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A Study of the Impacts of Calcitic Aggregates on the Properties of Air Lime Mortar

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ABSTRACT: Preliminary studies have been undertaken to assess the impact of limestone aggregate characteristics on the compressive/flexural strengths of air lime mortar. A number of Binder/Aggregate (B/Ag) ratios were used (1:2, 1:3, 1:4), at curing times of 14 and 28 days. It has been confirmed that different types of limestone can yield higher mortar compressive strengths when compared with the use of silicate aggregate. Some results were found to differ from previous research, despite having the same binder and aggregates, at the same B/Ag ratio and curing times. One key difference is the particle size distribution of the respective aggregates, and further studies will seek to establish the significance. Flexural strength results appear to be unaffected by aggregate type.

Keywords: compressive strength, particle size distribution, calcitic aggregate, air lime, limestone

NOTATION

H_2CO_3 carbonic acid;
$Ca(OH)_2$ calcium hydroxide;
$CaCO_3$ calcium carbonate;
B/Ag binder/aggregate;
SEM scanning electron microscopy;
MIP mercury intrusion porosimetry;
TGA thermo-gravimetric analysis

1 INTRODUCTION

The need for low energy materials has become increasingly recognised over the past decade; the construction industry accounts for around 50% of the UK's total carbon emissions so there is a demand for low energy materials that would help the government achieve their target of reducing emissions by 80% by 2050. Consequently, the current research is being undertaken at a crucial time, with one positive outcome being the wider usage of low energy materials, including lime mortar.

With that in mind, it is important to note that while low energy materials are valuable, the environmental benefits must not be at the cost of mortar properties. Air lime mortars have been found to have superior properties when compared with other binders (Lawrence, 2006) [1], and ideally the mortar properties can be optimised with careful mix design.

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The research originated as a result of findings that with the use of certain types of limestone as an aggregate, stronger mortars were produced than with the most commonly used silica sand mortars (Lawrence, 2006; Lanas and Alvarez, 2003; Arizzi and Cultrone, 2012) [1,2,3]. Since the silicate sand itself is stronger than limestone aggregate, these findings were unexpected. The primary role of aggregates in mortar and concrete is to give structure to the material (Farey, 2003) [4] and as silica sand is an inert material, there is little research that investigates the impact of aggregate properties on mortar.

Lime mortars are primarily used in restoration work due to their similarities and therefore compatibility with the original masonry being repaired/conserved. One of the key factors influencing this is the compressive strength of the lime mortar, which is much lower than that of cement mortar. If the mortar is too strong, then the original masonry can end up becoming damaged even further. This is due to higher strength mortars having less ability to accommodate movement in the masonry, therefore a build-up of stresses will cause the masonry to fail (Mosquera et al., 2002) [5].

Little is known about the higher strengths that have been observed with some limestone aggregates, which have been found to be 3x higher than silica sand mortars in some cases (Lawrence, 2006) [1], although suggestions have been made that a similarity between limestone aggregate and binder matrix may have caused a superior bond (Lanas and Alvarez, 2003) [2]. As a result, the current research is at the forefront in this field.

2 THE ROLE OF CARBONATION

Carbonation is the primary chemical reaction that occurs during setting of air lime mortar, and is influenced by a number of factors; water content of a sample, relative humidity (RH) of curing, temperature and the porosity of the material.

The carbonation process describes the evolution of a mortar through chemical hardening, and can be split into two stages. In the first stage, CO_2 reacts with moisture on the surface of the mortar or in the air to form carbonic acid. This can be summarized in the equation below.

$$H_2O + CO_2 \rightarrow H_2CO_3$$

$$(1)$$

Following this, the carbonic acid diffuses into the sample to react with the portlandite ($Ca(OH)_2$), transforming it into calcite ($CaCO_3$). Equation 2 summarizes this.

$$H_2CO_3 + Ca(OH)_2 \rightarrow CaCO_3 + 2H_2O$$

$$(2)$$

During this process, samples see a weight gain upon transformation of calcium hydroxide to calcium carbonate. Furthermore, the pore structure changes over the carbonation period as a result of the transformation of portlandite into calcite; a decrease in total pore volume can be seen (Van Balen and Van Gemert, 1994) [6].

Van Balen and Van Gemert (1994) [6] state that upon drying, 2 different stages are present regarding water transport. The first of these stages is the shortest and relates to capillary action, where carbonation can be expected to be negligible. The second relates to water vapour diffusion, where Fick's first law can be applied.

Fick's first law of diffusion can be used to describe the process of CO_2 penetrating the pores in the mortar:

$$J = -D\frac{\delta \phi}{\delta x}$$

$$(3)$$

Where J = diffusion flux, D = diffusion coefficient, ϕ = concentration of substance per unit volume and x = position in length.

Arandigoyen and Alvarez (2003) [7] demonstrated the part carbonation plays in the porosity of mortar. Hydrated lime/cement mortars were used here, and a porosity decrease of 10% was observed as a result of carbonation; it was found that fewer pores of 1µm can be found. Cizer et al. (2006) [8] suggested that with an increase in open porosity of the mortar, a subsequent increase in carbonation depth was noticed. Lawrence et al. (2007) [9] stated that the volume of 0.1µm pores increases as a result of carbonation in air lime mortars. Furthermore, Lawrence et al. (2006) [10] assert that the pore size distribution is likely to have an influence on the rate of carbonation reaction.

Lawrence et al. (2006) [10] say that even when the carbonation process is thought to have ended, a substantial amount of lime remains uncarbonated. This is due to calcite crystals forming around the portlandite crystals and thus preventing further diffusion of CO_2 into the mortar.

At low temperatures, Dheilly et al. (2002) [11] revealed that the carbonation process speeds up. This is likely due to higher temperatures causing a faster drying of the mortar and thus less moisture is available for the first stage of carbonation (Equation 1). Van Balen and Van Gemert (1994) [6] suggest that 20°C provides the optimum temperature.

A high RH has been observed by Lanas et al. (2005) [12] to allow a higher level of carbonation in both air and hydraulic lime mortars. This is due to the CO_2 reaction being improved as a result of the greater amount of water that is present in the atmosphere.

With the exception of a some studies (Lawrence et al., 2007; Lanas et al., 2005) [9,12], silicate sand is primarily used as aggregate in mortars. It is therefore beneficial to look further at calcitic aggregates, in order to explore their effect on carbonation in more depth.

Since carbonation is the reason for strength gain in air lime mortar, it would follow that mortars with higher strengths might have a higher level of carbonation. This could result from use of more porous aggregates which allow improved CO_2 diffusion leading to a greater depth of penetration than mortars with less porous aggregates. Carbonation also causes a decrease in the porosity of the mortar, which could also contribute to higher strengths.

3 MATERIALS AND METHODS

For the purposes of this study, 6 different limestone aggregates were compared against Standard sand, a silicate sand.

Since the focus of investigations was to assess the effect of aggregate type, the particle size distribution was kept constant. Table 1 shows the sieve sizes that were used and the percentage passing each sieve.

Table 1. Sieve sizes

Sieve size (mm)	% Passing
4	100
2	88
1.6	81.84
1	58.96
0.5	29.04
0.16	11.44
0.08	0

The Standard sand was graded as supplied, and the limestone aggregates were crushed and graded in the lab to match, with the addition of the 2mm fraction to all aggregates in order to reduce shrinkage in the mortars. Figure 1 below shows the particle size distribution.
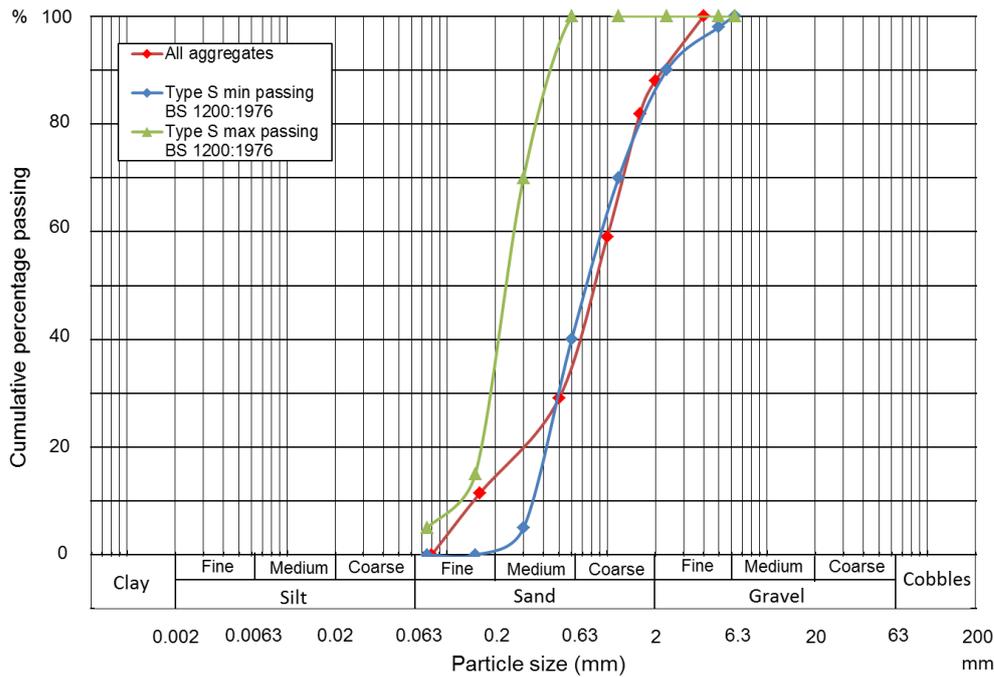
Figure 1. Particle size distibution compared with BS 1200:1976

As can be seen from Figure 1, the particle size distribution used in this study follows a similar trend to that specified in BS 1200:1976 [13].

In order to gain a wider range of results, 3 B/Ag ratios were used in this research; 1:2, 1:3 and 1:4 for each aggregate type at mortar curing times of both 14 and 28 days.

Due to the importance placed upon a mortar's workability as a result of the literature review, it was decided that rather than having a constant water/binder (w/b) ratio, a constant flow of 13cm would be used instead. This resulted from findings that if a mortar wasn't sufficiently workable, then a mason would add more water, thus altering the properties of the mortar. The flow table test was done in accordance with BS EN 1015-3:1999 [14].

Compressive/flexural testing of the samples were in accordance with BS EN 1015-11:1999 [15].

4   EXPERIMENTAL RESULTS AND DISCUSSION

Early results confirm that with use of a calcitic aggregate in air lime mortar, higher compressive strengths can be achieved than mortars with a silicate aggregate. Figure 2, below, shows the 14 and 28 day compressive strengths of all aggregates with a 1:2 B/Ag ratio.
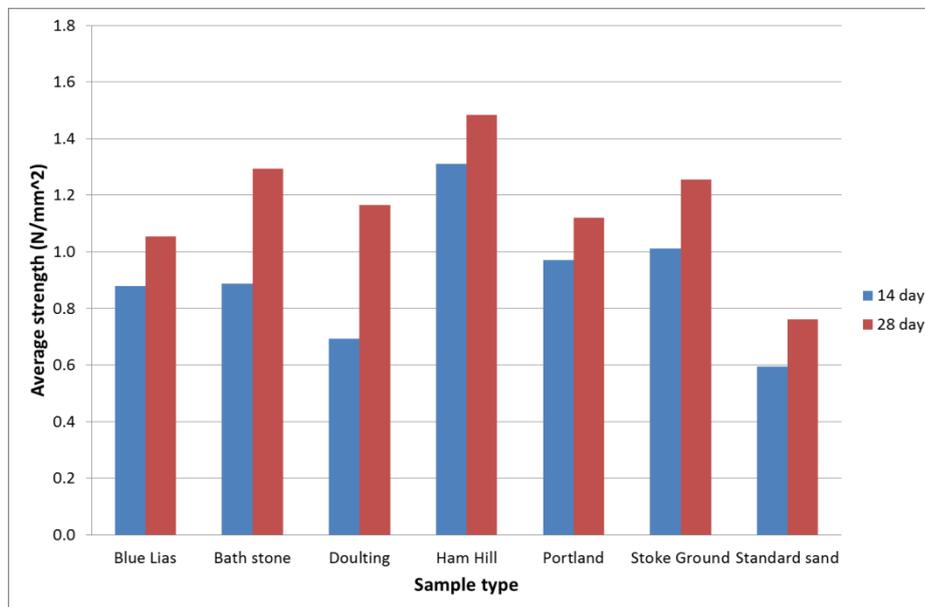

Figure 2. 14 and 28 day compressive strengths for 1:2 B/Ag

It is evident from Figure 2 that mortars made with Standard sand were the weakest when compared with the limestone aggregate mortars. This was confirmed with statistical testing at 99% confidence. Similar results were observed with the 1:3 mixes, which can be seen in Figures 3 and 4.
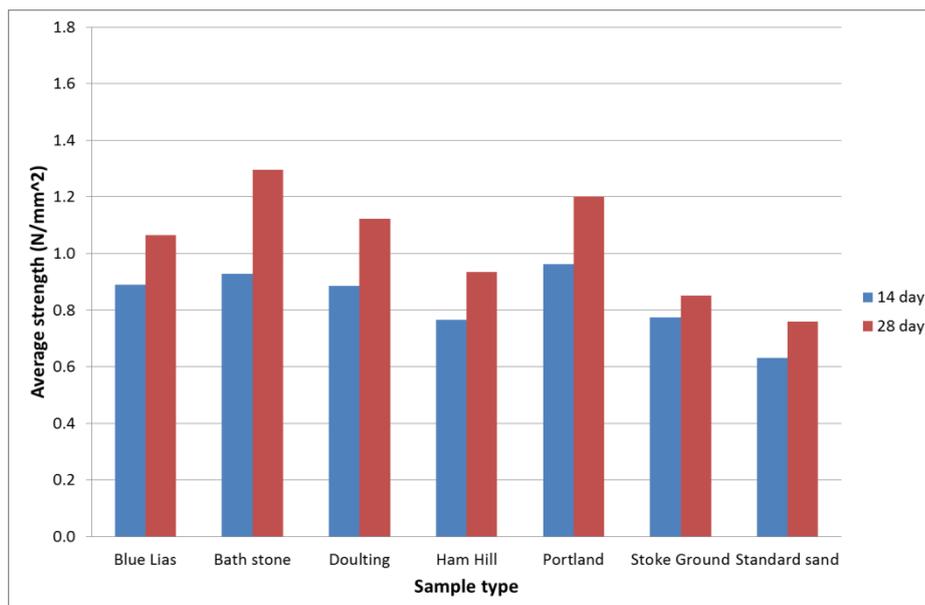

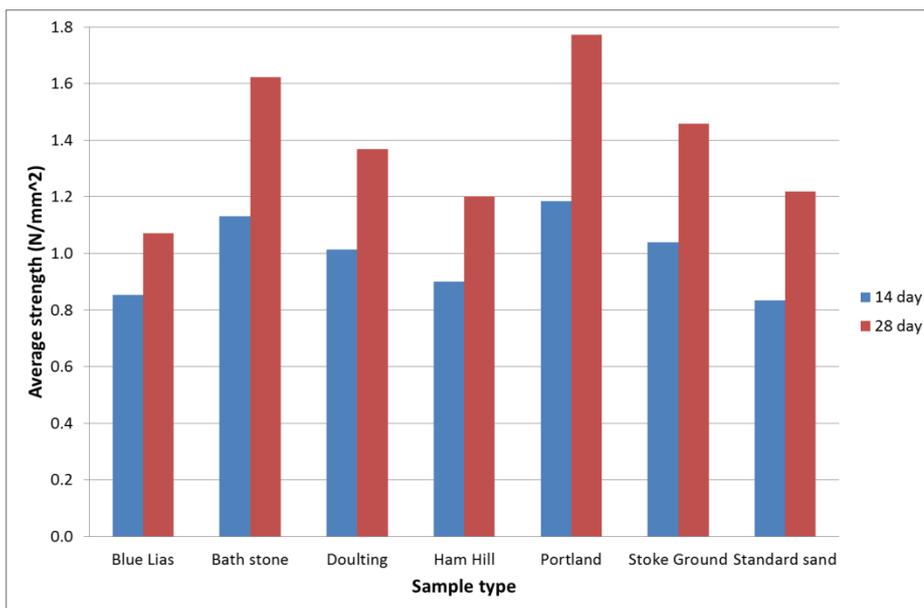**Figure 3.** 14 and 28 day compressive strengths for 1:3 B/Ag

Figure 4. 14 and 28 day compressive strengths for 1:4 B/Ag

While it has been found that the majority of the limestone aggregates produce higher compressive strengths in mortars than the silicate aggregate, the differences were not as large as found by Lawrence (2006) [1]. Figure 5 shows a comparison of the data obtained in the present study and that obtained by Lawrence (2006) [1].
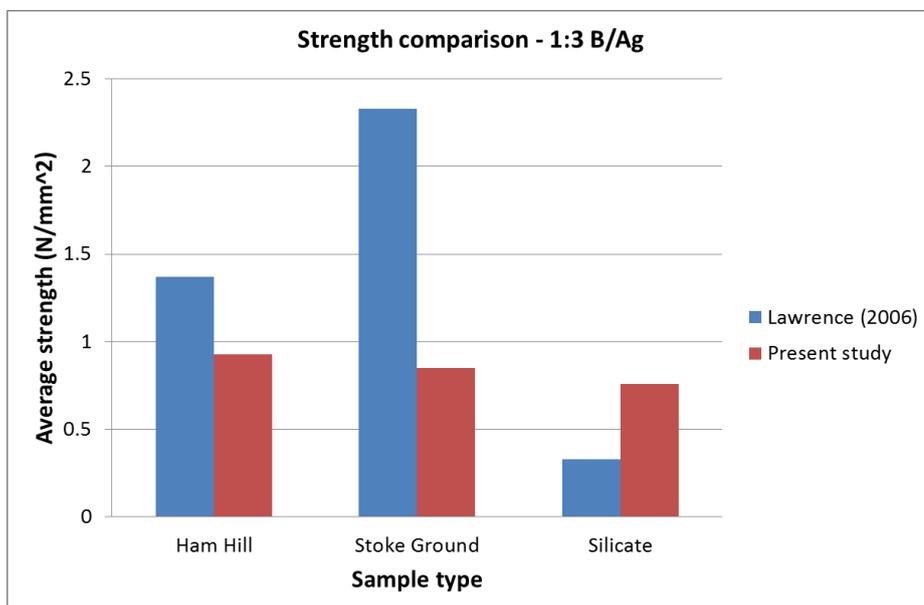

Figure 5. Comparison of strengths between current and previous work

Flexural strength testing did not show the same trends between different aggregates. There appears to be no relationship between aggregate type and flexural strength results. Figures 6-8 show flexural strength results.
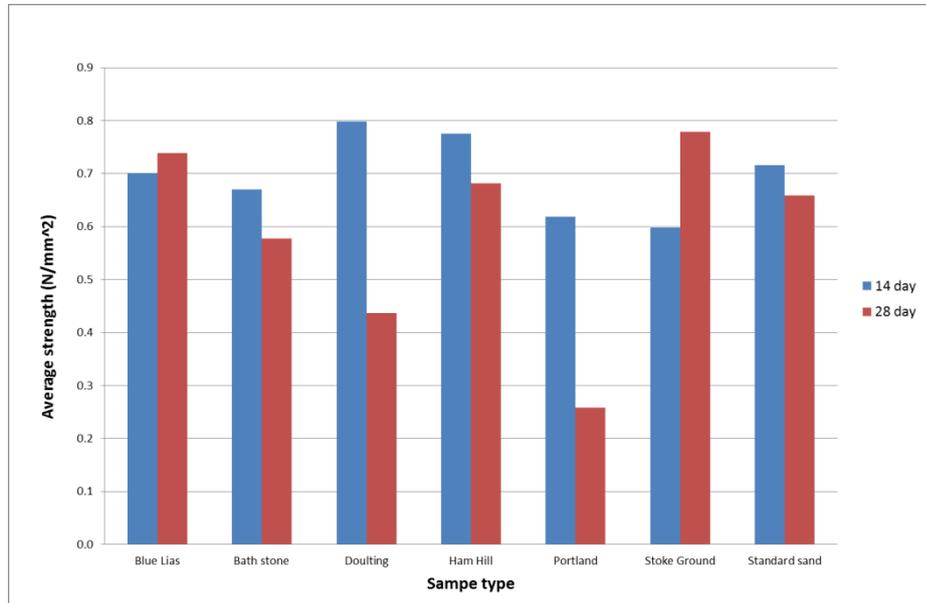

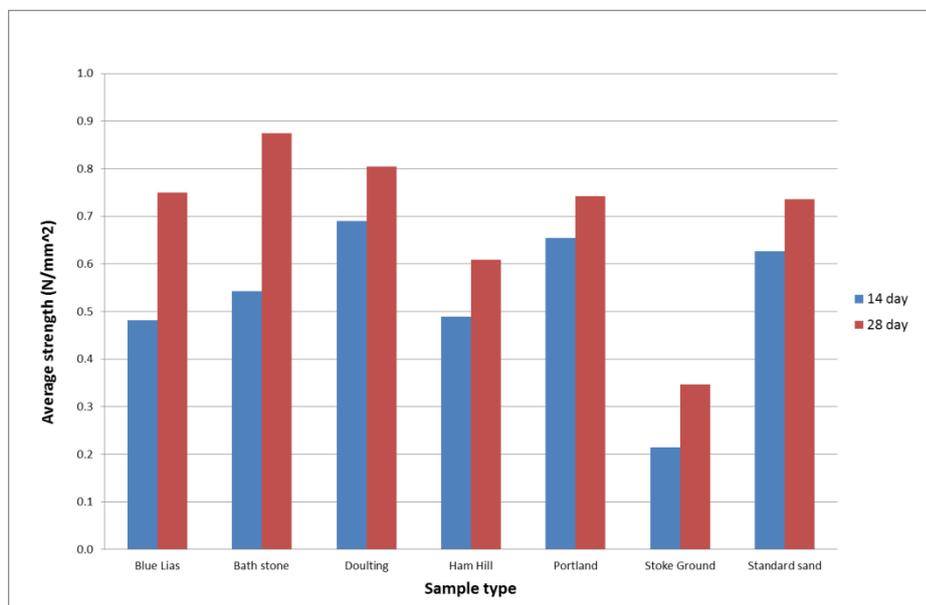**Figure 6.** 14 and 28 day flexural strengths for 1:2 B/Ag

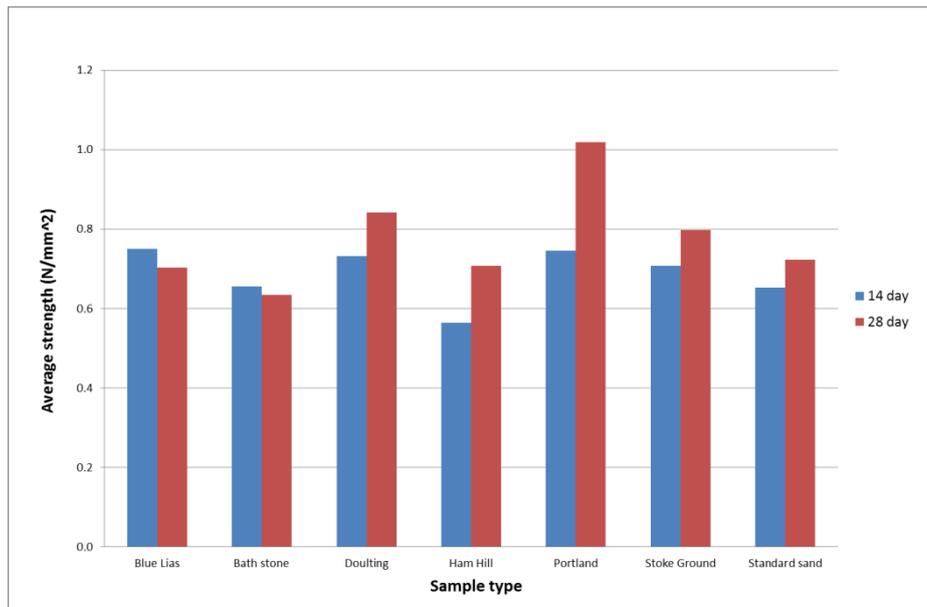**Figure 7.** 14 and 28 day flexural strengths for 1:3 B/Ag

Figure 8. 14 and 28 day flexural strengths for 1:4 B/Ag

Figure 9 shows the phenolphtalein staining on two of the samples from the 1:2 mix at 28 days. The left-hand image shows the Standard sand formulation (the weakest sample) while the right-hand sample contained Ham Hill limestone (the strongest sample).

Figure 9. Phenolphthalein staining at 28 days

Small differences can be observed between the two samples, which indicate that the mix containing Ham Hill is slightly more carbonated than than the mix containing Standard sand; the former has a smaller area of stained (uncarboanted) material. It is possible that this difference would be more noticeable over a longer curing period.

It is clear from the compressive strength results that despite using the same aggregate type and binder type at the same B/Ag ratio, Lawrence (2006) [1] obtained stronger mortars with both of the limestone aggregates. Conversely, the present study achieved stronger mortars with the silicate aggregate.

It has been identified that two differences exist in the mixes. Firstly, the water/binder ratio is different; however, the effect on strength has been assumed negligible due to work by Lawrence and Walker (2008) [16] that showed a minor effect of the water/binder ratio on compressive strength when

using air lime binder. Secondly, the particle size distribution is noticeably different between the two pieces of research. Lawrence (2006) [1] used a higher proportion of finer aggregate (for the limestones) than the current research, which could have led to the higher strengths observed. This would be in agreement with Fragata and Veiga (2010) [17] who also used calcitic aggregates compared against silicate aggregates, and found that the addition of fines led to an increase in both flexural and compressive strength of mortars.

There are several ways in which properties relating to aggregate type could have impacted the strength of the mortar. Failure in a mortar could occur as a result of low mechanical strength of the aggregate, where the aggregate would fail preferentially to the mortar, or due to poor binder/aggregate bond which would cause failure to occur at the interface of binder and aggregate. The latter would occur if the aggregate strength was higher than binder strength. Since it has been suggested that calcitic aggregates form a stronger bond with the binder (Lanas and Alvarez, 2003) [2], failure in these mortars may be occurring in the aggregate. It is possible that some of the paste is entering the pores of the aggrgegate so failure would be expected where no paste is in contact with the aggregate. Conversely, the silicate aggregate could be failing at the interface of binder and aggregate, if the bond wasn't as strong. SEM will be utilised to investigate further into the failure modes of the samples.

Furthermore, calcitic aggregates could have a better connected porosity (ie. improved gas and liquid diffusion) than silicate aggregates. This would allow better flow of CO_2 through the mortar, leading to a deeper level of carbonation and the potential for higher strengths. Figure 6 could be evidence of this and Mercury Intrusion Porosimetry (MIP) will give a better insight into the pore structure.

5 CONCLUSIONS AND FURTHER WORK

The research originally set out to verify whether use of calcitic aggregates in air lime mortars led to improved compressive strength over silicate sand mortars. It has been found that there was a significant difference between limestone mortar and silicate sand strengths. The majority of results showed higher strengths of limestone aggregate mortars at 95% significance or more. The exceptions were Stoke Ground at 1:3 B/Ag and Bath stone and Ham Hill, both at 1:4 B/Ag; these mixes were not found to be siginifically stronger than the respective Standard sand mortars.

Additionally, some differences have been observed between the level of carbonation of the strongest and weakest mortars respectively. These differences appear to correlate with strength; higher strengths have higher levels of carbonation.

Since the current research found strength increases to a lesser degree than Lawrence (2006) [1], further work is now seeking to establish reasons for this. Additional research will look in more detail at the effect the granulometry of the aggregate has on the mortar strength, as well as microstructural investigations using SEM, TGA and MIP.

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