Pore Structure as a Determinant of Flexibility in Sustainable Lime-Cement Mortar Composites

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Abstract — Flexibility of Lime Mortar resulting from its good permeability feature through its relatively large interconnected pore structure makes it popular as an old sustainable building material. This characteristic feature enhances its capability to suppress masonry deformation. However, its mechanical strength is weak. To address this shortcoming, cement is added to form a stronger composite. This study, therefore, evaluates the pore structure of the resulting lime-cement composite, with a view to assessing impact and sustainability of the lime’s flexibility in the overall performance of the composite. Adopting equal mix ratio (1:3) of Binder/Aggregate (B/A), mortars were prepared using lime as a lone binder, as well as other five different compositions in the ascending and descending order of lime and cement binders (i.e., 1:1, 1:2, 1:3, 2:1 and 3:1). Each composition was assessed in fresh state using Water/Binder ratio, Air Content and Bulk Density. For the hardened state assessments, mechanical characteristics and microstructural features evaluated through six (6) and twelve (12) month curing periods respectively, were considered. Results of the investigation reveal that low cumulative porosity (i.e., summation of the porosities) across the three pore regions of Inter-Nano pores (1 – 10 nm), Super-Nano pores (10 – 100 nm) and Sub-Micro pores (0.1 – 1 µm), for each of the mortars, is observed to be synonymous with low cement content, low Modulus of Elasticity, low (f/f0), and thus, high flexibility. This observation is supported by high linear coefficient of correlation (R2 = 0.89) for the equation describing both ‘cumulative porosity’ (within the referenced three pore ranges) and cement content. Thus, it could be inferred that despite cement addition with resulting alteration in the pore structure of the composite, presence of lime would retain flexibility property of the composites, with more flexibility in the descending order of cement contents. Considering the impacts of pandemic climate change, growing energy costs and human activities on the environment, this study therefore attempts to promote revival of an old but partially abandoned sustainable lime based mortars, with a view to facilitating improved lime mortar performances, conservation of energy resources and overall protection of the environment.

Key words — Climate Change, Flexibility, Lime Mortars, Pore Structure, Sustainability.

I. INTRODUCTION

Lime mortar is an old but partially abandoned building material. It is obtained from mixing lime, sand and water together in a proportional ratio. It is a flexible and slow to harden material, thereby allowing minute movements within the mortar joints, without impacting negatively on the masonry’s structural stability [1]-[4]. Its flexibility feature helps to cushion masonry joints, accommodate strains, inhibit cracking, thereby resulting in good flexural bond strengths [5], [6]. It is characterised with low elastic modulus, and therefore capable of more deformation on load application compared with Portland cement [7]. Because of its relatively large interconnected pore structures, it has excellent permeability features. As a building fabric, these pore structures are responsible for the mortar’s durability against environmental conditions as they accommodate growth of ice crystal within, in frost periods, without deteriorating the matrix [8], [9]. The permeability feature also reduces moisture entrapment within, a phenomenon known as “breathability” [8], [10]-[12]. Increased bond between mortar and substrate is facilitated through lime mortar’s enhanced workability resulting from its high water retention property [5].

While the manufacture of limes requires less energy and releases lower greenhouse gases (about 20% reduction, relative to Portland cement), most of the carbon dioxide released during its calcination is re-absorbed while exposed to the atmosphere as lime mortars, a process referred to as re-carbonation. This is in agreement with the current call for minimal energy consumption and reduced atmospheric CO2 concentration, with significant environmental usefulness [13]-[15]. Also, masonry laid with lime based mortar is characterised with lower bond strength (relative to cement) such that the masonry units can be prised off easily thereafter, thus encouraging material recycling, and hence, supporting overall sustainability. Additionally, building lime mortars finished structures do possess low thermal conductivity. This impacts positively on the interior surface temperatures of buildings and may therefore be considered for some insulating role [11], [16].

In general, lime mortar has several beneficial characteristics which may include good adhesion, ductility, useful porosity values [17] and therefore possesses greater water vapour permeability than Ordinary Portland Cement (OPC), which facilitates reduced moisture entrapment [18]. A lime binder facilitates good adhesion between surfaces, and ensures effective penetration into voids. Masonry joints would be cushioned to absorb strains, and therefore check cracking. It would constitute the primary route for moisture passage thereby making it permeable, to protect the overall structure. Thus, it acts sacrificially to protect the masonry units against the harmful impacts of salts and moisture [19]. However, in spite of these useful properties, lime mortar is known for long setting and hardening periods, low internal cohesion, high volumetric changes (i.e., shrinkage, particularly, aerial lime), relatively low mechanical strengths and a high water absorption capacity via capillarity. These have resulted in delayed project delivery periods leading to its relegation and relative disuse [20]-[22]. It is within this context that this experimental study has been carried out, to improve on lime mortar by replacing
specific quantities of lime with cement with a view to evolving a performance synergy between lime and cement. In this case, their advantageous features are maximised at the expense of their individual shortcomings. Adding cement to lime mortar increases the mortar’s strength. Alongside this is significant effect on the microstructural features of lime mortar as porosity, pore size distribution and vapour diffusion coefficient are progressively reduced (with increasing cement content) [23]. This may also lead to mortar’s increased modulus of elasticity considered unfit to accommodate masonry deformations [3], [21], [25]-[27]. This study, therefore, focuses on the evaluation of the pore structures of the resulting lime-cement composites, with a view to assessing sustainability of the lime’s flexibility on the overall performance of the evolving composite. This research has a potential to evolve a blended lime/cement mortar for improved mortar performance in new constructions, with attendant reduction on environmental impacts from construction industry.

II. EXPERIMENTAL PROCEDURE

A. Materials and Mortar Preparation

Considering material’s availability, relatively short setting time to attain optimal strength, low handling difficulties, and the need to maximise lime performance behaviour [28], [29], Natural Hydraulic Lime (NHL-5.0) was adopted for this study. This was obtained from St Astiers, UK. Also, Cement (CEM I 52.5) for this study was supplied by ‘Hanson Cement’, UK. Particle Size Distributions for both Lime and Cement obtained using Laser Diffraclometry Xmastersize (via laser diffraction method) are indicated in Table I. Also, Chemical compositions (by elements) for both lime and cement as determined by Energy Dispersive Spectrometry are given in Table I. Siliceous fine kiln dried sand obtained from ‘Fife Silica Sands’, UK, constituted the aggregate. In line with the British Standards [30] and ASTM C 136 [31], the aggregate was passed through a sieve analysis as the particle size distributions are as shown in Fig. 1. The sand had Particle Size Distribution of 0-2 mm (i.e., 0.05 mm < Ø < 2 mm), considered suitable, in accordance with ASTM C 33 [32].

Considering binder-aggregate (B/A) ratio of 1:3 (by volume) being the most prevalent in the literature [20], [23], [34], [35], preparation of mortars followed requirements of British Standards [33]. To ensure measurement precision during batching, volume proportions of components were converted to weights [6]. Using BS EN 1015-3 [33] and BS EN 1015-6 [36], workability of 145±5 mm was adopted for the mortars, to avoid stiffness or fluidity in accordance with the British Standards [37]. This was achieved through visual and physical assessments of the mixes during the trial experimentation.

As shown in Table III, using the adopted B/A ratio (i.e., 1:3), mortars were prepared with increasing/decreasing cement contents. Mixing was done in the laboratory mixer of 30 litres maximum capacity as explained in previous studies [4], [28] and [38]. Three prismatic specimens of 40×40×160 mm was prepared for each mortar mix. An average value for the three was considered for the specimens during evaluations (i.e., for microstructural analysis, and mechanical characteristics evaluations). The specimens were compacted with a vibration table after mould filling in prismatic casts [39], demolded 2 days later and allowed to cure at the laboratory ambient conditions of 21±4 °C (temperature) and 40±5% (relative humidity), until the test dates of 1, 3, 6, 9 and 12 months.

B. Analytical Methodology

Details of the microstructural characteristics evaluation (in respect of the total porosity, median pore diameter, bulk density and pore size distribution, using Mercury Intrusion Porosimetry (MIP) technique via AutoPore IV 9500 by Micrometrics (with pressure range up to 60000 psi)), mechanical properties assessment (using ELE AutoTest 2000 apparatus and INSTRON 3367) and some other relevant preliminary material testing (involving ‘Carl Zeiss EVO 50’ Scanning Electron Microscope and Laser Diffraclometry Xmastersize) are as documented in the previous studies [4], [28] and [38].
III. RESULTS AND DISCUSSION

A. Water/Binder (W/B) Ratio, Air Content and Bulk Density

7Results of the W/B ratios for lime, and lime-cement mortars batched using varying binders’ contents are shown in Fig. 2. A value of 1.53 was obtained for the reference mortar (lime mortar, L13), which indicates the ability of the mortar to retain water in the mix as it ensures plasticity and enhances workability [6], [41].

Adding cement content (to obtain the composites) reduces composites’ water demand (although non-linear). This may result in quicker formation of more hydration products of Calcium Silicate Hydrates and subsequent densification of the mortar matrix [42] [43]. Concerning the mortars’ Air Contents, the graph in Fig. 3 indicates air content value of 6.4% for lime mortar only.

With 25% addition of cement content to the mortar, air content value increased to 8.3%. Further increase in cement content resulted in subsequent reduction of the air content due to densification of the microstructure, a consequence of hydration processes. During the hydration, the volume of pores is gradually reduced by the newly created products of the reactions, leading to formation of more compact composites, hence the observed air content reduction. Also, for lime only, the bulk density was 1.98 g/ml (Fig. 4).

The bulk density increases as more cement content is added, (although not in a complete linear trend). This may arise from deposition of C–S–H gel and other hydration products that are usually characterised with increase in mass. Hence, the more the cement content, the higher the bulk density. This is in agreement with the trend observed for the air contents.

B. Microstructural Features

Microstructural data inclusive of Pore Size Distribution (PSD) curves for the six different composites at 1 and 6 months of curing are as shown in Figs. 5 (a) and (b), and Table IV.

As shown in Fig. 5 (a), a bimodal PSD curve with predominant sizes between (0.5-5 µm) and (5-20 µm) is observed for lime only at first month of curing. The mortar displayed both gel pores (i.e., 1 nm – 3 µm) and significant capillary pores (i.e., 3-30 µm) [44]. Because of high water-retaining nature of lime, and its associated long carbonation process, the observed large pores arise from evaporation of excess unbound kneading water. The pores cannot be dissociated from fine cracks usually resulting from drying shrinkage which is capable of expanding and widening [4],

| Cement Contents | Mortar Reference LD. (Slag Contents) | Volumetric Ratio (L:\C-\Sd) | Lime (L): Volume (Volume in 'ml'/ mass in 'g') | Cement (C): Volume (Volume in 'ml'/ mass in 'g') | Sand (Sd): Volume (Volume in 'ml'/ mass in 'g') |
|-----------------|-----------------------------------|-----------------------------|-----------------------------------------------|-----------------------------------------------|-----------------------------------------------|
| 0%              | L13 (0%)                          | 1-0-3                       | 1 (1700/ 1172)                                | 0 (0)                                         | 3 (4350/6444)                                |
| 25%             | LC13 (25%)                        | 3-1-12                      | 3 (770/528)                                  | 1 (415/442)                                  | 12 (4350/6444)                               |
| 33%             | LC21 (33%)                        | 2-1-9                       | 2 (665/458)                                  | 1 (540/574)                                  | 9 (4240/6283)                                |
| 50%             | LC11 (50%)                        | 1-1-6                       | 1 (850/886)                                  | 1 (830/883)                                  | 6 (4350/6444)                                |
| 66%             | LC12 (66%)                        | 1-2-9                       | 1 (510/352)                                  | 2 (995/1060)                                  | 9 (3920/5800)                                |
| 75%             | LC13 (75%)                        | 1-3-12                      | 1 (260/176)                                  | 3 (1250/1325)                                 | 12 (4350/6444)                               |
[28]. Despite continuous carbonation at 6-months, wider pore range is prevalent, showing a presence of more pores with larger sizes (between 0.5 µm and 40 µm) (Fig. 5 (b)). This is as a result of slow carbonation rate associated with hydraulic lime and possible effect of prevailing low relative humidity (40±5%) in the laboratory curing conditions. Nevertheless, decrease in the total pore volume (i.e., from 27.77 to 23.42%) is recorded during the curing period (Table IV). This shows replacement of some of the existing capillary voids by formation of the hydration and carbonation products (from hydraulic lime) over time. Also, arising from evaporation of excess kneading water and the drying shrinkage that accompanied that, the bulk density became reduced (from 1.75 to 1.72 g/ml).

On the contrary, addition of cement to the mortar resulted in unimodal PSD curve with narrow pore range, reduced intrusion peaks and presence of finer pores at 1 and 6 months (Fig. 5 (a) and (b)). This can be attributed to lower W/B ratio due to reduction in lime content in the composite, faster hydration associated with cement and subsequent replacement of larger pores by C–S–H gel and other hydration products [45]-[47]. This may account for the observed densification of the mortar matrices, reduced porosities, and prevalence of smaller pore sizes in lime mortar due to the addition of cement. Usually, quantity of kneading water becomes lower as cement content in the paste becomes higher (as observed in Section 3.1). The curves are therefore shifted towards the right (relative to lime mortar), which shows dominance of smaller pores (10-
500 nm range). These observations are clearly shown in Fig. 6 as the pronounced effects between lime mortar (L13) and the composite having the highest cement content (LC13) at 6-month of curing are comparably illustrated.

The observed reduced intrusion peak and dominance of finer pores can be related to replacement of larger pores by C–S–H gel and other hydration products, leading to presence of more smaller pore sizes and consequent reduced porosity in lime mortar. In particular, porosity is reduced by about 8% in LC13 over the 6-month period.

Results of the Total Water Absorption of the mortars are shown in Fig. 7, to further assess the mortars’ microstructural features. For lime only (L13), water absorption values were 13.84% and 13.68% at 1 and 12 months of curing respectively. This reduction relates to continuous formation of hydration and carbonation products from the hydraulic lime, leading to porosity reduction.

Progressive addition of cement results in relatively small but steady reduction in the total water absorption, leaving LC13 at 11.68% over 12 months. It can be inferred that addition of cement to lime mortar induced modification of the mortar microstructures with a reduction in porosity over time. This decrease in the pore volume is the result of filling of larger pores in lime mortar by the cement hydration products, particularly Calcium Silicates. Consequently, there is more restriction on water movements, leading to the observed reduced water absorption pattern (Fig. 7). However, within each mortar mix, reduction pattern in water absorption was low. This may also be associated with suspected low carbonation of the mortars after the initial hydration.

**C. Mechanical Properties**

Table V summarises results of the flexural strength, compressive strength, and modulus of elasticity, for the mortars over a 12-month curing period. Each result was obtained from the average of three similar specimens for flexural strength, and five similar specimens for both compressive strength and modulus of elasticity each. It could be noted that the Coefficients of variation (COV) fall substantially within the lower range (0-17%) with few outliers, however. This generally suggests consistent results.

For lime only, decrease in the flexural strength is observed (0.70 Mpa and 0.61 Mpa at the first and twelfth months of curing). This may result from formation of different pore sizes due to evaporation of excess unbound kneading water as observed in the microstructural analyses above. Progressive addition of cement to the mortar, raised the values for the flexural strength throughout the curing period, although not in a linear trend. This is because quicker hydration process, faster formation of C-S-H gel, reduced pore sizes and volume, associated with Cement, increased the flexural strength at a relatively shorter rate. This is applicable to all the composites. In this regard, of particular reference is LC12 which recorded the highest values of 4.12 Mpa and 4.23 Mpa at 1 month and 12 months of curing, respectively.

Compressive strength developments follow similar trends with the flexural strength. For lime only, compressive strength recorded 0.61 Mpa and 0.66 Mpa at 1 month and 12 months respectively. This marginal increase may also result from formation of different pore sizes resulting from evaporation of excess unbound kneading water as shown in the microstructural analyses above. As expected, these values significantly increased with increasing contents of cement. Highest values of 9.17 Mpa and 12.34 Mpa were recorded at 1 and 12 months of curing, for the mix with triple cement contents (LC13). Compared with the mix with no cement content (L13) over the same curing period, this represents a significant increase (in each case) above.
As revealed in the values obtained for their moduli of elasticity (E), mortars’ practical performance is related to the compressive strength recorded in each case. From Table V, after 12 months of curing, lowest ‘E’ value was recorded for L13 with the lowest compressive strength. Conversely, over the same curing period, the mix with the highest compressive strength, LC13, recorded the highest ‘E’ value. Nonetheless, both mixes have the tendencies to exhibit contrasting elastic behaviours (high and low respectively), as revealed in their stress-strain relationships at 1, 3, 6, 9 and 12 months of curing (Fig. 8 (a) to (e)), with varying capacities to absorb deformation. The pattern is also applicable to the other mixes. From the figures, as the slope of the curve becomes smaller, the better the elastic behaviour. Every mortar exhibits comparably varying slopes relative to lime mortar’s (L13). Compressive stress relates proportionally with increase in the content of cement as reflected on the behavioural patterns for the individual composites (Fig. 8 (a) to (e)). The specimen having the highest cement content (i.e., LC13 with 75% cement content) for example, sustained about 8 Mpa and 12 Mpa maximum compressive stresses after 1 and 12 months respectively. This is at variance significantly with LC31, the composite having the least cement content (i.e., having 25% cement content), capable of sustaining maximum compressive stresses of 2 Mpa and 3 Mpa only, over the same period respectively. Generally, increase in cement content is associated with higher slope of the curve. This is characterised with negative consequences on the strain, thereby reducing the ability of the sample to sustain deformation. This leads to decrease in flexibility. Thus, as LC13 curve is observed with the highest gradient, it is characterized with the least flexibility, and consequently, the least capability to sustain deformation before failure. This observation is further supported with the mortars’ Compressive/Flexural strength ratios (f/c), and Moduli of Elasticity relationships, with increasing cement contents as demonstrated in Fig. 9.

Fig. 9 confirms that the lower the E-value, so also the lower the (f/c) ratio, and the higher the flexibility of mortars. These descriptions are in agreement with the studies by [48] and [49], indicating that mortar’s ‘E’ value is inversely proportional to its elastic behaviour.

D. Further Result Analyses: Examining the Pore Structures

From Figs 8 (a)–(e), the patterns show by the composites indicate that they are relatively characterized by elastic behaviours compared with the basic lime-based mortar (specimen L13). To ascertain these behaviours, since nano structural and colloidal characteristics of lime, and the carbonation process by which it hardens strongly influence the ultimate performance of lime mortars [28], mortars’ pore structures were examined through further analyses of their MIP results. Since porosity is measured directly by a volume of mercury intrusion into the specimen, volumes of intruded mercury for corresponding pore ranges were subdivided into six pore size classifications and ranges (as recognised/applicable to AutoPore MIP software) in order to obtain total porosity for each range, as indicated in Table VI.

| S/N | Pore Size Classification | Adopted Acronym** | Pore range |
|-----|--------------------------|-------------------|------------|
| 1   | Inter-Nano pores         | Inp               | 1–10 nm    |
| 2   | Super-Nano pores         | Snp               | 10–100 nm  |
| 3   | Sub-Micro pores          | Smp               | 0.1–1 µm   |
| 4   | Inter-Micro pores        | Imp               | 1–10 µm    |
| 5   | Super-Micro pores        | SmP               | 10–100 µm  |
| 6   | Sub-Milli pore           | Smm               | > 100 µm   |

Source: MIP AutoPore software.
Note: ** - Acronym adopted solely for this analysis.

TABLE VI: ADOPTED CLASSIFICATION AND PORE RANGE
Fig. 8. Compressive Strength /Strain relationships for lime and composite mortars over a 12-month curing period: (a) At 1-month curing; (b) At 3-month curing; (c) At 6-month curing; (d) At 9-month curing; (e) At 12-month curing.

Fig. 9. Compressive/Flexural strength ratios ($f_{c}/f_{f}$) versus Moduli of Elasticity for lime and composite mortars over a 12-month curing period (general relationships).

Lime porosity with interconnected pores facilitate its ability to meet opposing moisture movement requirements of breathability in the large pores and capillary transport in the small pores [19]. Following the classifications in Table VI, lime’s predominant pore range is notably within Sub-Micro pores (SMP) and Inter-Micro pores (IMP) regions, the regions that are effective for moisture movement. This is important for dissolution of CO$_2$ and eventual carbonation reactions. Fig.10 therefore shows volumes of intruded mercury for each pore range (based on the classification in Table VI) in all the mortar samples. The data were collected after 6-months of curing.

Since the study is primarily based on B/A ratio of 1:3, sample L13 (lime only, with 0% cement content) is used as...
the reference sample for the comparative analyses. Referencing Fig. 10, the dotted line indicates direction (downward) of higher cumulative porosity (i.e., summation of the porosities) in the first three pore regions of Inter-Nano pores (1-10 nm), Super-Nano pores (10-100 nm) and Sub-Micro pores (0.1-1 µm), for each of the mortars. Low cumulative porosity across the first three pore regions in a mortar is observed to be synonymous with low cement content, low Modulus of Elasticity, low (fc/ff), and thus, high flexibility. This observed trend as noted in Fig. 10 is well stated in Table VII.

As mortar’s Modulus of Elasticity is a function of its flexibility, the lower the E-value, the higher the flexibility. Fig. 11 therefore shows comparative tendencies of the mortars to exhibit flexibility, relative to the reference sample, L13. It could be observed that the cumulative porosity across the first three pore regions in a mortar is proportional to cement content and the E-value, as the pattern is illustrated using dotted arrow in Fig. 11.

From Fig. 11, it can be observed that the lower the cumulative porosity, the lower the E-value, thus higher flexibility (and vice versa). This observation is also supported by the apparent linear coefficient of correlation ($R^2 = 0.89$) for the equation describing both ‘cumulative porosity’ (within the referenced three pore ranges) and cement content. This relationship can be expressed as:

$$C_p = 55.212c + 7.421$$

where $C_p$ represents cumulative porosity, and $c$ represents cement content.

Thus, it could be inferred that the mortars would exhibit flexibility in the descending order of LC31, LC21, LC11, LC12 and LC13 for lime-cement composites (with LC13 being the least flexible i.e., with the highest E-value of 780.83 MPa) as shown in Fig. 11. These results were expected, as composites with highest lime contents (i.e., LC31) was expected to exhibit highest flexibility in lime-cement composite. It could therefore be demonstrated that the pore structure, as defined by the volume of mercury intrusion into the specimen, across the three low pore regions of Inter-Nano pores (1-10 nm), Super-Nano pores (10-100 nm) and Sub-Micro pores (0.1-1 µm) of lime-cement composite is a function of the composite’s flexibility. The cumulative porosity across the three low pore regions has been shown to be synonymous with low (fc/ff), thereby defining a measure of flexibility. Accordingly, the composites have shown to exhibit more flexibility in the descending order of cement content.
IV. CONCLUSION

This study attempted to evaluate pore structures of lime composites in order to determine lime based mortars’ flexibility tendencies. This is particularly useful in reviving sustainable eco-friendly lime based mortars relative to predominant polymer based cementitious products. Using ‘lime-cement’ composites, efforts are being made to evolve low carbon composite construction materials, with cement serving as a partial replacement for lime in each case. This is particularly useful with a view to promoting flexibility property of age-long building lime, known for its building structural failure accommodation. While the flexibility property of lime in the composites (via their pore structures) is explored, addition of cement to lime, to form a composite, increases lime’s both compressive and flexural strengths. This is however characterized with reduced flexibility, subject to the cement content in the composite.

CONFLICT OF INTEREST

The author declares no conflict of interest.

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