Chemistry and Materials Research, Vol.5 2013
Special Issue for International Congress on Materials & Structural Stability, Rabat, Morocco, 27-30 November 2013

A Critical Review of the Effect of Calcitic Aggregate on Air Lime Mortar

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Abstract. In recent years, the importance of aggregate type on the properties of mortars has become increasingly recognised. In the context of restoration, it is particularly important to achieve the optimum properties that provide the best compatibility between the repair mortar and the existing masonry. With that in mind, the properties of the aggregate should be given priority when designing the repair mortar mix. A critical analysis of the current state of the art is presented, identifying the areas of research that have not yet been explored thoroughly. The role of calcitic aggregates in mortar is one such area, and the paper presented here examines the notion that calcitic aggregates cause an increase in the strength of lime mortar. The review establishes the limited amount of knowledge existing on the subject and seeks to determine methods that will enable validation of the claim.

1 Introduction

There are three primary constituents of mortar; binder, aggregate (usually siliceous) and water. Aggregates have been found to have an effect on the properties of mortar, which could be attributed to the mineralogy, shape, surface roughness and porosity.

In the past few years, the similarities/differences between the aggregate and mortar composition have been thought to impact the mortar's properties. Lanas and Alvarez (2003) [1] make reference to this regarding the use of calcitic aggregate in air lime mortar, stating that the similarities between binder matrix and aggregate structure could be responsible for the higher strengths observed with the calcitic aggregate.

Based on current findings from the literature, it is clear that a gap exists in the knowledge surrounding the impact of calcitic aggregates on the performance of air lime mortars. It has been found that the use of calcitic aggregate in air lime mortars exhibits a greater strength than was to be expected (Lanas and Alvarez, 2003), (Lawrence, 2006) and (Arizzi and Cultrone, 2012) [1-3], and this is worth exploring further as it may have a positive impact on the use of air lime mortars in industry.

The paper will give a brief description of the types of aggregate used in construction, namely silicate-based and limestone respectively. Further to this, the current state of the art is assessed, with a focus on what research has been done so far. Based on this, suggestions of important tests are put forward, with a justification for their usage and what is hoped to be achieved.

2 Types of aggregate

Aggregate makes up the majority of a mortar mix, by volume, and its primary role is in reducing the drying shrinkage which could otherwise lead to severe cracking.

The most commonly used aggregate in mortars is natural sand, which generally has high silica content. Since it has been found that the use of calcitic aggregates in lime mortars can result in higher mortar strengths, their use could become more commonplace, but it is expected that this would only occur when more is known about the mechanisms responsible for the higher strengths.

It is worth noting here that different types of limestone have a different composition and pore structure, and for the purpose of this study, the focus will be on the oolitic Bath Stone. Previous work by Lawrence (2006) [2] found that the use of oolitic aggregate produced mortars with superior properties when compared with the use of bioclastic aggregate (also calcitic); hence, the interest in Bath Stone.

2.1 Siliceous aggregate

Silica sand is often in the form of quartz and has the chemical composition SiO_2 (silicon dioxide). It is a hard, chemically inert material.

2.2 Calcitic aggregate

Calcitic aggregate can either compose of angular or rounded grains, and has the chemical composition $CaCO_3$ (calcite). There are many different forms of calcitic aggregate; for example Bath Stone is an oolitic variety. Oolitic grains are round in shape and are 0.25-2mm in diameter.

Lawrence (2006) [2] found that mortars made from crushed oolitic stone were four times as strong as those made using silicate aggregates. This could be due to the similar pore structure that exists between the aggregate and mortar respectively (Lanas and Alvarez, 2003) [1].

It is possible that if both aggregate and mortar have a similar porosity, CO_2 movement through the sample would be more constant, potentially leading to a faster and more complete carbonation.

3 Effect of aggregate type on mortar

It has been found by Lanas and Alvarez (2003) and Arizzi and Cultrone (2012) [1,3] that pure limestone aggregates yield mortars with higher strengths than those containing siliceous aggregates. This is possibly due to the limestone aggregate structure being similar to the calcitic binder matrix Lanas and Alvarez (2003) [1], which results in a more uniform mortar composition.

Conversely, Pavia and Toomey (2007) [4] suggest that the two sands containing the highest amount of calcite actually produced the weakest mortars. However, it has been suggested that the reason for this could be due to the use of chalk, where low mechanical strength is an intrinsic property. It was found that the highly siliceous aggregate produced the strongest mortar.

It was noted by Carlos et al. (2010) [5] that with an increase in the proportion of fine limestone in relation to cement and gravel content, shrinkage was reduced. This may also be the case for lime mortars.

Naik et al. (2006) [6] compared the use of crushed dolomitic limestone against crushed quartzite in concrete and found that at early ages, the limestone was weaker but at later stages, it was either a similar strength or higher than the quartzite. Dolomitic limestone was also observed to yield the lowest autogeneous shrinkage and lowest resistance to chloride ion penetration.

4 Scope for future investigation

Based on the limited amount of literature surrounding the effect of different types of aggregate (specifically limestone aggregate), and the lack of knowledge of the possible mechanisms surrounding what knowledge does exist, there is clearly a lot of scope for further investigations to be undertaken.

The following tests are thought to be the most appropriate when considering repair mortars comparing different aggregate types.

4.1 Rheology

4.1.1 Mortar flow

Mortar flow is of primary importance, particularly concerning ease of application for the mason. Measurement of flow involves looking at the mean diameter of the spread of mortar, after being subjected to vertical impacts on a flow table, as described in BS EN 1015-3: 1999 [7].

De Vekey (2005) [8] suggested that mortar should be able to flow freely, whilst still maintaining an adequate viscosity. Additionally, workability should remain for a few minutes after being applied to the stones, before starting to stiffen.

Bauer et al. (2007) [9] propose that there is a consensus suggesting that use of flow table is insufficient for defining workability, and as such, suggest also using the laboratory Vane test to measure yield stress to assist in the understanding of flow table results.

With the use of different aggregate types, it is expected that the water/binder (w/b) ratio will vary in order to keep the desired flow consistent. Porosity of the aggregate and surface roughness may contribute to this effect.

4.1.2 Setting time

The setting time of lime is considerably longer than that of cement and consequently, cement is favoured in modern construction. The more rapid setting time enables masons to be more efficient during construction.

Subsequently, if the setting time of lime could be reduced, this may promote its use in modern construction rather than primarily as a restoration mortar. It is possible that the unexplained strength increase with use of calcitic over silicate aggregate may be a result of more rapid carbonation. As a result, the setting time of the mortar may be reduced.

4.1.3 Yield stress

A common method of measuring the yield stress of mortar is the vane test (Bauer et al., 2007) and (Hendrickx et al., 2008) [9-10].

Hendrickx (2008) [11] underline the importance of yield stress in helping to understand flow table results when assessing workability.

Furthermore, research by Hendrickx et al. (2009) [12] found that air lime mortar has almost 3x higher yield stress than cement mortar.

No mention has been made, to the author's knowledge, of a relationship between yield stress and aggregate properties.

4.1.4 Drying shrinkage

It has been found that aggregate type can have an effect on drying shrinkage. Naik et al. (2006) [6] state that when using dolomitic limestone as aggregate, both autogeneous and drying shrinkage were reduced in comparison with using river gravel or quartzite stone. It is worth bearing in mind that concrete was used in this study, however, this study can still prove useful as a comparison between different aggregate types.

In addition, Hughes et al. [13] showed that NHL 5 mortars made with silica sand observed increased shrinkage corresponding to an increase in fines content. Conversely, when carbonate ($CaCO_3$) sands were used, the highest shrinkage was found with the lowest fines content. Further investigations into the mechanisms would be beneficial.

4.2 Hardened mortar properties

4.2.1 Compressive/flexural strength

The primary focus of previous research on the use of limestone aggregates in mortar has involved looking at the compressive/flexural strengths of the mortars. Consequently, these tests are vital if any useful comparison is to be made, or claims are to be validated.

It is in general agreement that the compressive strength of air lime mortar increases more than double after the first 28 days up to a year of curing (Lanas and Alvarez, 2003) and (Moropoulou at al., 2005) [1,14].

Lanas and Alvarez (2003), Lawrence (2006) and Arizzi and Cultrone (2012) [1-3] found that pure limestone aggregates yield mortars with higher strengths than those containing siliceous aggregates. This is possibly due to the limestone aggregate structure being similar to the calcitic binder matrix (Lanas and Alvarez, 2003) [1], which results in a lack of discontinuity.

Arizzi and Cultrone (2012) [3] point out that there is a noticeable difference in the textural properties of calcareous and siliceous aggregates, with the former having more angular, rough and porous surface. It has been suggested that this impacts the degree of cohesion between aggregate surface and binder, possibly leading to the higher strengths exhibited with the use of calcareous aggregate.

4.2.2 Open porosity and pore size distribution

Carbonation is the primary chemical reaction that takes place in air lime mortars. The carbonation process describes the evolution of a mortar through chemical hardening, and for calcitic mortars it can be summarized by the following equation:

$$Ca(OH)_2 + CO_2 \leftrightarrow CaCO_3 + H_2O$$
(1)

During the carbonation process, the microstructure changes, as a result of the transformation of portlandite into calcite.

Arandigoyen et al. (2006) [16] demonstrated the part carbonation plays in the porosity of mortar. Hydrated lime/cement mortars were used here, and a porosity decrease of around 10% was observed as a result of carbonation; it was found that fewer pores of 1µm can be found.

Furthermore, Lawrence et al. (2006) [17] assert that the pore size distribution is likely to have an influence on the rate of the carbonation reaction.

As a result, it is suggested that both open porosity and pore size distribution should be ascertained, in order to gain an insight into the pore structure. It is hoped that the influence of different types of aggregate on the carbonation of the mortar will be clearly evident.

With the exception of Lawrence et al. (2007) and Lanas et al. (2005) [18-19], it has been found that silicate sand is primarily used as aggregate. Consequently, a further at calcitic aggregates would be beneficial, in order to explore their effects on carbonation in more depth.

4.2.3 Elastic modulus

A mortar that has a high modulus of elasticity is not appropriate for conservation; the modulus of elasticity of cement mortar is almost 3x that of stone (Marevalaki-Kalaitzaki, 2007) [20]. On the other hand, lime mortars also have a plastic zone that is not present in cement-only mortars (Arandigoyen and Alvarez, 2007) [21]. As a result, they are much more capable of accommodating movements of ancient masonry.

Furthermore, aggregates can have an impact on the modulus of elasticity. Winnefeld and Böttger (2006) [22] note that when a higher proportion of clayey fines is incorporated, the elastic modulus is reduced by up to 50%. The authors suggest that this may be attributed to the increase in w/b ratio required.

Limestone aggregates were also found to have an impact on modulus of elasticity; a higher percentage in concrete leads to a higher elastic modulus Carlos et al. (2010) [5]. It was suggested that during the plastic stage, paste could have entered the pores of the aggregate. This could affect the elastic modulus. Although these findings were in concrete, it is possible that similar results would also be found with lime mortars, and it is therefore worth investigating further.

4.2.4 Salt crystallization

Resistance to salt crystallization is an important factor to consider when designing mortars for the repair of historic masonry, as soluble salts can be very damaging. Lime mortars have a particular tendency to suffer from salt crystallization due to their high porosity (Henriques and Charola, 2000) [23].

Again, the author has found no mention about whether the type of aggregate used has an effect on the mortar’s resistance to salt crystallization. This is surprising, as it is evident that the type of lime binder was found to have an effect.

Pavia and Treacy (2006) [24] compared non-hydraulic and feebly-hydraulic lime, concluding that the former is more resistant to salt crystallization. The paper added that it wasn’t possible to examine the process of decay due to the rapidity of the decay. In contrast, Stewart et al. (2001) [25] assert that hydraulic lime had a better resistance to salt crystallization.

4.2.6 Water absorption

Water absorption has been defined as the ratio of the mass of water a mortar can retain, to the dry mass of the mortar Pavia and Toomey (2007) [4].

Pavia and Toomey (2007) [4] also suggest that aggregate properties can have an impact on the water absorption of the mortar, although not a substantial effect. The highest water absorption occurred in mortars that contained coarser, more rounded aggregate, with inferior grading. In contrast, the minimum water absorption occurred in mortars with the best grading, sharpest particle size and finest average particle size. Pavia and Toomey (2007) [4] also noted that calcite content did not affect water absorption. It is worth mentioning that the limestone aggregate contained both calcite and quartz, but the proportions of these were not specified in the paper.

Therefore, it may be beneficial to take a closer look at a variety of calcitic aggregates to determine whether this is true for a wide range, or indeed just one or two types.

4.3 Analysis

In order to establish the mechanisms behind the different mortar properties due to aggregate type, it is likely that microstructural analyses will be required. There are a few techniques that could prove useful here.

4.3.1 Scanning electron microscopy (SEM)

SEM is a useful technique for determining the morphology of the microstructure in mortars (Lanas and Alvarez, 2003), (Arandigoyen and Alvarez, 2006) and (Tuncoku and Caner-Saltik, 2006) [1,25,27], in addition to phases that are present in the mortar (Lanas and Alvarez, 2003) [1].

Figure 1 shows an SEM image of the exterior of air lime mortar made with oolitic aggregate (Lawrence, 2006) [2].

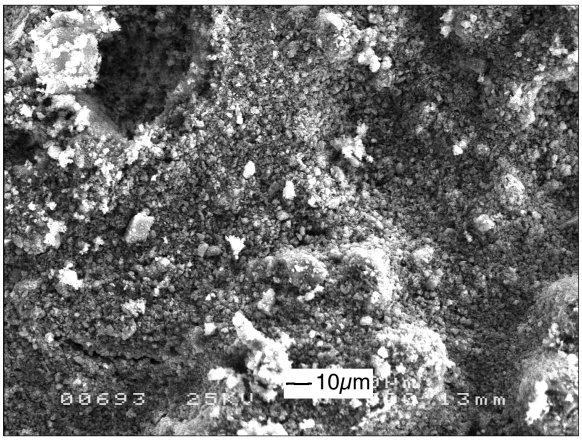

Fig. 1. SEM of exterior of oolitic mortar x500 [2]

When comparing this with Figure 2 which is air lime mortar made using sand (Lawrence, 2006) [2], there are clear differences.

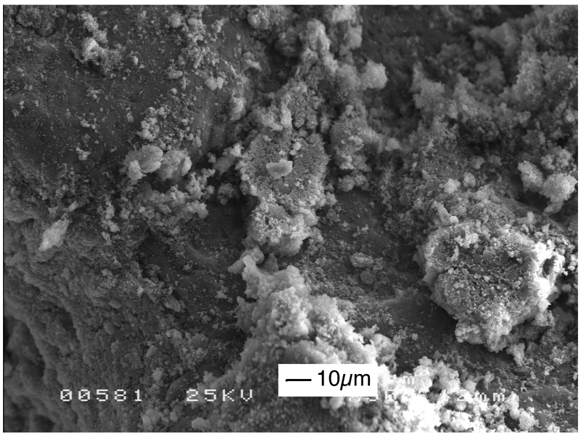

Fig. 2. SEM of exterior of sand mortar x500 [2]

Consequently, it is expected that SEM will be able to highlight differences between the use of calcitic and silicate aggregate at the B/Ag interface.

In the case of air lime mortar there are no hydraulic reactions taking place, due to the absence of silica, therefore other interactions must be taken into consideration.

As previously mentioned, a link has been suggested between the roughness and porosity of the aggregate surface, and the aggregate's cohesion with the mortar. SEM would be a good technique to use in order to examine the interface of aggregate and binder matrix, with the aim of comparing the calcitic aggregate/binder interface and the silicate aggregate/binder interface.

Use of SEM would enable verification of the extent of chemical reaction at the aggregate surface, in addition to being able to establish whether a significant amount of binder has entered the pores of the aggregate.

4.3.2 X-ray diffraction (XRD)

XRD is frequently used to determine the mineralogical phases that occur during the carbonation process (Lanas and Alvarez, 2003), (Arandigoyen and Alvarez, 2006) and (Tuncoku and Caner-Saltik, 2006) [1,26-27]. Additionally, the morphology and crystallinity can be established (Hansen et al., 2000) [28].

Lawrence et al. (2007) [18] noted that carbonation can be compared as a result of a obtaining a semi-quantitative relationship between the intensity of the peaks of calcium carbonate and calcium hydroxide.

4.3.3 Phenolphthalein

Phenolphthalein staining is a useful technique in assessing the depth of carbonation in a mortar sample (Lawrence, 2006) [2]. As previously mentioned, carbonation is of utmost importance in air lime mortars.

When used together with XRD, the two techniques provide detailed information about the carbonation process, which can be used to determine the extent of the difference in carbonation between mortars with calcitic and with siliceous aggregates.

4.3.4 Thermogravimetry (TG)/differential thermal analysis (DTA)

TG is another method that can be used to establish the mineralogical composition of lime (Arandigoyen et al., 2005) [29]. Marquez et al. (2006) [30] claim that DTA/TGA is very reliable for the identification of compounds in aerial lime mortar and suggest that for hydraulic-based mortars, it is less accurate due to the increased complexity of the material.

5 Conclusions

It is clear from the existing research that there is a gap in knowledge about the effect of aggregate type on the properties of air lime mortars.

While it has been found that aggregate type can affect strength, drying shrinkage and modulus of elasticity, little is known about the mechanisms behind these findings.

The author plans further research, seeking to establish the mechanisms behind this, by firstly undertaking a number of tests on the properties of the mortar. The calcareous aggregate will be compared with a siliceous aggregate; several B/Ag ratios will be used, in addition to a number of different curing times. Subsequently, microstructural analyses will be conducted, in order to establish the differences between aggregate type (calcitic and siliceous), that may have influenced the properties of the mortar. Particular focus is on the strength of the mortar.

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