen and specified by architects after 1931, consisting of hydrated lime, Portland cement and sand).

Laboratory research has also reported NHL mortars as highly workable materials of low shrinkage, that can develop a good bond with masonry units and display high deformability and water vapor permeability and are resistant to salt and frost damage (Lanas a et al. (2004), Hanley and Pavia (2008), Ball et al. (2011) and Grilo et al. (2014 a)). Other authors have improved the mechanical properties of NHL mortars using fibers: Chan and Bindiganavile (2010) used polypropylene micro-fibers in NHL2 mortars to impart post-peak stress carrying capacity in compression, flexure and shear; and increase their flexural toughness. However, despite a considerable amount of literature devoted to the cement with pozzolanic or latent hydraulic
additives, there is a paucity on research publications on NHL mortars with latent hydraulic or pozzolanic materials. Former authors have investigated their mechanical strength Grist et al. (2013) and Grilo et al. (2014 b). This paper contributes to understanding the properties of NHL mortars with additives, partially replacing the NHL binder, by measuring the mechanical and hygric properties of NHL5 mortars with Ground Granulated Blastfurnace Slag (GGBS), rice husk ash (RHA) and Portland stone dust (PSD) in an effort to improve the characteristics and performance of NHL mortars.

Latent hydraulic materials such as GGBS and pozzolanic materials such as RHA have been successfully blended with PC for decades to enhance properties and durability of concrete. Their reactivity and impact on the properties of hydrated lime pastes have also been investigated. Walker and Pavía (2010, 2011) demonstrated that, out of 9 additives, GGBS and RHA were amongst the most reactive due to their high amorphousness; and that GGBS (and metakaolin) produced the highest strength followed by the high-silica pozzolans RHA and micro-silica (with a strength 68% lower). More details of mineral composition, particle size and specific surface area of GGBS and RHA can be found in Walker and Pavía (2010 and 2011). The PSD consists mainly of calcite (CaCO₃), with traces of silica in the form of quartz (SiO₂) (Godden 2012). A siliceous sand similar in grading and composition to the European CEN standard sand was used as aggregate. The composition and water demand of the mortars studied are shown in Table 1. The mortars were mixed to a constant initial flow of 165±5 mm (measured with a flow table in accordance with EN 459-2), with a binder:aggregate ratio of 1:3 by weight, in order to attribute variation of mortar properties to the type of additive. Two additive-binder replacements (10 and 20%-by weight) were investigated.

<table>
<thead>
<tr>
<th>Mortar designation</th>
<th>NHL5 (%)</th>
<th>GGBS (%)</th>
<th>RHA (%)</th>
<th>PSD (%)</th>
<th>W/B</th>
</tr>
</thead>
<tbody>
<tr>
<td>100% NHL5</td>
<td>100</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0.65</td>
</tr>
<tr>
<td>10% GGBS</td>
<td>90</td>
<td>10</td>
<td>0</td>
<td>0</td>
<td>0.65</td>
</tr>
<tr>
<td>20% GGBS</td>
<td>80</td>
<td>20</td>
<td>0</td>
<td>0</td>
<td>0.66</td>
</tr>
<tr>
<td>10% RHA</td>
<td>90</td>
<td>0</td>
<td>10</td>
<td>0</td>
<td>0.73</td>
</tr>
<tr>
<td>20% RHA</td>
<td>80</td>
<td>0</td>
<td>20</td>
<td>0</td>
<td>0.75</td>
</tr>
<tr>
<td>10% PSD</td>
<td>90</td>
<td>0</td>
<td>0</td>
<td>10</td>
<td>0.60</td>
</tr>
<tr>
<td>20% PSD</td>
<td>80</td>
<td>0</td>
<td>0</td>
<td>20</td>
<td>0.6</td>
</tr>
</tbody>
</table>

2 MATERIALS AND METHODS

2.1 Sample preparation and curing

The lime, additives and aggregate were dry mixed for 2 min. Water was then added and mixed for 2 min at low speed and finally at high speed for 1 min. As aforementioned, water was added to achieve a 165±5 mm flow diameter. The mortars were molded and compacted on a vibration table according to EN459-2. They were initially covered with damp hessian to prevent shrinkage cracking, de-molded after 3 days and cured for 25 days at c. 90% humidity and 20 ± 2°C temperature. Each property measured is the arithmetic mean of three specimens.
2.2 Mechanical strength and hygric properties

The compressive ($F_c$) and flexural strength ($F_f$) were measured according to EN 1015-11 (1999). The flexural test was performed on 40x40x160 mm mortar prisms using a Zwick testing machine at rates of loading of 1 mm/min. Compression strength tests were carried out on the half prisms using a loading rate of 1 mm/min. The porosity and bulk density of the mortars was tested according to RILEM recommendations (RILEM 1980). The water absorption was measured according to UNE (Jung and Lamb 2004). Capillary suction was measured according to EN 1925 (1999). Here, the dry samples were immersed to a depth of 3 ± 1 mm; removed and weighed at specific time intervals of 1, 3, 5, 15, 30 and 60 minutes. The coefficient of water absorption by capillarity was then calculated.

3 RESULTS AND DISCUSSION

3.1 Mechanical properties

The compressive and flexural strength of the mortars are shown in Table 2. The low coefficient of variation (COV) indicates consistent results. As it can be seen here, replacement of NHL5 by GGBS, RHA and PSD enhances the compressive and flexural strength of NHL5 mortars (at 28 days); agreeing with results by Grist (2013) and Işıkdağ and Topçu (2013).

In NHL-pozzolan mortars, there are several simultaneous processes that contribute to setting and strength development. The hydration of the clinkers comprising the NHL5 binder begins early. In contrast, pozzolanic reaction is often reported in the literature as beginning late and progressing slowly. However, if the particle size of the pozzolan is very fine such as RHA, pozzolanic hydrates were recorded after 24 hours, increasing in size and amount at 3 and 7 days and linking to each other, forming continuous networks throughout the paste after 14 days (Pavía et al. 2014). Therefore, hydration of the NHL5 clinkers probably occurs simultaneously to the pozzolanic reaction. In the pozzolanic reaction, RHA reacts with the free lime ($\text{Ca(OH)}_2$-portlandite) comprising the NHL5 to form pozzolanic cements. As aforementioned, the NHL5 binder contributes a significant amount of free lime to the system: ≥15% by mass. The hydration of the NHL5 clinkers progresses more or less fast forming early cementing hydrates and releasing additional portlandite which subsequently reacts with the RHA pozzolans in the presence of water to form additional pozzolanic cements.

The 20%GGBS mortar achieved the highest compressive strength, with an approximately 125% increase when compared to the control mortar. This was expected as GGBS is actually a latent hydraulic binder rather than a pozzolan. The hydration reaction involves activation of the GGBS by alkalis to form its own hydration products. Some of these combine with the lime hydration products to form further hydrates which have a pore blocking effect. This results in a lime mortar with more of very small gel pores, and fewer of the much larger capillary pores for the same total pore volume, that greatly improves the strength and durability of the mortar (Siddique and Bennacer 2012).

Replacing NHL5 by RHA also enhances the compressive strength of NHL5 mortars however to a smaller extent: by approximately 26% (at 10% replacement) and 47% (at 20% replacement).

The results are consistent with former authors reporting that GGBS produced lime pastes with the highest strength, followed by high-silica pozzolans (RHA) with a 68% reduction (Walker and Pavía 2010, 2011). The results also agree with Pavía et al. 2014 who reported that rising
RHA content increased compressive and flexural strength of hydrated lime (CL90s) mortars. The authors reported CL90s:RHA (1:3) mortars as being over 37 times stronger in compression and nearly 5 times stronger in flexion than hydrated lime mixes.

A 10% increase in NHL5 replacement by GGBS approximately doubles the compressive strength of the mortars (from 10 to 21.5 MPa) whereas a 10% increase in RHA raises compressive strength slightly (by about 20%). However, the flexural strength of the NHL5/GGBS mortars is higher at lower replacement level (it drops from 2.03 to 1.59 MPa as replacement increases by 10%). This seems contradictory and can be due to testing inconsistencies.

With respect to the mortars including Portland stone dust (PSD), it was noted that strength increase is higher at low-level replacements: the 10% PSD mortar increased compressive and flexural strength by 37 and 38% respectively, while the 20% PSD mix increased by 24% and 0% when compared to the control mortar. These findings agree with Menéndez et al. (2003) and (Courard and Michel 2014) who concluded that the compressive strength of cement mortar decreases by increasing the ratio of limestone filler. Menéndez et al. (2003) found that cement replacement with limestone powder by 20% slightly reduces the strength (by 4% at 28 days) when compared to cement mortar with 10% limestone powder. According to previous authors (Péra et al. 1999) and Bonavetti (2001), the addition of limestone powder to cement mortar completes the fine fraction in the particle size distribution curve of cement without an increment on water demand that improves the packing density; it blocks the capillary pores and increases the strength of the composites. This finding was also noted in the NHL5/PDS mortars (Tables 3 and 4). Here, the replacement of NHL5 by 10 and 20% PSD slightly decreased the water demand of the NHL5 mortar by 7% and increased strength when compared to the control mortar. Furthermore, the above authors reported that the limestone dust constitutes nucleation points for calcium hydroxide crystals at early hydration ages, accelerating the hydration of clinker particles thus increasing early strength. However, the stone dust does not have pozzolanic properties and does not produce C-S-H (calcium silicate hydrates).

Table 2. Compressive (F_C) and flexural (F_f) strength of mortars at 28 days.

<table>
<thead>
<tr>
<th>Mortar Type</th>
<th>F_C (MPa)</th>
<th>COV%</th>
<th>F_f (MPa)</th>
<th>COV%</th>
</tr>
</thead>
<tbody>
<tr>
<td>100% NHL5 (control)</td>
<td>9.5</td>
<td>1.8</td>
<td>0.9</td>
<td>1.8</td>
</tr>
<tr>
<td>10% GGBS (G1)</td>
<td>9.9</td>
<td>7.5</td>
<td>2.0</td>
<td>0.9</td>
</tr>
<tr>
<td>20% GGBS (G2)</td>
<td>21.5</td>
<td>2.7</td>
<td>1.5</td>
<td>4.7</td>
</tr>
<tr>
<td>10% RHA (R1)</td>
<td>12.0</td>
<td>2.9</td>
<td>1.4</td>
<td>0.9</td>
</tr>
<tr>
<td>20% RHA (R2)</td>
<td>14.1</td>
<td>2.8</td>
<td>1.7</td>
<td>2.9</td>
</tr>
<tr>
<td>10% PSD (D1)</td>
<td>13.1</td>
<td>7.5</td>
<td>1.3</td>
<td>5.8</td>
</tr>
<tr>
<td>20% PSD (D2)</td>
<td>11.8</td>
<td>2.7</td>
<td>0.9</td>
<td>4.5</td>
</tr>
</tbody>
</table>
3.2 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 (Table 3), the porosity of the RHA mixes and the NHL5 (control) mortar are similar, comparable to the 18-40% values documented by Moropoulou et al. (2005) for hydraulic lime mortars from ancient structures in the Mediterranean Basin. However, the GGBS and stone dust mortars have lower porosities. The porosity of the control mortar agrees with results by Gulotta (2013) who reported c.21% values for NHL5 mortar, with standard siliceous (quartz) sand at B/A ratio of 1:3 by weight.

The results showed that replacing NHL5 with GGBS significantly lowers the porosity of NHL5 mortars: an approximately 46 and 33% decrease was noted for the 20 and 10% GGBS mixes respectively when compared to the NHL5 (control) mortar. This agrees with observations made by Griffin (2004), who reported that addition of GGBS to non-hydraulic lime mortars decreased porosity by c.25%. According to Siddique and Bennacer (2012), the addition of GGBS to lime mortar results on smaller gel pores and fewer larger capillary pores; a finer pore structure that lowers porosity and enhances durability.

The replacement of NHL5 by 10 and 20% PSD reduced the porosity of the control mortar by about 16%. This decrease can be on account of the filler effect of PSD that enhances the density of the lime matrix reducing porosity. The presence of stone dust acting as nuclei thus facilitating clinker hydration may have also contributed to porosity reduction.

In contrast with the GGBS replacement significantly lowering the porosity of NHL5 mortars, replacement of NHL5 by RHA does not significantly impact porosity (Table 3). This is probably due to the much greater specific surface (Walker and Pavía 2010, 2011) and higher water demand of the RHA (Tables 1) increasing the amount of pores. Although, both GGBS and RHA have a similar particle size, RHA has a much greater specific surface (Walker and Pavía 2010, 2011) and a higher water demand (Tables 1). It is well known in concrete technology that many pores are remnants of space once filled with water, and therefore increasing water/binder ratio increases porosity. Papayianni and Stefanidou (2006) stated that, in lime mortars, water/binder ratio is the most important factor influencing porosity.

Table 3. Hygric properties of mortars at 28 days

<table>
<thead>
<tr>
<th></th>
<th>Porosity (%)</th>
<th>COV%</th>
<th>Water absorption (%)</th>
<th>COV%</th>
<th>Capillary suction (kg/m².min⁰.⁵)</th>
<th>COV%</th>
</tr>
</thead>
<tbody>
<tr>
<td>100% NHL5 (control)</td>
<td>20.0</td>
<td>2.4</td>
<td>10.6</td>
<td>2.7</td>
<td>1.2</td>
<td>2.9</td>
</tr>
<tr>
<td>10% GGBS (G1)</td>
<td>13.3</td>
<td>2.4</td>
<td>6.8</td>
<td>2.6</td>
<td>0.6</td>
<td>1.5</td>
</tr>
<tr>
<td>20% GGBS (G2)</td>
<td>10.6</td>
<td>1.7</td>
<td>5.3</td>
<td>1.7</td>
<td>0.4</td>
<td>4.2</td>
</tr>
<tr>
<td>10% RHA (R1)</td>
<td>20.7</td>
<td>0.5</td>
<td>11.1</td>
<td>0.7</td>
<td>0.9</td>
<td>3.4</td>
</tr>
<tr>
<td>20% RHA (R2)</td>
<td>19.7</td>
<td>1.2</td>
<td>10.6</td>
<td>1.4</td>
<td>0.7</td>
<td>0.3</td>
</tr>
<tr>
<td>10% PSD (D1)</td>
<td>16.3</td>
<td>3.0</td>
<td>8.5</td>
<td>3.2</td>
<td>0.9</td>
<td>6.7</td>
</tr>
<tr>
<td>20% PSD (D2)</td>
<td>16.8</td>
<td>3.4</td>
<td>8.7</td>
<td>5.5</td>
<td>1.0</td>
<td>3.4</td>
</tr>
</tbody>
</table>
It is generally accepted that there is a direct relationship between increasing strength and decreasing the porosity. This was evident from GGBS and PDS results however, in the RHA mortars, the increase in strength is not coupled to a drop in porosity.

A similar trend was observed for water absorption, as this property strongly relates to porosity. According to the results in table 5, 19% water absorption decrease was observed for the 10% and 20%PSD mixes and over 49% decrease for the 20%GGBS mortars (with the lowest porosity). The NHL5/RHA mortars, with porosity similar to the control mix, show either slightly higher or similar absorption than the control mortar.

All additions investigated lower the capillary suction of NHL5 mortars. GGBS and RHA are generally more efficient at lowering suction than the stone dust, and this is probably due to the additional cementitious hydration products blocking capillary pores. As it can be seen from the results in Table 5, the suction of the GGBS mortars is the lowest, agreeing with their lowest porosity and water absorption. The 20%GGBS mortar shows the lowest suction (with a 64% decrease with respect to the control mortar) followed by the 10%GGBS and 20% RHA mixes with a 49 and 41% decrease in suction (respectively). Although RHA replacement lowered neither porosity nor water absorption, it lowers capillary suction, this is probably due to the pozzolanic hydrates blocking capillary pores.

4. CONCLUSION

The replacement of NHL5 with GGBS, RHA and stone dust enhances the compressive and flexural strength of NHL5 mortars at 28 days. Replacement of NHL by 20% GGBS increases compressive strength by 125% and either doubles or increases by 50% its flexural strength. Replacing 10-20% NHL5 with RHA enhances compressive strength of NHL5 mortars to a smaller extent (by 26 -47%) and significantly enhances flexural strength.

A 10% increase in GGBS replacing NHL5 doubles the compressive strength of the NHL5 mortars (from 10 to 21.5 MPa) whereas a 10% increase in RHA raises strength by about 20%.

GGBS, RHA and stone dust lower capillary suction of NHL5 mortars. GGBS and RHA are more efficient at lowering suction than the filler as additional cementitious hydration products block capillary pores.

Replacing 20 and 10% NHL5 with GGBS significantly lowers the porosity of NHL5 mortars (46 and 33% decrease respectively); their capillary suction (49and 64% drop) and water absorption (c.49% drop for the 20%GGBS).

Replacing NHL5 with RHA does not lower porosity or water absorption but reduces capillary suction (the 20%RHA mix shows a 41% decrease). The higher porosity/water absorption of the NHL5/RHA mortars are probably due to the much greater specific surface and higher water demand of the RHA increasing the amount of pores.

Replacing NHL5 with stone dust rises strength and lowers porosity/water absorption (by c.16% and 19% for 10 and 20% replacements respectively) of NHL5 mortars; probably due to the filler effect increasing packing density and providing nuclei that accelerate NHL5 clinker hydration.

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