

Use of supplementary aggregates in mortars produced using a novel lime drying technique

STARINIERI, Vincenzo <<http://orcid.org/0000-0002-7556-0702>>, ILLINGWORTH, James M and HUGHES, David C <<http://orcid.org/0000-0002-4922-4086>>

Available from Sheffield Hallam University Research Archive (SHURA) at:
<http://shura.shu.ac.uk/16992/>

This document is the author deposited version. You are advised to consult the publisher's version if you wish to cite from it.

Published version

STARINIERI, Vincenzo, ILLINGWORTH, James M and HUGHES, David C (2019). Use of supplementary aggregates in mortars produced using a novel lime drying technique. *Proceedings of the Institution of Civil Engineers - Construction Materials*, 172 (6), 305-313.

Copyright and re-use policy

See <http://shura.shu.ac.uk/information.html>

13/09/2017

**Use of supplementary aggregates in mortars produced using a novel lime drying
technique**

Vincenzo Starinieri*, PhD

Research Fellow, Materials and Engineering Research Institute, Sheffield Hallam
University, Sheffield, United Kingdom, Orcid no. 0000-0002-7556-0702

James M Illingworth, PhD

Research Assistant, Bradford Centre for Sustainable Environments, University of
Bradford, Bradford, United Kingdom

David C Hughes, PhD

Emeritus Professor of Construction Materials, Bradford Centre for Sustainable
Environments, University of Bradford, Bradford, United Kingdom, Orcid no. 0000-0002-
4922-4086.

* Corresponding author at Materials and Engineering Research Institute,
Sheffield Hallam University, City Campus, Howard Street, Sheffield S1 1WB, UK.
Tel.: +44 (114) 225 3712; Email: v.starinieri@shu.ac.uk

Words: 4,539; Figures: 7; Tables: 5

Abstract

Formulated Lime mortars suitable for long-term storage in silos or bags can be produced by adding quicklime to wet, as-received quarried sand. However, sands with high water content may require the addition of so much quicklime that would alter the mortar

proportioning. This work investigates the possibility to replace part of the lime-dried sand with dry crushed mixed glass cullet and calcium carbonate aggregate to allow greater control over mortar formulations. It is shown that the use of glass yields a similar or slightly weaker product, depending upon curing regime, than the control whilst calcium carbonate generates the strongest mortar.

Keywords: Granular materials, Materials technology, Sustainability

1 Introduction

In order to produce mortars suitable for long-term storage in silos prior to use, dry sand is required to prevent hydration reactions in the binder phase. Whilst this may be achieved using conventional kilns the process is energy intensive. An alternative technique has been developed in which quicklime is added to the as-delivered sand which removes the free water by a combination of quicklime slaking and evaporation resulting from the associated exothermic reaction. The resulting slaked lime forms part of the binder phase in a Formulated Lime mortar.

The process has been validated and the principal process-control factors have been identified as lime addition based upon a ratio of the stoichiometric requirements for complete slaking of the quicklime, free moisture content of the sand and mixing time of the combined sand and quicklime blend followed by a 24 hour “rest period” (Hughes and Illingworth, 2015). Formulated Lime mortars have been successfully produced with M1 and M5 classifications (Hughes et al., 2015). BS EN 459-1 (BSI, 2010) specifies this product as being a lime with hydraulic properties and comprising lime and added

material with hydraulic and/or pozzolanic properties; in the current mortar the hydraulic phase is Ground Granulated Blast-Furnace Slag (ggbS). The strength classes M1 and M5 (compressive strengths of 1 MPa and 5 MPa respectively at an age of 28 days) are specified in BS EN 998-2 (BSI, 2016). The principal factors which control mechanical properties are the ratio of ggbS to slaked lime produced during the drying process, the binder:sand ratio and the water:binder ratio.

During the mortar study limitations were identified to the maximum moisture content of the sand without compromising mortar proportioning or the critical process parameter of mixing time during the initial lime-drying process. This is best explained by reference to Fig 1.

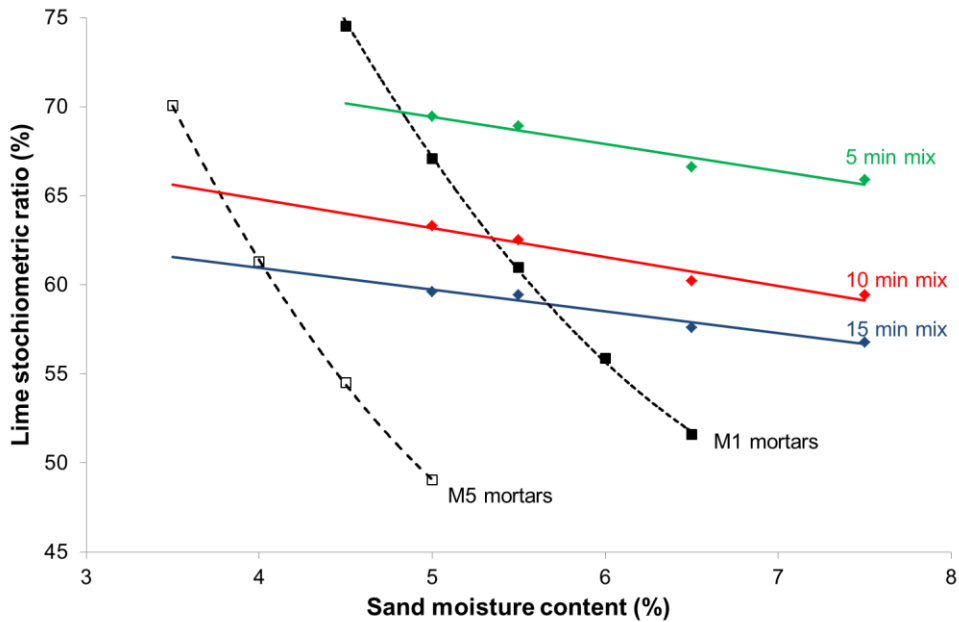


Figure 1 Example of mix design for optimum mortars at sand:binder ratio of 2.25:1. CH = Calcium Hydroxide.

Since the drying process combines both chemical combination of the free water and its evaporation it is not necessary to include the stoichiometric amount of quicklime to fully

combine with the free moisture within the sand (i.e. 3.113 g of Calcium Oxide being required to combine with 1 g of water). Thus, the term “stoichiometric ratio” has been adopted to express the amount of quicklime required (e.g. 50% stoichiometric ratio refers to 1.556 g of Calcium Oxide for each 1 g of free water). Fig. 1 shows that the optimum stoichiometric ratio for quicklime addition to dry the sand to achieve a specified mortar designation (ggb/slaked lime ratios of 0.2 and 1.0 for M1 and M5 respectively) decreases with increases in either the sand moisture content or the mixing time of the wet sand and quicklime – a complete description of its derivation may be found in Hughes and Illingworth (2015). At a given binder:sand ratio a sand with a high water content as delivered may require the addition of so much quicklime that the required ggb content cannot be incorporated without altering the mortar proportioning to yield high binder contents. Whilst such high water content sands could be used, the period of mixing during the drying process would have to be extended to such an extent that it would exceed acceptable commercial parameters, in this case specified as 15 minutes. Thus, the maximum free water content of the as-delivered sand to produce an M5 mortar is only some 5%.

One solution is to include a portion of dry sand as a partial replacement for wet sand to reduce the overall quicklime requirement which would permit greater variation in the moisture content of the as-received quarried aggregates. The key to this approach is to control the amount of slaked lime produced during the drying process so that sufficient ggb can be included whilst maintain the specified ratio of sand to binder. The addition of a dry supplementary aggregate to the material thus allows a greater addition of hydraulic material and, hence, greater control over mortar formulations produced by the lime drying process. Furthermore, the use of dry recycled materials such as crushed glass cullet is in keeping with the ‘sustainable’ credentials of these mortars.

This paper describes the inclusion of crushed glass cullet and calcium carbonate as supplementary aggregates. Its aim is to scientifically validate and specify the use of these materials in lime-dried mortars with a view to provide mortar specifiers with a greater control over mortar formulations produced by the lime-drying process.

2 Materials and Methods

2.1 Materials

The sand used for mortar production was a siliceous sharp sand which was oven dried to constant weight at 110°C prior to use. The supplementary aggregates were crushed mixed glass cullet supplied by Ecominerals Ltd and Minfil LG12 calcium carbonate aggregate supplied by Omya Ltd; these were used as a partial replacement of the lime-dried sand. The size distributions of all aggregates are shown in Figure 2.

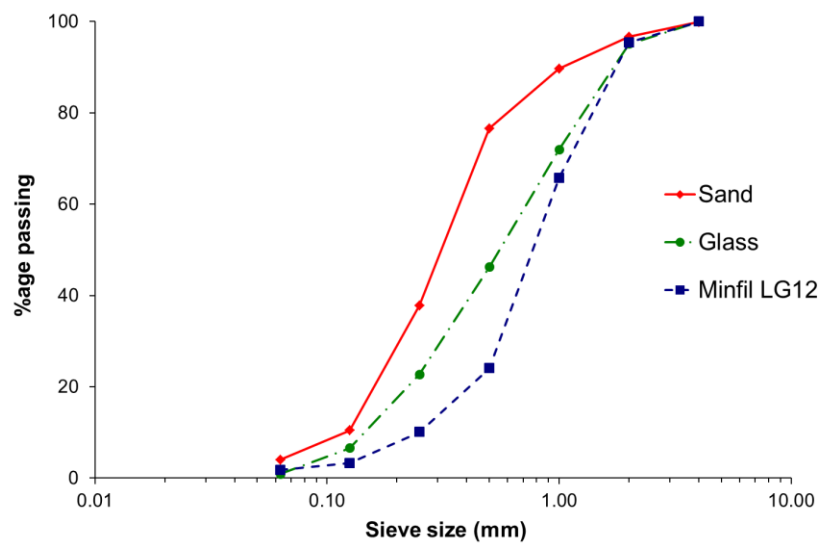


Figure 2 Grading of aggregates.

GGBS was supplied by Civil & Marine Slag Cement from the Frodingham plant, UK. Quicklime E (Hughes and Illingworth, 2015) was used for the preparation of the lime-dried mortars. Its chemical and physical properties, together with properties of the slaked limes, may be found elsewhere (Hughes and Illingworth, 2015); Table 1 summarises the key properties of the slaked lime.

Lime	E
Free lime (% CaO)	68.7
Insolubles (wt%)	4.8
CaO (wt%)	0
Ca(OH) ₂ (wt%)	90.8
CaCO ₃ (wt%)	6.9
A _{BET} (m ² /g)	18.16
V _{TOT} (cm ³ /g)	0.109
Bulk Density (kg/m ³)	559

Table 1: Properties of slaked lime from quicklime E.

2.2 Mortar Production

2.2.1 Mix design of lime-dried mortars

The mix design process has been detailed elsewhere and it is apparent that as the mortar specification is raised from M1 to M5 the maximum permissible moisture content of the sand is reduced (Hughes et al., 2015). Thus, only M5 mortars, requiring a ggbs:slaked lime ratio of 1.0 (Hughes and Illingworth, 2009) have been produced in this part of the study and are detailed in Table 2. In accordance with the terminology

previously described (Hughes and Illingworth, 2015) all mortars are considered optimally dried. Account must be taken of the observation that whilst high water contents in the feed aggregate require higher addition of quicklime this is accompanied by a lower stoichiometric ratio in order to satisfactorily dry the aggregate (see Fig 5 of Hughes and Illingworth, 2015). The approach taken was to fix the proportion of the total sand content comprising the supplementary aggregate (in this case 27%) the permissible control parameters of stoichiometric ratio and sand free moisture content may be determined to produce an M5 mortar of sand:binder ratio 2.25:1 utilising a 15 minute mixing time during the lime-drying process.

Code	Stoich ratio	Sand m/c (%)	Supplementary aggregate		w/b ratio	
	(%)		Type	% vol	“Fresh”	“Stored”
M5-1	60.9	4.03	-	-	1.03	1.01
M5-2	58.87	5.69	Glass	27	1.01	1.00
M5-3	58.87	5.69	CaCO ₃	27	0.99	0.98

Table 2: Mix design for M5 mortars.

2.2.2 Preparation of lime-dried mortars

Ten kg batches of sand were prepared containing known amounts of free water (wt.% basis) as specified in Table 2. A fundamental feature of the lime-drying concept is that water which is not contained within the pore structure of the sand, i.e. free water, rather than the total water content is the one which must be controlled. In this scenario the datum state of the aggregate is its saturated surface dry (SSD) condition in which the sand pores are completely filled whilst the free moisture is zero.

The wet sand was then mixed in a Hobart mixer with appropriate quantities of quicklime for the specified period of time of 15 min. Following mixing, the lime-dried material was stored in sealed containers for 24 hours to ensure complete hydration of the quicklime; this step was included after previous observations indicated the presence of free water in the blend at the completion of the initial mixing (Hughes and Illingworth, 2015). The required quantity of ggbs and supplementary aggregate were then added in order to provide the desired mix proportions. At this stage, the dry mortar mix was divided into two equal portions. One portion was stored in a sealed plastic container and retained under laboratory conditions (20°C, 50% RH) for 10 weeks; the other portion of the mix was prepared immediately. These mortars have been termed “stored” and “fresh” respectively.

2.2.3 Production of plastic mortars

All plastic mortars were produced in a Hobart mixer with a mixing time of 15 minutes. Appropriate quantities of water were added to give a flow table value (BSI, 1999a) of 170mm (+/- 5mm). This value was considered to provide materials with workability consistent with that observed in typical construction practice (Zhou et al., 2008). The required free water:binder ratio to achieve the specified flow value is included in Table 2.

2.2.4 Curing of mortars

Following preparation, the plastic mortars were cast into steel moulds of the required geometries (see section 2.3). The samples were initially cured within the moulds for 3 days at 20°C and covered by a polythene sheet to minimise evaporation. The moulds

were then stripped and the samples cured for a further 4 days under the same conditions. Unless stated otherwise, extended curing was then carried out at 20°C (\pm 2°C) and 65% RH (\pm 5%), hereafter referred to as air curing. The level of carbon dioxide in the curing room was estimated to be the standard atmospheric concentration (0.033 \pm 0.001% by volume). For comparison, some samples (for strength testing only) were subject to extended curing under water, also at 20°C.

2.3 Analytical Methodology

2.3.1 Compressive and Flexural Strength

The test specimens were prepared in steel moulds of 40 x 40 x 160 mm dimensions. Mortar was placed in 2 layers and vibration compacted. The three-point flexural and compressive strengths of the hardened mortars were determined using an Instron 4206. Crosshead speeds of 0.5 mm/min and 1 mm/min were used for flexural and compressive testing respectively. Measurement of compressive strength was carried out at 7, 28, 91 and 365 days. These testing ages were selected to comply with BS EN 196-1 (BSI, 2005) (7 and 28 days) and to investigate the strength development of the mortars at later ages (91 and 365 days). The compression tests were carried out on beam halves obtained from the flexural tests. For air-cured samples, flexural strengths were determined at 7 and 91 days whereas water-cured materials were tested at 28 and 91 days. A 7 day test was only required once since all samples were subject to the same curing regime in the first 7 days (see section 2.2.4).

2.3.2 Density and Porosity

The bulk density and total open porosity of the hardened mortars were assessed in accordance with BS EN 1015-10 (BSI, 1999b) and BS EN 1936 (vacuum saturation method) (BSI, 2006) respectively. The reported values are the mean of three individual samples.

2.3.3 Sorptivity

The water sorptivity of the hardened mortars was determined according to the methodology described in detail by Hall (1989). The test specimens were prepared in 70 mm steel cubes and cured as described in section 2.2.4. Prior to testing, the samples were conditioned at 60°C to constant weight in an atmosphere circulated over silica gel and soda lime. The sorptivity was measured through the moulded bottom surface. The reported values represent the mean of three individual samples.

2.3.4 Water Vapour Permeability

The water vapour permeability of the hardened mortars was determined using a modified version of the methodology described in BS EN 1015-19 (BSI, 1999c). The test specimens were cast in circular steel moulds, producing specimens of 180 mm diameter and 20 mm thickness. The mortar discs were then subjected to the curing regime specified in section 2.2.4. After curing, the samples were prepared for evaluation at 60°C in an atmosphere circulated over silica gel and soda lime. The samples were then placed in stainless steel test cups containing a saturated solution of potassium nitrate and sealed in-situ with molten paraffin wax. The air gap between the base of the samples and the solution was 12 mm (± 2 mm). The KNO_3 solution generates a relative humidity within the air gap of 93.2% at 20°C. The samples were then placed in a fan-

assisted storage chamber maintained at 20°C ($\pm 2^\circ\text{C}$) and 50% RH ($\pm 5\%$) and weighed every 24 hours until steady state vapour transmission was achieved. The reported values represent the mean of three individual samples.

2.3.5 Mercury Intrusion Porosimetry (MIP)

The pore size distributions of the mortars were determined using MIP at an age of 91 days (air cured). The measurements were carried out with a Micromeritics 9400 series instrument over a range of pressures between 3.9×10^{-3} MPa and 410 MPa. Assuming a contact angle of 140° and a mercury surface tension of 484×10^{-3} N/m, pore diameters ranging from 375 μm to 3.8×10^{-3} μm were characterized. Specimens with an approximate volume of 2 cm^3 were cut from the mortar prisms using a low speed saw. Prior to analysis, the samples were dried to constant weight at 60°C in an atmosphere circulated over silica gel and soda lime.

3. Properties of “fresh” mortars

3.1 Pore structure

Values of the total open porosity and dry bulk density are shown in Table 3. The tabled data is the average of three determinations. Following water-curing all 3 mortars display a similar trend of gradual reduction in open porosity with age. In contrast, air-curing produces little or no change in the control mortar and that containing glass cullet (M5-1 and M5-2 respectively) whilst the use of calcium carbonate yields a small reduction with age such that at ages of 91 and 365 days the porosity is independent of curing regime.

This suggests that the influence of the calcium carbonate is principally on the carbonation reactions.

Mortar	Open Porosity (%)			Dry Bulk Density (kg/m ³)		
	28 d	91 d	365 d	28 d	91 d	365 d
M5-1	29.3	29.3	29.1	1767	1805	1805
	<i>29.9</i>	<i>28.5</i>	<i>27.8</i>	<i>1733</i>	<i>1746</i>	<i>1779</i>
M5-2	28.6	28.5	28.6	1765	1753	1769
	<i>29.6</i>	<i>28.0</i>	<i>27.7</i>	<i>1732</i>	<i>1739</i>	<i>1768</i>
M5-3	28.3	27.9	27.6	1804	1826	1834
	<i>29.5</i>	<i>28.0</i>	<i>27.4</i>	<i>1772</i>	<i>1788</i>	<i>1809</i>

Table 3: Porosity and density of mortars at ages of 28, 91 and 365 days (air curing in Normal font, water curing in Italics).

The pore size distributions of the mortars (91 days of air curing) are shown in Figure 3. The bulk of the porosity is made up of pores in the region of 0.05 μm diameter. However, the principal difference between the use of glass and calcium carbonate is that the latter yields a lower volume in this range with a greater volume of the finest pores in the range 0.005 – 0.01 μm .

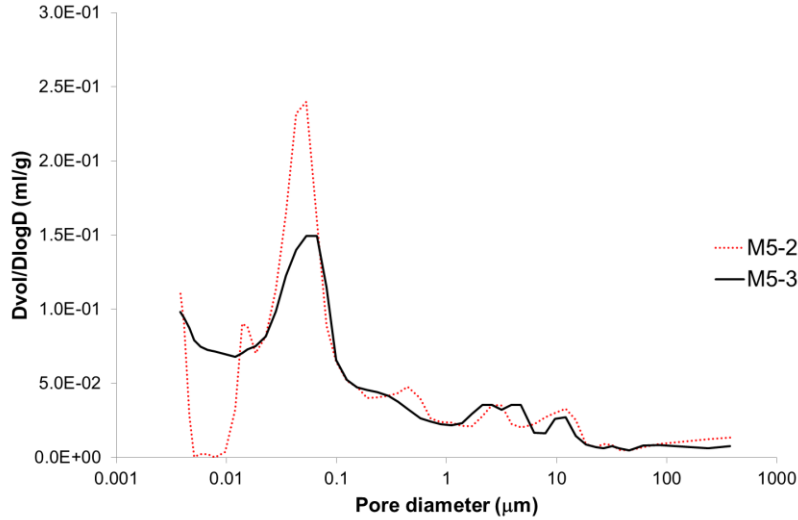


Figure 3 Pore size distribution of M5 mortars with supplementary aggregates after 91 days of air-curing.

3.2 Compressive and flexural strength

The data in Figures 4a and 4b show the evolution of flexural and compressive strength respectively. It is apparent that there are subtle differences in the performance of the carbonate (M5-3) and glass mortars (M5-2) when compared to the control mortar (M5-1).

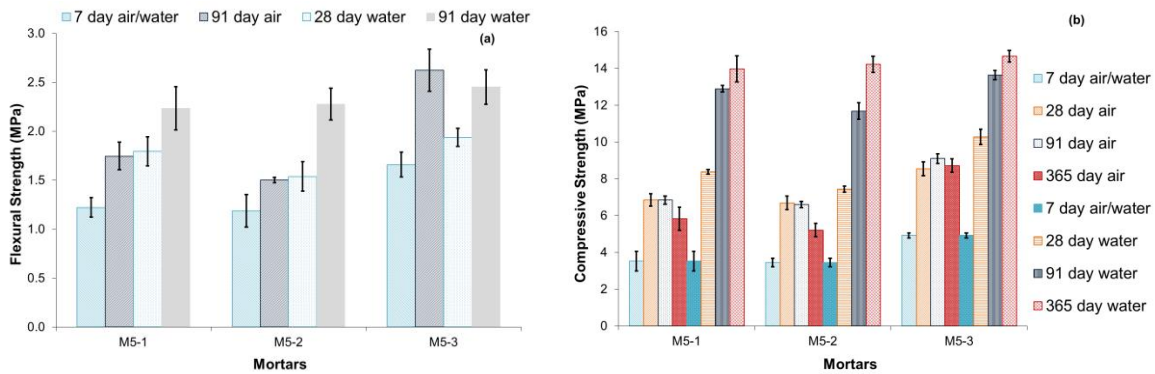


Figure 4 Strengths of M5 mortars – (a) flexural strength, (b) compressive strength.

Under conditions of air curing the carbonate mortar is stronger than the control at all ages in both flexure and compression. In contrast, the glass mortars are similar to the controls. All significance is tested at 95% confidence levels. Previous work (Hughes et al., 2015) has shown a degradation in strength of lime-dried Formulated Lime mortars when air cured, which was explained by the high shrinkage and micro-cracking associated with calcium hydroxide activated slag binder systems. Carbonation may be an additional mechanism for strength loss. Bernal et al (2010) observed strength reductions in sodium-based activated slag systems subject to accelerated carbonation conditions; however, later they concluded that such accelerated experimental conditions may be “unduly aggressive and test results must be cautiously interpreted” (Bernal et al., 2012).

It can be seen in Fig 4b that the previously observed strength loss phenomenon has occurred in the glass mortar whilst the inclusion of calcium carbonate has moderated any degradation and yields no strength change between 28 and 365 days. Similar strength degradation has been reported in air-cured calcium hydroxide activated slag mortars in which the alkali activator content was a relatively modest 7.5%. In this case the strength was stabilised by the inclusion of auxiliary activators of either sodium silicate or sodium carbonate (Yang et al., 2012) and barium hydroxide (Yang et al., 2010). Such a solution should be explored for the current Formulated Lime mortars.

Following water curing a different relationship is observed. In all mortars there is a continuous increase in strength up to an age of 365 days. There is no influence of the supplementary aggregate on flexural strength. However, whilst the compressive strength of all mortars is the same after 365 days, the glass mortar is slightly weaker than the

control mortar at 28 and 91 days whilst the carbonate mortar is stronger at the same ages.

Water curing yields higher strengths than air curing with enhancement being in the range 70 – 170% at 365 days. The lowest enhancement was observed in the mortar with the supplementary carbonate aggregate which is in line with the earlier observation of its influence on porosity (see section 3.1).

3.3 Moisture transport

The breathability of mortars is an important property, particularly where used in restoration works on historic buildings; see section 3.3 of Hughes et al. (2015) for more discussion of this topic. In the current work, breathability was assessed by sorptivity and water vapour permeability (WVP) measurements following air curing and the results are presented in Figures 5 and 6 respectively.

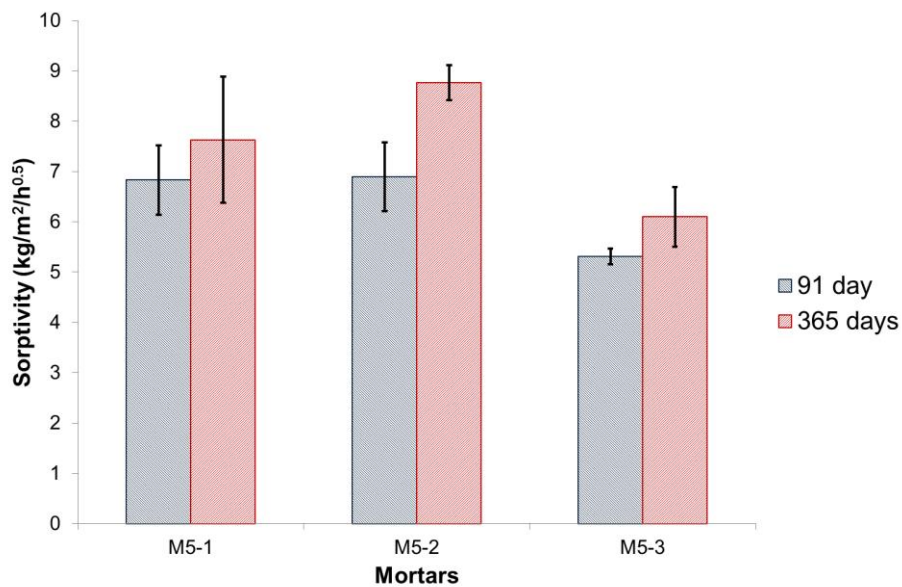


Figure 5 Sorptivity of M5 mortars.

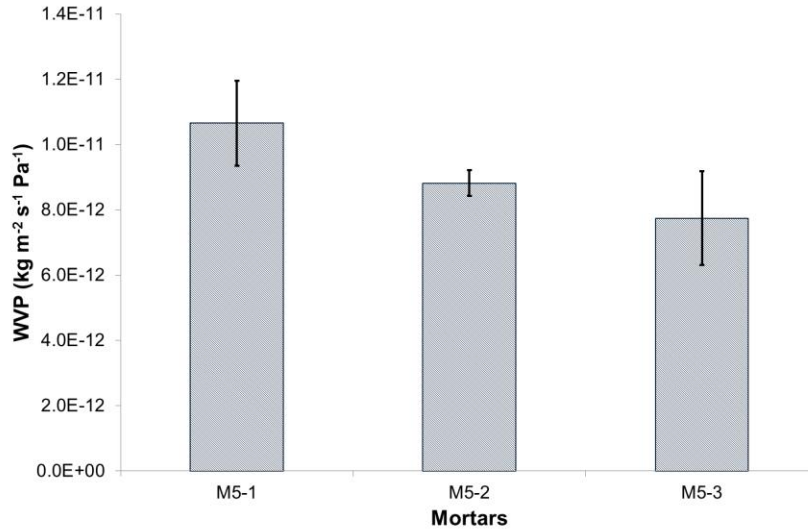


Figure 6 Water Vapour Permeability of M5 mortars after 91 days.

At an age of 91 days there is no significant difference in sorptivity and WVP of the glass mortar when compared to the control mortar; the same holds for the WVP of the carbonate mortar. However, the sorptivity of the carbonate mortar is lower than that of the control as would be expected from the pore structure. After 365 days the sorptivity of the mortars with supplementary aggregates is not significantly different from the control mortar; however, the sorptivity of the carbonate mortar is lower than that of the glass mortar. Unlike the control and carbonate mortars, the increase in sorptivity shown by the glass mortar between 91 and 365 days is significant.

3.4 Discussion

The data show that the inclusion of supplementary aggregates may influence the strength development of the lime-dried mortars. The inclusion of calcium carbonate aggregate (mortar M5-3) leads to a significant increase in strength in comparison with the control mortar with the exception of mortars water cured for 365 days. This is

represented in Table 4 with the strength of the carbonate mortar being expressed as a percentage of the control mortar at the same age. The strength enhancement is higher than that observed by Rashad (2015) in sodium sulfate-activated slag in which 5% limestone dust was used as a replacement for the slag. In contrast, the inclusion of recycled glass cullet produces similar results to mixes without a supplementary aggregate. It might have been expected that a pozzolanic benefit would have been observed; however, the grading of the glass is relatively coarse with few fines (Fig 2).

Curing	Age			
	7 d	28 d	91 d	365 d
Water	139%	123%	106%	105%
Air	139%	125%	134%	150%

Table 4: Strength of M5-3 mortars expressed as a percentage of their M5-1 counterparts.

The carbonate aggregate results in a slight reduction in the water demand of the plastic mortar (Table 2), presumably as a consequence of the coarser grading of this particular aggregate. However, it seems unlikely that this effect can be solely responsible for the relatively large strength differences observed; it should be noted that some of the increase in relative strengths at later ages is a consequence of the strength reductions in the air-cured control mortars (see Fig 4b). Other workers have also reported enhanced strength development in lime-based mortars produced using limestone aggregates. For example, Lanas *et al.* (2004) demonstrated that limestone aggregates generate higher strengths than siliceous aggregates in otherwise identical mortar formulations. It was suggested that 'the similarity between a calcitic binder matrix and a limestone aggregate structure could be related to this increase in strength and, during portlandite carbonation,

the calcite of the aggregate provides nucleating sites for crystal growth'. The data in Figure 4 appears to offer some support to this view as the strength differential between water and air-cured samples is lower for the M5-3 mortar, the carbonation process being absent during water curing. Scannel *et al.* (2014) found slightly higher depths of carbonation in mortars made with carbonate sand than in identical mortars made with silica sand. It was suggested that the higher porosity of carbonate sand allows a quicker rate of carbon dioxide diffusion, thus leading to a quicker rate of carbonation.

Studies of the use of calcium carbonate as an addition to Portland cement (Matschei *et al.*, 2007) show that fine calcium carbonate can be both a reactive component as well as a filler. Ingram and Daugherty (1991) reported that the chemical influence of calcium carbonate is a function of the tricalcium aluminate content of the cement, being more apparent at higher tricalcium aluminate levels. In contrast, there is little chemical influence in an alkali activated slag (sodium based activation) which was explained by an insufficient supply of calcium and/or aluminate (Gao *et al.*, 2015); the extent to which this may apply to the Formulated Lime mortars is currently conjecture. Many authors state that the fineness of the limestone is an important parameter (e.g. Rakhimova, 2016). However, in the current programme the calcite supplementary aggregate is relatively coarse with little fines content (Fig 2). That said, there is some evidence of a small "filler effect" as seen by the higher density of mortar M5-3 at even the earliest age of 28 days (Table 3). Further, whilst the addition of 1-3% fine calcite has been found to yield a small increase in the early age strength of an alkali-activated cement, no or even a negative impact occurred at later ages (Wang *et al.*, 1994). Little modern research has been undertaken on the lime-ggbs system and certainly not in the low slag contents currently being considered and further fundamental research is required to examine the role of calcium carbonate within this system.

4 The effect of storage on lime-dried mortar mixes

The effect of storage on strength development of either bagged or silo-based mortars has received little attention. Whilst the use of kiln-dried sands should minimise the potential for degradation during storage, the lime-drying technique raises this potential as the presence of any residual water which has neither been chemically combined nor evaporated may be a factor. Mortars were produced from material which had been stored for 10 weeks after the application of the lime-drying process (see 2.2.2). It is apparent that the “stored” mortars required a slightly lower w/b ratio to achieve the desired flow (see Table 2).

Figure 7 shows the compressive strength development of the M5 series of “stored” mortars (both water and air-cured) in comparison to the “freshly” prepared materials described in section 3.2; the “fresh” data is a duplication of the strengths in Fig 4b and are included here for ease of comparison. The “stored” mortars show the same strength development profiles as those of their “fresh” counterparts, i.e. a strength decline at 365 days for air-cured mortars whilst water curing yields a continuous increase. However, subtle differences are apparent. Under air curing the strengths of the “fresh” and “stored” mortars are similar up to and including 91 days reflecting the similar w/b ratios. The principal effect of storage is an increase in the strength loss between 91 and 365 days for the air-cured mortars with the largest loss being observed in the M5-3 mortar. In contrast, water curing yields “stored” mortars of similar or higher strength than the comparable “fresh” mortar.

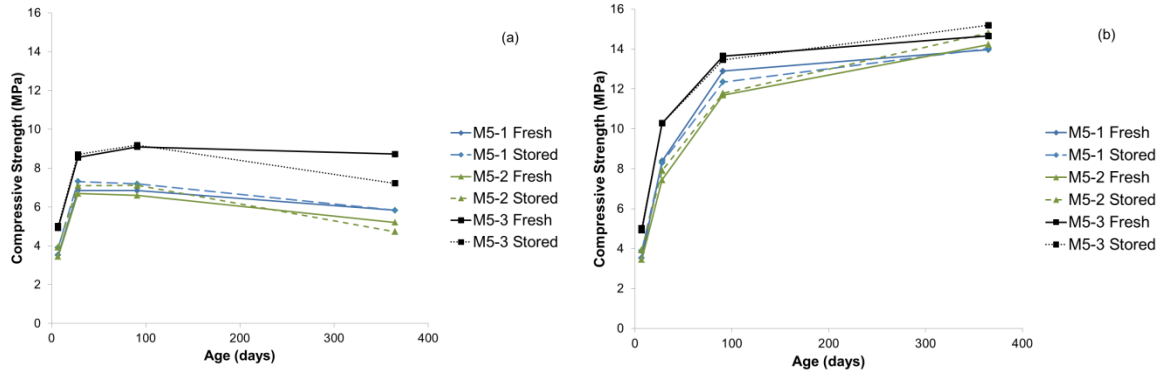


Figure 7 Compressive strengths of stored and fresh mortars; (a) air cured, (b) water cured.

Sample	H ₂ O (wt.%)	A _{BET} (m ² /g)	% Ca(OH) ₂ (as CaO)	% CaCO ₃ (as CaO)	% CaO
Fresh	5.1	23.7	89.5	5.8	1.1
Stored 8wk	1.7	20.3	89.0	6.5	0.9
Stored 12wk	1.2	16.0	88.4	6.9	1.0

Table 5: Influence of storage on the lime

Table 5 is an extract from Table 9 of Hughes and Illingworth (2015) and shows the characteristics of the lime following storage of a lime-dried silica sand for various periods of time. The small differences between the “fresh” lime in Table 5 and the lime cited in Table 1 may be accounted for by the lack of the 24 hour rest period and the use of a higher free moisture content in the sand for the latter lime sample. The Table shows that there is a small carbonation of the hydrated lime accompanied by a reduction in the BET (Brunauer–Emmett–Teller) surface area. It is not known how representative this data is of lime-drying of a carbonate sand and, hence, of any differences between the storage of M5-3 mortar and that of M5-1 and M5-2. There is the possibility that the M5-3 mortar contained more carbonated lime than the other mortars. It has been shown (Adams and Race, 1990; Palm et al., 2016; Baron and Douvre, 1987; Ramachandran and Zhang,

1986) that the addition of limestone in an hydraulic cement can increase the amount of drying shrinkage and should this phenomenon hold for the Formulated Lime then this could contribute to the additional strength loss of the M5-3 mortar.

The principal conclusion is that the use of glass and calcium carbonate as supplementary aggregates in stored mortars is not detrimental to mortar strength. Indeed, the use of calcium carbonate is beneficial.

5 Conclusions and significance for practical mortars

This work has shown that dry supplementary aggregates in the form of crushed glass cullet or calcium carbonate can be used as partial replacement of the lime-dried sand to allow greater control over mortar formulations produced by the lime-drying process.

It has been shown that:

- Carbonate aggregate yields a lower volume of pores in the region of 0.05 μm and a greater volume of the finest pores in the range 0.005 - 0.01 μm in "fresh" mortars cured in air for 91 days.
- The inclusion of calcium carbonate aggregate leads to a significant increase in strength in air cured mortars in comparison to the other mortars within the M5 series, whereas the inclusion of recycled glass cullet produces similar results to mixes without a supplementary aggregate. In contrast, water curing yields mortars of similar strength at an age of 365 days but with slightly different strength development profiles at earlier ages. Thus, the influence of the calcium carbonate is principally on the carbonation reactions.
- In "fresh" mortars the inclusion of calcium carbonate moderates the degradation in compressive strength which was observed between 91- 365 days of air curing.

- The use of supplementary aggregates does not affect negatively the breathability (assessed by sorptivity and wvp) of "fresh" lime dried mortars.
- Under air curing the strengths of the "fresh" and "stored" mortars are similar up to and including 91 days. The principal effect of storage is an increase in the strength loss between 91 and 365 days for the air-cured mortars with the largest loss being observed in the M5-3 mortar. In contrast, water curing yields "stored" mortars of similar or higher strength than the comparable "fresh" mortar.

Thus, lime-dried materials can be silo-stored prior to use without changes to the mortar classification and dry supplementary aggregates introduced for additional control over mortar formulations. The resulting mortars will combine the advantages of pre-blended dry silo mortars (i.e. accurate gravimetric batching of the mix components, reduced wastage, lower labour costs and cleaner, quieter construction practice) with low embodied energy deriving from the use of quicklime rather than the energy intensive kiln drying. The use of a processed waste product (glass cullet) is an additional benefit.

A practical application of the Formulated Lime mortars produced in this study is the bonding of masonry units. No research has been undertaken on the performance of lime-dried mortars in the bonding of masonry units and further research is required to examine the influence of supplementary aggregates on the mortar-masonry bond strength. Particularly, the water retention properties of the mortars, which affect the moisture flow from fresh mortar to brick and consequently the strength of the mortar-brick bond (Groot, 1993), will have to be investigated.

Acknowledgments

The authors are grateful for the funding provided by the Engineering and Physical Sciences Research Council (EP/D025036/1), Technology Strategy Board (Prog. No: 876) and Lime Technology Ltd and Castle Cement Ltd, Civil and Marine Ltd and Lhoist UK Ltd for the supply of materials. Thanks are also due to Dr. Mike Lawrence at the University of Bath for his assistance with the mercury porosimetry analyses.

References

Adams LD and Race RM (1990) Effect of limestone additions upon drying shrinkage of Portland cement mortar. In *Carbonate Additions to Cement* (Klieger P and Hooton R (eds)). ASTM International, West Conshohocken, USA, pp. 41-50.

Baron J and Douvre C (1987) Technical and Economical Aspects of the Use of Limestone Filter Additions in Cement. In *World Cement*, No. 3, vol. 18, pp. 100-104.

Bernal SA, de Guttierrez RM, Provis JL and Rose V (2010) Effect of silicate modulus and metakaolin incorporation on the carbonation of alkali silicate-activated slags. *Cement and Concrete Research* **40**: 898-907.

Bernal SA, Provis JL, Brice DG et al. (2012) Accelerated carbonation testing of alkali-activated binders significantly underestimates service life: The role of pore solution chemistry. *Cement and Concrete Research* **42**:1327-1326.

BSI (2005) EN 196-1:2005. Methods of testing cement - Part 1: Determination of strength. BSI, London, UK.

BSI (1999a) EN 1015-3:1999. Methods of test for mortar for masonry - Part 3: Determination of consistence of fresh mortar (by flow table). BSI, London, UK.

BSI (1999b) EN 1015-10:1999. Methods of test for mortar for masonry: Part 10: Determination of dry bulk density of hardened mortar. BSI, London, UK.

BSI (1999c) EN 1015-19:1999. Methods of test for mortar for masonry. Part 19: Determination of water vapour permeability of hardened rendering and plastering mortars. BSI, London, UK.

BSI (2006) EN1936:2006. Natural stone test methods – Determination of real density and apparent density, and of total and open porosity. BSI, London, UK.

BSI (2010) EN 459-1:2010. Building Lime, Part 1: definitions, specifications and conformity criteria. BSI, London, UK.

BSI (2016) EN 998-2:2016. Specification for mortar for masonry. Part 2: Masonry Mortar. BSI, London, UK.

Gao X, Yu QL and Brouwers HJH (2015) Properties of alkali activated slag-fly ash blends with limestone addition. *Cement and Concrete Composites* **59**:119-128.

Groot C (1993) Effects of water on mortar-brick bond, PhD Thesis, University of Delft, Delft, The Netherlands.

Hall C (1989) Water sorptivity of mortars and concretes: a review. Magazine of Concrete Research **41**:51-61.

Hughes DC and Illingworth JM (2009) Dry-silo mortars based on a novel lime-drying process. In *Proceedings of the 11th International Conference on Non-conventional Materials and Technologies* (Walker P, Ghavami K, Paine K, Heath A, Lawrence M and Fodde E (eds)). Bath, UK.

Hughes DC and Illingworth JM (2015) Low energy pre-blended mortars: Part 1 – control of the drying process using a lime drying technique. Construction and Building Materials **101**:466-473.

Hughes DC, Illingworth JM and Starinieri V (2015) Low Energy Pre-blended Mortars: Part 2 – Production and characterisation of mortars using a novel lime drying technique. Construction and Building Materials **101**:710-720.

Ingram KD and Daugherty KE (1991) A review of limestone additions to Portland cement and concrete. Cement and Concrete Composites **13**:165-170.

Lanas J, Perez-Bernal MA, Bell MA and Alvarez Galindo JI (2004) Mechanical Properties of hydraulic lime based mortars. Cement and Concrete Research **34**:2191-2201.

Matschei T, Lothenbach B and Glasser FP (2007) The role of calcium carbonate in cement hydration. Cement and Concrete Research **37**:551-558.

Palm S, Proske T, Rezvani M et al. (2016) Cements with a high limestone content – Mechanical properties, durability and ecological characteristics of the concrete. *Construction and Building Materials* **119**:308-318.

Rakhimova NR, Rakhimova RZ, Naumkina NL and Khuzin AF (2016) Influence of limestone content, fineness, and composition on the properties and microstructure of alkali-activated slag cement. *Cement and Concrete Composites* **72**:268-274.

Ramachandran VS. and Zhang C (1986) Influence of CaCO₃ on Hydration and Microstructural Characteristics of Tricalcium Silicate. In *il Cemento*, vol. 83, No. 3.

Rashad AM (2015) Influence of different additives on the properties of sodium sulfate activated slag. *Construction and Building Materials* **79**:379-389.

Scannell S, Lawrence M and Walker P (2014) Impact of aggregate type on air lime mortar properties. *Energy Procedia* **62**:81-90.

Wang S-D., Scrivener KL and Pratt PL (1994) Factors affecting the strength of alkali-activated slag. *Cement and Concrete Research* **24**:1033-1043.

Yang KH, Sim JI and Nam SH (2010) Enhancement of reactivity of calcium hydroxide-activated slag mortars by the addition of barium hydroxide. *Construction and Building Materials* **24**:241-251.

Yang HK, Cho AR, Song JK and Nam SH (2012) Hydration products and strength development of calcium hydroxide-based alkali-activated slag mortars. *Construction and Building Materials* **29**:410-419.

Zhou Z, Walker P and D'Ayala D (2008) Strength characteristics of hydraulic lime mortared brickwork. *Construction Materials* **161**:139-146.