

# DEVELOPMENT OF SUSTAINABLE MORTAR AND CONCRETE MADE OF LIMESTONE BLENDED CEMENT – INFLUENCE OF PARTICLE SIZE

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**Abstract:** Mortars and concretes with 20% substitution of cement by limestone were studied. Four different limestone powders having different particle diameters and surface areas were examined: one size smaller with high surface area, two sizes of larger with low surface area and one size similar to the original cement particles. It was shown that the compressive strength of the mortar and concrete made with limestone of different particle sizes were affected significantly by the surface area and size of the particles. Blended mortar and concrete made of medium limestone particle size had shown the highest density and hence the greatest compressive strength, while blended mortar and concrete with the fine limestone particles, which are commonly used in the construction industry, exhibited the lowest density and hence the smallest compressive strength. By controlling the size of the limestone particles and surface area, concrete and mortars with higher compressive strength can be achieved compared to those reported in the literature with similar clinker substitution content. Therefore the current work results have not only important scientific implications but also practical ones.

## 1 INTRODUCTION

Mortars and concretes are widespread materials in the construction industry, due to their good workability, high compressive strength and durability. Yet, manufacturing of cement, the binder of concretes and mortars, has significant energy demand and CO<sub>2</sub> emission footprint [1]. In order to reduce the environmental damage and costs, partial substitution of clinker, the active component of the cement, by materials with lower environmental footprint, is practicable. Limestone is one of the most widespread used materials for clinker substitution due to its low price and high availability.

In the last decade, comprehensive research took place to examine limestone as a partial substitute for clinker [2-6]. Acceleration of the hydration process at early age with low strength and durability due to reduction of the active

material was reported. Grinding limestone together with clinker is a common way in the cement manufacture to prepare blended cements. Due to the soft nature of the limestone compared to that of the stiff nature of the clinker, the resulting limestone particle sizes are significantly smaller than those of the clinker. The small size of the limestone particles is of high influence on the limestone particles distribution in the cement paste as they tend to agglomerate in the cement powder, lowering the limestone efficiency and hence the hardened cement paste compressive strength [3,4]. Previous studies examined cement pastes with partial substitution of the clinker by limestone with controlled particle size [7-8]. It was found that the surface of the limestone particles influences the hydration rate of the cement paste, as limestone particles increase the nucleation sites for the hydration reactions.

Also the compressive strength of the hardened cement paste was influenced by limestone particle size. Up to date most studies examined the behavior and properties of limestone-based blended cements having different particles sizes whereas hardly any works were explored the influences of limestone particle size on mortar and concrete behavior, which is the basis of this work.

The goal of this work was to develop mortars and concretes with partial substitution of clinker by limestone with different particle sizes. The limestone was grinded separately from the clinker in order to obtain controlled particle sizes which are smaller, bigger or similar to the clinker particle size. The fresh and harden properties of mortar and concrete mixtures made of blended cements of different limestone particle sizes were investigated, including measurements of air content, harden mixture density and compression strengths.

## 2 MATERIALS AND METHODS

**Powders:** CEM I 52.5 R was partially replaced with 20% (wt.) limestone powders (> 99.8% CaCO<sub>3</sub>, (CC)) of varying particle sizes. Limestone powders of four different particle diameters and surface areas relative to the cement particles were examined: smaller with greater surface area – one type, larger with lower surface area – two types, and similar – one type, to that of the original cement, as well as plain cement without addition of limestone. The two large particle limestone size powders were prepared by sieving the limestone powder through 90 micron mesh, where only the fraction under 90 microns was used, and by sieving the limestone powder between 90 and 45 micron mesh. Note that the source of the large particle powders was waste of the cement industry quarry (of Nesher Ltd).

The surface area and particle size distribution were examined for the four different limestone systems and the original reference cement. The surface area of each individual powder was determined using the BET technique with N<sub>2</sub>. The surface areas were calculated by multiplying the cumulative relative weight of each powder by the surface

area of the individual component in the powder mixture. The particle size distribution was determined by Laser Granulometer (ANALYSETTE 22 MicroTec Plus laser diffraction). All limestone powders were dried at 110 °C for 48 hours before blended cement preparation.

**Mortar and concrete:** Mortar and concrete mixtures were prepared with the blended cements according to C-109 ASTM and to IS (Israel Standard) 118, respectively. The basic mixes design of the mortar and concrete are presented in Table 1 with w/c ratios of 0.49, 0.62, for the mortar and concrete, respectively. Reference mixtures of mortar and concrete without limestone were also prepared for comparison.

**Table 1:** Mix design of mortar and concrete with limestone in Kg/m<sup>3</sup>.

|          | Agg.<br>9.5mm | Agg.<br>19mm | Sand | CC  | CEM |
|----------|---------------|--------------|------|-----|-----|
| Mortar   | -             | -            | 1468 | 107 | 429 |
| Concrete | 369           | 743          | 555  | 68  | 272 |

The mortar and concrete mixtures were cast to 5cmX5cm and 10cmX10cm cube molds, respectively, for compression tests. Then cured in tap water for 7 days and stored in room temperature and relative humidity (RH) of 60% until testing in compression at age of 28 days.

Fresh mortar and concrete mixtures properties were determined: workability, bulk density and air content, according to ASTM C 1437 and IS 26, respectively. At the harden stage the compressive strength was measured on a test machine with maximum load capacity of 2000 KN following ASTM C-109. The testing rates were 500 N/s and 1000 N/s for the mortar and concrete respectively. The density of the harden mixtures was evaluated by weight to volume. Four specimens of each mixture with the various blended cements of both mortar and concrete were tested in compression.

The microstructure of the different specimens was characterized using polished cross sectional specimens to evaluate the limestone particle distribution and their

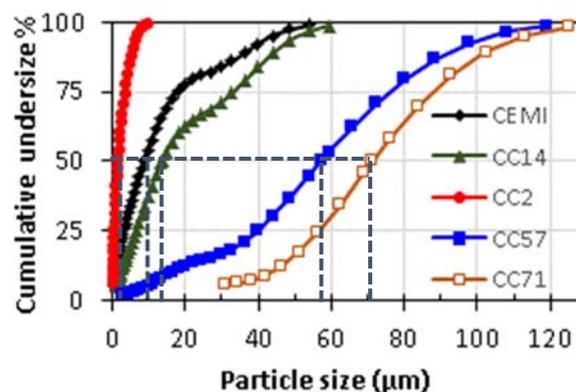
influences on the microstructure of the mortar and concrete, using A FEI Quanta 200 scanning electron microscope (SEM). The cross-sectioned samples were initially ground in five steps, using grinding paper containing silicon-carbide particles with 30 $\mu\text{m}$ , 16 $\mu\text{m}$ , 9 $\mu\text{m}$ , 5 $\mu\text{m}$  and 3 $\mu\text{m}$  grit sizes, after which they were further polished with 1 $\mu\text{m}$  diamond paste. The samples were then cleaned with acetone in a supersonic cleaner for 180 sec.

### 3 RESULTS AND DISCUSSION

#### 3.1 Powders

The particle size distribution (PSD) of the four tested limestone powders and the original cement is presented in Figure 1, showing the particle size as a function of the cumulative volume of the solid. Median particle size of each powder, based on the PSD analysis, was determined by volume of the particles and represented by the dashed lines. The four different limestone sizes used had median particle sizes of 71  $\mu\text{m}$ , 57  $\mu\text{m}$ , 14  $\mu\text{m}$  and 2  $\mu\text{m}$ , each of which is signified by CCX, such that X stands for the particle size, e.g., CC2 is the limestone powder with median particle size 2  $\mu\text{m}$ . The original cement has average particle size of  $\sim 9$ . In which CC2 has smaller average particle size than that of the original cement, CC71 and CC57 have larger average particle sizes than the original cement, while CC14 shows similar particle size distribution as the original cement. In the case of the limestone powder with the smallest average particle size of 2  $\mu\text{m}$ , most of its particles are smaller than those of the original cement. In the case of the two limestone powders with the large particles big portion of the particles is larger than the original cement, in which CC57 exhibits wider particle size distribution. The average particle size of the different powders and their surface areas are provided in Table 2. The surface area of the fine limestone powder is the highest (CC2) and the lowest surface is for the limestone with the largest particle size (CC71), as expected. The medium particle size limestone exhibits similar surface area as that of the original cement. The high surface area of the

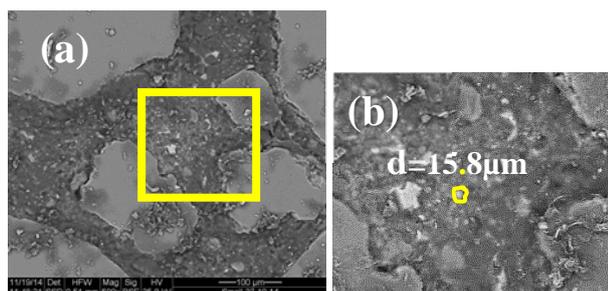
fine limestone particles led to aggregation of the particles (Figure 2) before adding to the mixture. Aggregation of the fine limestone particles in the mortar and concrete mixtures was also observed at their harden stage by SEM (Figure 3). Limestone particle size of above 15  $\mu\text{m}$  is seen, although the original average size is of 2  $\mu\text{m}$ . Note that for the fine limestone less than 0.2% of the total particles were reaching particle size of 10  $\mu\text{m}$ , the size of most particles was much lower than that. Therefore such high limestone diameter of 15  $\mu\text{m}$  can indicate agglomeration of the fine particles when they are part of the mortar and concrete mixtures. Note that similar agglomerates were seen through the entire CC2 specimen cross section of both mortar and concrete mixtures. In the other blended mixtures the measured size of the limestone was correlating with their PSD results. Poor limestone particle dispersion is expected to influence the properties of the mortar and concrete at their fresh and harden stages.



**Figure 1:** Particle size distribution (PSD) of the examined powders (cement and limestone).

**Table 2:** Average particle size and surface area of the examined powders.

|                                      | CEM  | CC71 | CC57 | CC14 | CC2  |
|--------------------------------------|------|------|------|------|------|
| Particle size, $\mu\text{m}$         | 8.9  | 71.3 | 57.2 | 13.6 | 1.8  |
| Surface area, $\text{m}^2/\text{gr}$ | 1.53 | 0.24 | 0.39 | 1.37 | 6.22 |

**Figure 2:** (a) As received limestone powder of CC2  $\mu\text{m}$ , (b&c) microstructure of mortar made of fine limestone, showing limestone agglomeration.**Figure 3:** SEM images of mortar made of fine limestone, showing limestone agglomeration: (a) X500, (b) X1200

### 3.2 Mortars and concretes

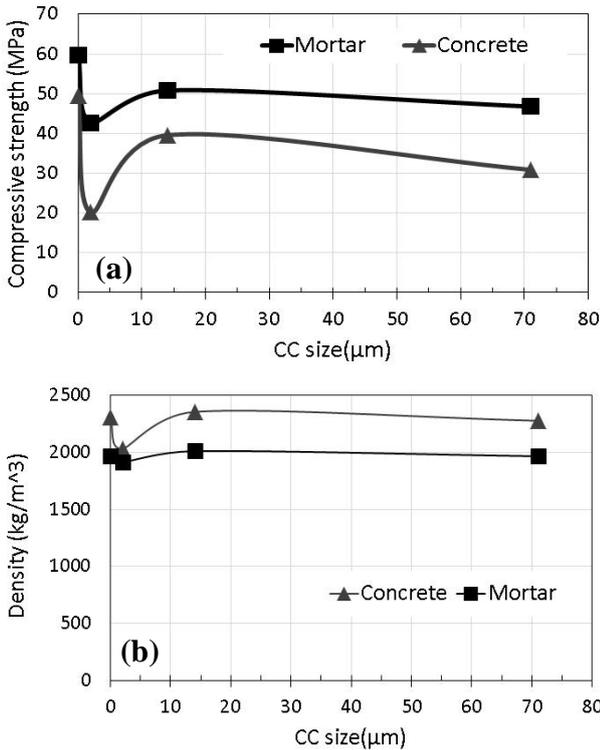
The compression strengths vs. limestone particle size of the mortar and concrete mixtures are presented in Figure 4a. It is obvious that limestone particle size influences the compressive strength of both, mortar and concrete mixtures, in a similar manner. In which the fine limestone particle CC2 offers the lowest compression strengths and the medium particle size CC14 provides the greatest compression strength of both mortar and concrete made of blended cement. Note that the references concrete and mortar mixtures without limestone observed the greatest compression strengths as expected, as 20% of

the active component (clinker) was replaced with the non-active component, i.e., limestone.

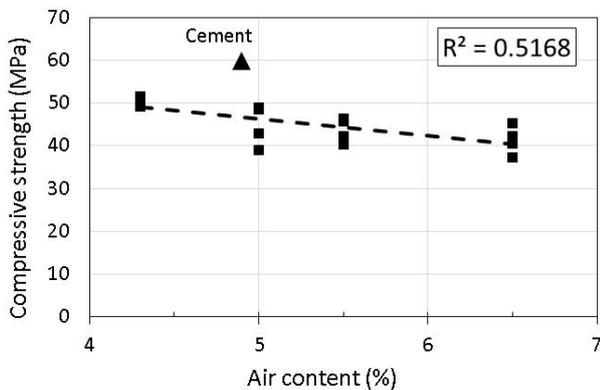
The density of the harden mixtures at age of 28 days (Figure 4b) indicates similar trend as that of the compression strength, low density for the mortar and concrete with the fine limestone powder CC2 and high density for the mortar and concrete with the medium size limestone particles among the blended-cement-based mixtures. The differences in the harden mixtures density can be related to the limestone particle size as this is the only dissimilarity between the various mixtures (with the limestone). This can be explained based on limestone particles dispersion within the mortar and concrete mixtures and the related packing density of these mixtures. The fine particles (CC2) agglomerate within the mixture due to their high surface area as was discussed above (Figures 2,3) leading to low packing density of the entire mixture and thus to reduced compression strength. While the medium size limestone particles (CC14) disperse more efficiency in the mortar and concrete mixture; leading to high packing density and thus to greater compression strength.

Based on the above discussion it can be concluded that the limestone particles packed differently within the mortar and concrete mixtures depends on their size and surface area, where high packing density leads to greater compression strength. This behavior is clearly observed in Figure 5 that shows a linear correlation between the air content at the fresh stage of the mortars and their compression strength; greater air content leads to lower compression strength. Note that the highest air content is for the CC2 mortar and the lowest is for the CC14 mortar where the two in the middle are for the big limestone particle mortars (CC57 and CC71). Also note that the reference mortar observed relatively high air content as compared to those with the limestone, excluding the CC14 mortar that provides the greatest mixture packing. Similar trend was also observed for the concrete mixtures but to less extent. In this work the medium particle size limestone mixture observed the lowest air content at the fresh stage with the greatest density at the harden

stage, thus providing the highest compression strength among all limestone-based mixtures.



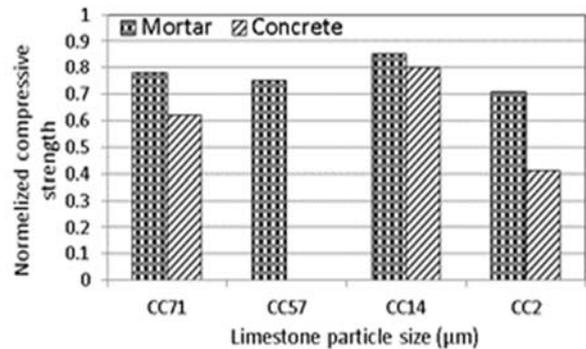
**Figure 4:** Mortar and concrete mixtures properties at the age of 28 days vs. limestone particle size: (a) compression strengths, (b) density. Zero presents the reference mixtures without limestone.



**Figure 5:** Compression strengths of the mortars with the limestone of different particle sizes vs. the air content of the fresh mixture.

A comparison between the compression strengths of the mortars and concretes with the limestone relative to the references without limestone is presented in Figure 6, via normalized values. The figure indicates a significant reduction in the compression strengths of 26% for the mortar and as high as

60% for the concrete with the fine limestone particles (CC2). Similar reductions but of slight less extent is seen for the mixtures with the big limestone particles (CC71 and CC57). Note that similar reductions for concrete and mortar with same limestone contents were also reported in the literature [3,6]. However, much smaller loss in the compression strengths is observed for the medium size limestone particle (CC14) if only 15% and 20% for the mortar and concrete, respectively. This is very attractive when considering the replacement amount of the active original cement with the non-active limestone, which was 20%. This means that by controlling the particle size of the limestone it is possible to increase the efficiency of the limestone and produces blended mortar and concrete mixtures with improved performance.



**Figure 6:** Normalized compression strengths of the concretes and mortars with the limestone relative to the references without limestone.

The basic role of fracture of a concrete specimen is presented in Figure 7. Such mode of fracture was observed for all tested systems, with no significant difference between those with and without limestone.



**Figure 7:** Role of fracture of concrete

It should be remembered that today the common way in the cement industry to produce blended cements is to grind the limestone together with the clinker. However due to the soft nature of the limestone compared to the stiff nature of the clinker, the resulting limestone particle sizes are significantly smaller than those of the clinker, which are presented in this work by the CC2. However small size of limestone particles are tend to agglomerate, lowering the mortar and concrete mixtures packing density and hence their compressive strengths as was observed also in this work.

#### 4 CONCLUSIONS

It was shown that limestone powder particle size has a major influence on the packing density of both fresh and hardened mortars and concretes, leading to improved compressive strength. By controlling the size of the limestone particles, concrete and mortars with higher compressive strength can be achieved compared to those reported in the literature with similar clinker substitution content. Therefore the current work results have not only important scientific implications but also practical ones.

Based on this work it can be concluded that sustainable mortars and concretes can be produced with significant percentage of an environmentally friendly, inert, available, and economical material, by substituting the high energy demand and CO<sub>2</sub> footprint clinker with controlled particle size limestone.

#### 5 ACKNOWLEDGMENTS

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