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Impact of aggregate type on air lime mortar properties

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Abstract

In recent years, the need for low energy materials has become increasingly recognised. Government targets aim to achieve a decrease in carbon emissions by 80% before 2050. With the construction industry being responsible for approximately 50% of UK carbon emissions, an increased use of low energy materials could go a long way to achieving this target. With this in mind, it is also important that materials still have adequate properties to fit their purpose. For this study, four limestone aggregates were compared with a silicate aggregate in order to assess the impact of the aggregate type on the properties of air lime mortar (CL90). The primary focus was to assess the differences in compressive strength, and investigate reasons behind the measured differences. Without exception, the mortars made with limestone aggregate have higher compressive strengths than those made with silicate sand. Phenolphthalein staining shows slight differences in carbonation levels at 28 days, which could help to explain the strength differences observed. Furthermore, SEM analysis has revealed differences at the binder/aggregate interface between limestone aggregate mortars and silicate sand mortars.

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Keywords: limestone aggregate; silicate sand; air lime; compressive strength; scanning electron microscopy (SEM)

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1. Introduction

Lime mortar has been used for centuries in masonry construction. The past few decades have seen an increase in restoration work on old structures, where the compatibility of old and new materials is of paramount importance. This means that cement-based materials are inappropriate as a repair material due to the significantly higher strengths they reach over lime-based materials; a mortar with a higher strength than the original masonry would lead to more damage due to having less ability to accommodate movement. As a result, a build-up of stresses would cause the masonry to fail (Mosquera et al. 2002) [1].

Lime mortars are inherently weak in compression, and research has shown that higher strengths can be obtained with use of limestone aggregate over silicate aggregate (Lawrence, 2006) [2]. Since low strengths are synonymous with poor durability, higher strengths could lead to longer-lasting mortars. The higher strengths obtained are still much lower than cement mortar strengths so should not have a detrimental effect on existing masonry. Aggregates are primarily used to provide structure to a mortar (Farey et al., 2003) [3] and their role in mortar strength has been largely underestimated. Despite various studies concluding that limestone aggregates can produce higher strength air lime mortars (Lawrence, 2006; Lanas and Alvarez, 2003; Arizzi and Cultrone, 2012) [2,4,5], little is known about the reasons why. Additionally, while adequate strength is required for durability of a mortar, it is also vital to ensure other characteristics are sufficient; porosity, water retentivity and plasticity are just a few of the important properties. In the current climate, it is becoming increasingly recognized that carbon emissions need to be reduced; as the construction industry is responsible for around 50% of the UK's emissions (BIS, 2010) [6], the use of low energy materials can contribute to this reduction. Air lime is a low energy binder due to the fact that during curing, almost all of the CO_2 that was emitted during the manufacturing process is reabsorbed during carbonation (limetechnology, accessed 2013) [7]. Carbonation gives a mortar strength through the transformation of $Ca(OH)_2$ into $CaCO_3$. It is the primary chemical reaction that occurs during setting of air lime mortar, and is a self-limiting process. This is due to the formation of calcite crystals around the calcium hydroxide particles, which block CO_2 penetration and subsequently some portlandite ($Ca(OH)_2$) always remains uncarbonated (Houst and Wittmann, 2002) [8].

The research originated due to lack of knowledge surrounding the effect of aggregate type on mortar properties, particularly compressive strength. Consequently, four limestone aggregates were compared against a silicate sand (CEN Standard sand) to determine firstly the compressive strengths, and secondly whether there were any differences at a microstructural level for the different mixes. Scanning electron microscopy (SEM) was used for this analysis.

Nomenclature

SEM	scanning electron microscopy
$Ca(OH)_2$	position of
$CaCO_3$	calcium carbonate
$CaMg(CO_3)_2$	calcium magnesium carbonate (dolomite)
SiO_2	silicon dioxide
B/Ag	binder/aggregate

2. Effects of aggregate type

Several aspects relating to aggregates can have an impact on the strength of the mortar. The most commonly used aggregate is silicate aggregate, which is hard and chemically inert. Limestone aggregates can be calcitic or dolomitic; calcitic aggregates are in the form $CaCO_3$ whereas dolomitic aggregates are $CaMg(CO_3)_2$. Calcitic aggregates are used in the current study (as well as silica sand) and can either compose of angular or rounded grains. Differences in the porosity of aggregates can have an impact on overall mortar strength, due to differences in the diffusion of CO_2 through the sample. Aggregates with a higher porosity would allow a quicker rate of CO_2 diffusion, thus leading to a quicker rate of carbonation. Since carbonation is the primary strength mechanism in air lime mortar, strength would be expected to be increased.

If the aggregate has a low mechanical strength, failure of the mortar is likely to occur through the aggregate. Conversely, aggregates with high mechanical strength will result in failure at the binder/aggregate interface, assuming the aggregate is stronger than the bond.

It has been suggested in previous research (Lanas and Alvarez, 2003) [4] that a similarity between the limestone aggregate and binder matrix can also form a superior bond (over silicate aggregate) and as a result strengths are higher.

3. Materials and methods

Air lime mortar specimens (using CL90 hydrated lime) of dimensions 40x40x160mm were made in accordance with BS EN 1015-11: 1999 [9], in order for compressive/flexural strength testing to be undertaken. Samples were cured for a period of 28 days in air. Four of the samples were made with different limestone aggregates (Monks Park Bath stone, Doulting, Portland and Stoke Ground), while the fifth was made with a silicate sand. A binder/aggregate (B/Ag) ratio of 1:2 was used for all mixes. In order for the tests to be able to isolate differences in mortar properties based on aggregate type, all limestone aggregates were made to match the silicate sand (Standard sand CEN 196-1). An additional 2mm fraction was added to all five aggregates in order to help reduce shrinkage in the mortar specimens. Table 1 shows the sieve sizes used.

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

Figure 1 shows the particle size distribution used in this study, compared with BS 1200: 1976 [10]. It is clear that a similar trend exists.

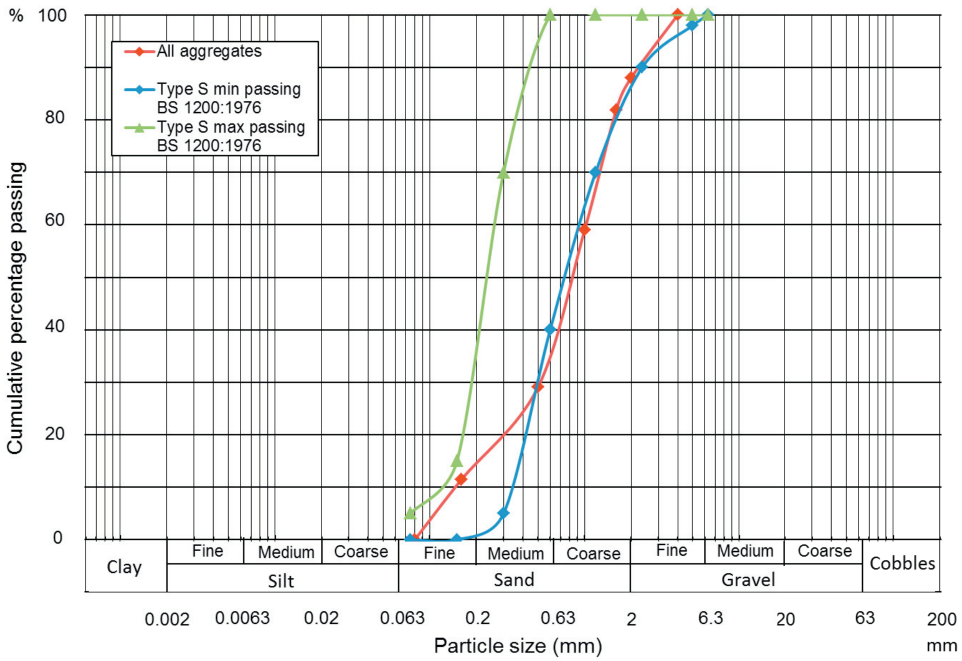

Figure 1. Particle size distribution

Due to the importance of workability of a mortar, it was decided that rather than having a constant water/binder (w/b) ratio, the flow would be kept constant at 13cm +/- 0.5cm. The main reason behind this was due to findings that if a mortar wasn't deemed workable by the mason, more water would be added to the mix, thus altering the properties (Gunn, 2005) [11].

The flow table test measures consistence of a mortar; mortar is tamped down into a truncated cone then once the cone is removed, 15 drops of the table are made at a rate of one per second. The diameter of the mortar spread is then measured. This was done in accordance with BS EN 1015-3: 1999 [12].

Table 2 shows the mix specifications used for each of the mixes in order to achieve a 13cm flow.

Table 2. Mix design

Sample	Lime (g)	Agg (g)	Water (g)	w/b
Bath stone	250	2050	539	2.16
Doulting	250	2238	513	2.05
Portland	250	2781	493	1.97
Stoke Ground	250	2279	535	2.14
Standard sand	250	1536	250	1.00

The w/b ratio is known to impact strength of cement mortars and hydraulic lime mortars. Abrams' Law shows the relationship between the strength of fully compacted concrete and the w/b ratio (Equation 1).

$$f_c = K_1/K_2^{w/c} \tag{1}$$

where f_c is the water/cement ratio, K_1 and K_2 are constants. Compressive strength in cementitious mortars is known to follow Abram's Law, and is inversely proportional to water/cement ratio (Neville, 2005) [13]. Lawrence and Walker (2008) [14] have shown that for air lime mortars, with the exception of the lowest water/lime ratio, there is very little difference in the compressive strengths of the mortars with increasing water content.

4. Experimental results

Results from the compressive strength testing confirmed the findings from literature that limestone aggregates (Bath stone, Doulting, Portland and Stoke Ground) can produce stronger mortars than silicate sand (Standard sand), as can be seen in Figure 2. While it is possible that the angular nature of the crushed limestone could be providing greater mortar strengths than the more rounded silicate sand, SEM testing has suggested something more complex.

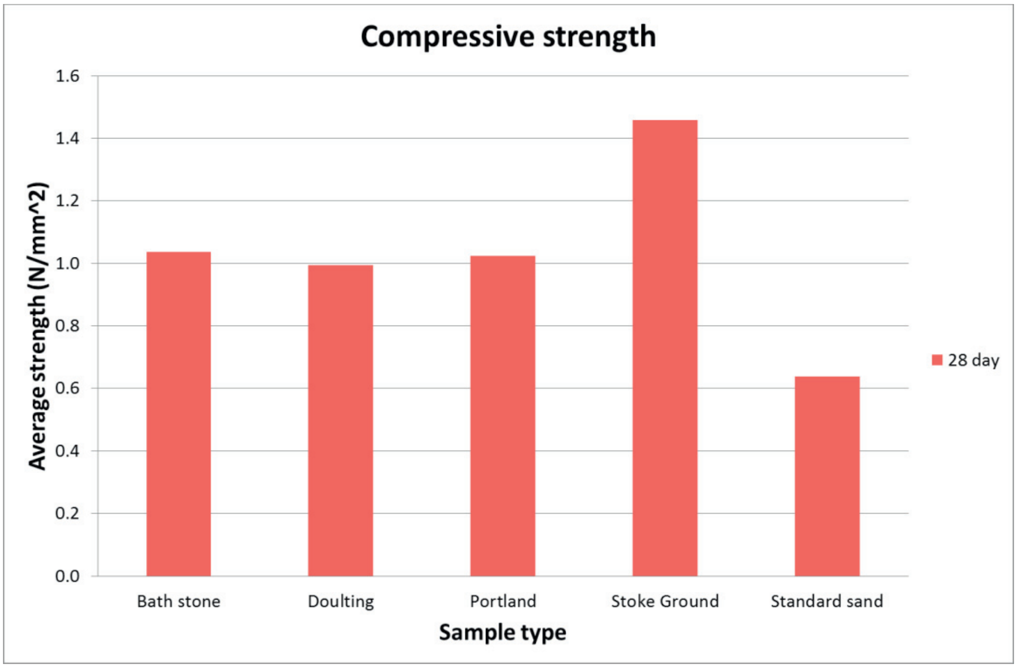

Figure 2. 28 day compressive strengths

Having confirmed previous knowledge, samples were taken from the outer edge of each of the fractured specimens in order to undertake SEM analysis to try to help understand the mechanisms behind the strength differences. The SEM used was JEOL SEM6480LV. It was used under low vacuum conditions so that the samples didn't require coating. BSE (back-scattered electron) mode was used, in combination with EDX which enables identification of the elements that make up the sample.

Figure 3 shows a sample of the specimen made with Portland aggregate. The lighter area indicates a heavier element; in this case it is expected that the lighter area contains $CaCO_3$ and the surrounding darker areas consist of $Ca(OH)_2$. This seems to be confirmed by the EDX analysis which indicates a higher proportion of oxygen in the lighter areas. The large area of lighter material appears to be an aggregate particle that is almost entirely coated with calcium carbonate (calcite) crystals. This supports the notion that calcitic aggregate can act as a nucleation site for calcite crystals (Lanas and Alvarez, 2003) [4].

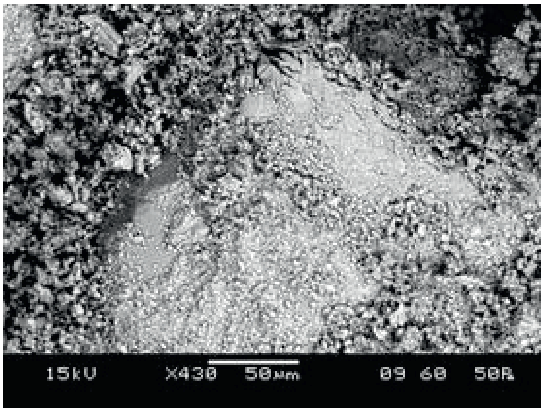

Figure 3. SEM image of sample containing Portland limestone aggregate

If more calcite crystals are forming, this suggests the carbonation is of a better quality than samples where aggregates are coated in fewer crystals. This could be contributing to the higher strengths that are being achieved with limestone aggregate mortars.

Figure 4 (below) shows a specimen containing silicate sand (Standard sand) at the same magnification as Figure 3. The aggregate particle can be seen much more easily than the Portland aggregate in Figure 3, which suggests there are less calcite crystals forming on the aggregate surface. Furthermore, the aggregate in Figure 4 appears to exhibit a larger amount of space between it and the binder. This implies that an inferior bond exists when compared with the Portland sample in Figure 3, which could be responsible for the lower strength achieved. A similarity between binder matrix and aggregate has been linked with higher mortar strengths (Lanas and Alvarez, 2003) [4] in limestone aggregate mortars; the findings shown in Figures 3 and 4 imply that this is a valid hypothesis.

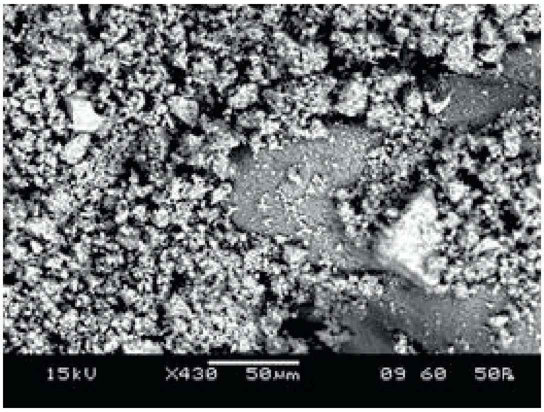

Figure 4. SEM image of sample containing Standard sand

Figure 5 shows a sample made with Doulting aggregate. Again, the lighter area appears to be an aggregate particle. Cracking is evident on aggregate, as well as between the aggregate and the binder, suggesting a weaker bond than that obtained from the Portland sample. When no cracks exist at the binder/aggregate interface, cracking in the aggregate indicates that the aggregate itself is weaker than the bond that has formed at the interface. In this instance, there are cracks both within the aggregate and at the interface between binder and aggregate, which could point to similar compressive strength of aggregate and mortar respectively.

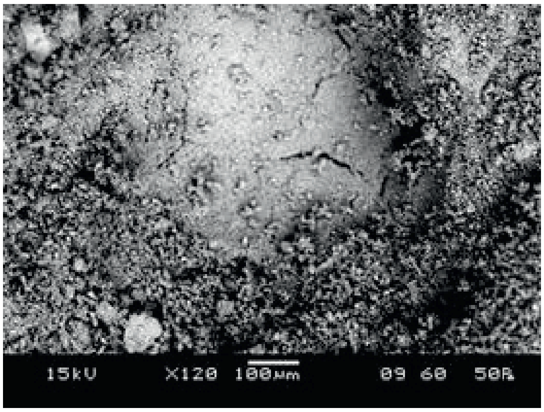

Figure 5. SEM image of sample containing Doulting limestone

The Stoke Ground sample in Figure 6 (the strongest mortar) has less discontinuity than the other samples between binder and aggregate, although some cracking is evident in the aggregate itself (right hand side of Figure 6). It is possible that the Stoke Ground aggregate is causing a more thorough level of carbonation at the binder/aggregate interface. A combination of X-ray diffraction (XRD) and thermogravimetric analysis (TGA) could help to establish this.

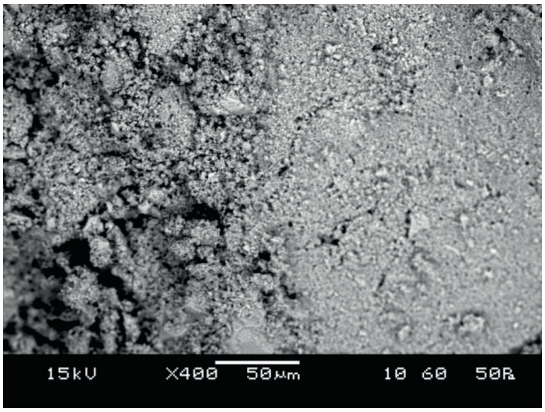

Figure 6. SEM image of sample containing Stoke Ground limestone

In Figure 7 (below), the lighter area shows a particle of Bath stone aggregate, with some calcite crystals on the surface. It is evident that there are fewer crystals than with Portland limestone, although the average compressive strengths are quite similar. Despite this, there is good continuity between the aggregate and binder, suggesting a good bond.

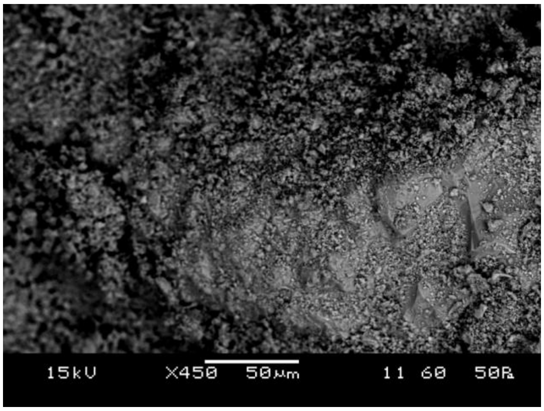

Figure 7. SEM image showing sample containing Monks Park Bath stone

Phenolphthalein staining was used to determine levels of carbonation in the samples. Figure 8 below shows a Standard sand sample and a Portland sample.

Figure 8. Left - Standard sand sample; Right - Portland sample

The dark area shows uncarbonated material in the samples; it can be seen that the sample containing Portland has a slightly smaller area of uncarbonated material, which could contribute to strength differences. It is also possible that the limestone aggregates allow quicker carbonation of the samples, due to having intrinsically higher levels of porosity.

Table 3 shows the average carbonation depth of each sample after 28 days curing. All mixes had a binder/aggregate ratio of 1:2 as well as identical partical size distribution (Figure 1).

Table 3. Average carbonation depth

Sample	Depth (mm)
Bath stone	6
Doulting	7
Portland	6
Stoke Ground	6
Standard sand	5

It is evident that there is a slight variation between the different mortar mixes. However, it is clear that these differences are not directly related to the strength values that can be seen in Figure 2. While a 2mm difference in carbonation depth is evident between the Doulting sample and Standard sand sample (the value for Doulting is 1.4x higher), the Doulting sample is 1.3x stronger than Standard sand sample. Furthermore, the Stoke Ground sample was the strongest in compression but does not have the greatest depth of carbonation suggesting that while the different aggregates are causing small differences in carbonation depth of the mortar, it is unlikely that the variances have had an impact on the compressive strength. It is more feasible that the higher porosity of the limestone aggregate is causing improved binder/aggregate bond rather than higher levels of carbonation. Mercury intrusion porosimetry (MIP) will help determine whether porosity is influencing strength results.

5. Discussion

It is clear from the SEM analysis that differences exist in the microstructure between the different aggregates used in the mortar. Most noticeably, the difference between the binder/aggregate interface of the silicate sand mortar and limestone aggregate mortars respectively.

On the surface of the silicate aggregate there were far fewer calcite crystals developing after 28 days when compared with the limestone aggregate. A possible reason for this could be due to the lower porosity of the silciate aggregate, meaning binder couldn't enter the pores in the same way as it could with limestone aggregate, which has a higher porosity. Binder entering the pores would create a better bond and consequently a higher compressive strength.

Ideas have been put forward about reasons for higher strengths (Lawrence, 2006; Lanas and Alvarez, 2004) [2,4] including the notion that calcitic aggregates act as a nucleation site for calcite crystals.

6. Conclusions

The current research has shown that with the use of different aggregate types in air lime mortar, the compressive strengths can be quite different; limestone aggregates have produced higher strength mortars. SEM analysis has revealed that there are also differences on a microstructural level. An important finding is that there are more calcite crystals forming on the surface of the limestone aggregates than the silicate sand, and still some differences between those as well. Furthermore, some of the aggregates were found to exhibit cracking within themselves and some were also found to have discontinuity between binder and aggregate. Finally, it has been found that while carbonation depths have slight differences with use of different aggregates in the mortar, it is unlikely that these are related to compressive strengths.

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