Improving the performance of crushed limestone concrete utilizing supplementary cementitious materials: A case study in Iraq

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Abstract. This paper presents an experimental work conducted to study the properties of concrete incorporating (0-100) % of local (in Iraq) crushed limestone (CLS) as a partial replacement of coarse aggregate. Supplementary Cementitious Materials (SCM) such as High Reactivity Metacaoline (HRM) and Rice Husk Ash (RHA) were added to the mixtures to improve the properties of fresh and hardened concrete. The selective mixtures in this study were arranged into three groups, the first without (SCM), the second with (RHA), and the third with (HRM). Each of them (being with different ratios of (CLS) as a coarse aggregate) was tested for comparison with the reference mixtures. The slump and air contents were evaluated for the fresh concrete. The absorption as well as compressive strength (CS) and flexural strength (FS) were evaluated for the hardened specimens after 7 and 28 days. According to the results, the concrete with CLS as a coarse aggregate gained less performance than the ordinary concrete but the (SCM) improved the properties of the concrete mixtures. The results show that the HRM was more effective than the RHA with respect to the mechanical properties.

1. Introduction

Concrete is one of the important constructional materials as far as strength and durability characteristics are concerned [1]. It can be considered an attractive material for a wide range of structural applications. The aggregate is an important constituent in concrete and its role is more important. It occupies most of the volume of concrete and has a significant impact on concrete performance [2, 3]. However, the concrete industry is problematic one due to many and environmental concerns, for example, it has been proved that the cement industry is a major source of harmful gases that could cause global warming [4-6], the latter is responsible for water shortage [7-11], and water pollution [12-14]. Besides, the produced water from concrete plants is heavily polluted with a wide range of pollutants [15-17], such as organic matter and suspended solids [18-21]. Therefore such wastewaters require efficient treatment technologies, such as coagulation [22-
25], filtration [26-31], electrochemical [32-38], or even hybrid methods [39-44]. Furthermore, the extensive use of natural aggregates results in depletion of natural resources. Therefore, serious search has been initiated to find eco-friendly alternatives for both cement and aggregates. Limestone is the most widely available rock in Iraq and particularly in the Governorate of Mosul, where it can be extensively used in construction activities. It consists essentially of calcium carbonate, with which there is generally some magnesium carbonate and siliceous matter such as quartz grains [45, 46]. Introducing limestone aggregates in concrete mixtures can reduce environmental pollution. The production of limestone fillers requires much less energy than producing cement which also means that carbon dioxide is much lower in limestone production than cement production. Limestone fillers result in more stable concrete quality thus reducing the amount of waste and improving durability which leads to a longer life for concrete [47-50].

The ratio of limestone as aggregate in concrete was only 1.3% (in 1974). Since then, the shipments and the usage of limestone have increased every year with the demand for concrete and the shortage of other natural aggregates [51]. The coarse aggregate can significantly affect the concrete due to its gradation and the bond with the surrounding mortar. The impact of the transition zone depends on its microstructural itself [52]. The coarse aggregate parameters such as the size distribution and content of aggregate play a significant effect on the compressive strength of concrete. The engineering properties and suitability of the crushed stone aggregate for use in bituminous mixes were studied using two resources of crushed limestone in Iraq. It was observed that the limestone itself meets the Marshall test requirements of a bituminous mix and can be used to produce these mixes. It is well known that the engineering materials (sand, gravel) are limited. They must be replaced by another available alternative. The crushed limestone as a coarse aggregate and its dust which is resulting in the crushing process can somehow fulfill the demand for aggregate in construction. Kürklü, Görhan and Materials [53] investigated the impact of limestone dust in terms of compressive strength, flexural strength, impact resistance, absorption, water permeability, and other properties. They concluded that the fine aggregate could be replaced by the dust of the crushed limestone without affecting the properties of concrete. The stone dust is a suitable choice as fine aggregate in concrete construction.

The use of supplementary cementing materials such as ground blast furnace slag, silica fume, metakaolin, coal fly ash, and natural pozzolan can produce a very significant effect on the pore solution chemistry of concrete, depending on the dosage and composition of these supplementary cementing materials. The influence of mineral and chemical admixtures on the strength development and chloride ion permeability of high-performance concrete (HPC) was studied in the literature. The results indicate that unique combinations of micro silica and superplasticizer exist for HPC mixes with negligible to very low chloride ion permeability [54]. On the other hand, concrete with the addition of SCM has been reported with good properties, emphasizing the favorable impact of the SCM in the performance of its resistance and durability, as well as in the environmental benefit involved.

Crushed limestone was investigated in this research study as a substitute to gravel for many reasons including (local availability, low price, and its ability to reduce environmental pollution). To the best of the authors’ knowledge, this is the first time (in Iraq) to study crushed limestone as a substitute to coarse aggregate in concrete mixes. The objectives of this research are:

1. To study the effects of locally (in Iraq) crushed limestone as a coarse aggregate on some properties of concrete.
2. To investigate the improvement which can be achieved on the properties of limestone concrete by introducing two types of supplementary cementitious materials (SCM), specifically High Reactivity Metacaoline (HRM) and Rice Husk Ash (RHA).
2. Materials and Methods

Materials used in this research include the ordinary Portland cement which conforms to the Iraq specification No.5:1984 (Iraqi Organization of Standards, IOS5:1993, for Portland Cement). The cement has a specific gravity of 3.14; the initial and final setting times are 120 min and 255 min respectively. The sand has a specific gravity of 2.62 with particle size ranging from 200μm - 4.75mm. The natural aggregates proceed from crushed rock (coarse), the natural gravel, and the natural sand, with an adequate particle size gradation according to the limits established by ASTM C 33 / C33M-18. The RHA and HRM used in this work conform to the chemical and physical requirements of ASTM C618-15. The Specific gravities of RHA and HRM were 2.14 and 2.63 respectively. Tables (1) and (2) describe the chemical and physical properties of each of RHA and HRM. Limestone was supplied from Dukan, situated in the Northeast of Sulaymaniah Governorate in Iraq. Alumina ratio (AR) and silica ratio were 1.291 and 2.868 respectively. The maximum size for each crushed limestone and gravel is (19 mm) and the bulk density test for each crushed limestone and gravel were done according to ASTM C127-12. Tables (3) and (4) describe the properties of crushed limestone and gravel. The chemical and physical tests of the RHA, HRM, and CLS were made by the National Center for Construction Laboratories and Research in Baghdad.

**Table 1.** Chemical analysis of HRM and RHA.

| Oxide composition | Oxide content % |
|-------------------|-----------------|
|                   | HRM  | RHA |
| SiO₂              | 53.66| 86.53 |
| Al₂O₃             | 35.56| 0.12 |
| Fe₂O₃             | 1.52 | 0.30 |
| SO₃               | -    | 0.10 |
| Na₂O              | 0.03 | 1.45 |

**Table 2.** Physical Properties of HRM and RHA.

| Physical Properties | HRM | RHA |
|---------------------|-----|-----|
| Strength activity Index | 149 | 138 |
| Flow table test     | 93  | 110 |
| Specific gravity    | 2.61| 2.12|

**Table 3.** Chemical components of crushed limestone (CLS) and gravel.

| The aggregate   | The component (%) |
|-----------------|--------------------|
|                 | SiO₂HP | Al₂O₃ | Fe₂O₃ | CaO | MgO |
| Crushed limestone | 0.814  | 0.168 | 0.126 | 53.728 | 1.018 |
| Gravel          | 84.81  | 6.16  | 1.66  | 1.72 | 0.06 |

**Table 4.** Physical properties of crushed limestone (CLS) and gravel.

| Aggregate type   | Bulk Density (g/cm³) | Specific Gravity | Porosity (%) | Moisture Content (%) | Crushing strength (Kg/cm²) |
|------------------|----------------------|------------------|--------------|----------------------|---------------------------|
| Crushed limestone| 1.27                 | 2.59             | 4.6          | 0.23                 | 971.2                     |
| Gravel           | 1.61                 | 2.68             | 1.42         | 0.03                 | 1855                      |
2.1 Test Methods
Tests were conducted according to the following specifications for fresh and hardened concrete:
- Slump: ASTM C143.
- Air content: ASTM C173/C173M.
- Compressive strength (CS): ASTM C39/C39M.
- Flexural strength (FS): (Three-point load test), ASTM C 78 –02.
- Absorption: BS 1881:122-1983

Tests of the mixtures for each of the CS and FS were done by using three specimens, then the mean value was dependent.

2.2 Mix proportioning
The mixtures were designed and prepared to study the properties of concrete with different increments of limestone in the mixtures. The (CLS) was added to the mixtures as a percentage varying from 25% to 100% by weight of coarse aggregate, while 0% replacement served as the control.

For the purpose of comparison, the reference mixtures were produced without crushed limestone but with natural locally available gravel. The reference mixtures (without SCM or CLS) were designed in the laboratory according to the guidelines of ACI-211.1-91 to comply with the requirements of workability and CS. These mixtures are composed of ordinary cement, natural sand, and gravel. RHA was added as 20% by weight of cement, as recommended by ASTM C311-02. The most suitable percentage of HRM as a partial replacement of cement was (8%) in view of strength as concluded by Alhadithy [55]. These ratios of RHA and HRM were suggested to serve the required properties. Here, we define:
- RF = reference mixture without CLS and SCM.
- RF-20RHA = reference mixture with 20% RHA but without CLS.
- RF-8HRM = reference mixture with 8% HRM but without CLS.

It was referred to the mixtures containing 25%, 50%, 75%, and 100% of coarse CLS as CLS25, CLS50, CLS75, and CLS100 respectively. The symbol 20RHA refers to the mixtures incorporating 20% of rice husk ash, and 8HRM referred to the mixtures incorporating 8% of high reactivity Metacoline. Generally, the mixtures in this study can be classified into three groups:
- The first group: mixtures with different ratios of CLS but without SCM
- The second group: mixtures with different ratios of CLS and 20% RHA
- The third group: mixtures with different ratios of CLS and 8% HRM.

The mix proportion of the three groups of mixtures is listed in tables 5, 6, and 7.

**Table 5.** The mix proportion without SCM (kg/m³).

| The mixtures | Cement | Sand | Gravel | Crushed limestone | Water | Water-cement ratio (W/CM) |
|--------------|--------|------|--------|--------------------|-------|--------------------------|
| RF           | 450    | 675  | 1025   | --                 | 216   | 0.48                     |
| CLS25        | 450    | 675  | 769    | 256                | 225   | 0.50                     |
| CLS50        | 450    | 675  | 513    | 512                | 235   | 0.52                     |
| CLS75        | 450    | 675  | 257    | 768                | 245   | 0.54                     |
| CLS100       | 450    | 675  | -      | 1025               | 250   | 0.55                     |
Table 6. The mix proportion with 20% RHA (kg/m³).

| The mixtures  | Cement | RHA% | Sand | Gravel | Crushed limestone | Water | W/CM |
|---------------|--------|------|------|--------|-------------------|-------|------|
| RF-20RHA      | 360    | 90   | 675  | 1025   | --                | 207   | 0.46 |
| CLS25-20RHA   | 360    | 90   | 675  | 769    | 256               | 216   | 0.48 |
| CLS50-20RHA   | 360    | 90   | 675  | 513    | 512               | 221   | 0.49 |
| CLS75-20RHA   | 360    | 90   | 675  | 257    | 768               | 234   | 0.52 |
| CLS100-20RHA  | 360    | 90   | 675  | -      | 1025              | 243   | 0.54 |

Table 7. The mix proportion with 8% HRM (kg/m³).

| The mixtures   | Cement | HRM | Sand | Gravel | Crushed limestone | Water | W/CM |
|----------------|--------|-----|------|--------|-------------------|-------|------|
| RF-8HRM        | 414    | 36  | 675  | 1025   | --                | 207   | 0.46 |
| CLS25-8HRM     | 414    | 36  | 675  | 769    | 256               | 212   | 0.47 |
| CLS50-8HRM     | 414    | 36  | 675  | 513    | 512               | 212   | 0.47 |
| CLS75-8HRM     | 414    | 36  | 675  | 257    | 768               | 225   | 0.50 |
| CLS100-8HRM    | 414    | 36  | 675  | -      | 1025              | 234   | 0.52 |

3. Results and Discussion

Tables 8, 9, and 10 describe the slump, air content, CS, FS, and absorption of the hardened concrete for the different mixtures as described in section 2. The mean value of the properties for each of the three groups of the specified mixtures is listed in table 11.

Table 8. Properties of mixtures without SCM.

| Mixtures | Slump (mm) | Air content (%) | CS (MPa) | FS (MPa) | Absorption (%) |
|----------|------------|-----------------|----------|----------|----------------|
|          |            |                 | 7 days   | 28 days  | 7 days         | 28 days |
| RF1      | 103        | 2.5             | 23.93    | 33.51    | 4.56           | 6.38    | 6.12 | 4.5  |
| CLS25    | 91         | 4.3             | 18.11    | 27.31    | 3.67           | 5.83    | 8.31 | 6.13 |
| CLS50    | 85         | 5.6             | 14.88    | 23.98    | 3.82           | 5.33    | 9.62 | 7.69 |
| CLS75    | 87         | 6.8             | 15.12    | 21.55    | 3.55           | 4.98    | 9.40 | 8.91 |
| CLS100   | 74         | 7.1             | 13.98    | 19.62    | 3.13           | 4.11    | 10.13 | 9.32 |

Table 9. Properties of the mixtures with 20% RHA.

| Mixtures  | Slump (mm) | Air content (%) | CS (MPa) | FS (MPa) | Absorption (%) |
|-----------|------------|-----------------|----------|----------|----------------|
|           |            |                 | 7 days   | 28 days  | 7 days         | 28 days |
| RF2-20RHA | 93         | 2.41            | 23.85    | 35.61    | 5.10           | 7.14    | 5.72 | 4.08 |
| CLS25-20RHA | 89       | 3.57            | 19.81    | 29.71    | 4.66           | 6.52    | 7.88 | 5.62 |
| CLS50-20RHA | 81       | 4.54            | 16.22    | 24.33    | 4.01           | 5.61    | 8.53 | 6.19 |
| CLS75-20RHA | 76       | 6.13            | 16.83    | 23.74    | 3.89           | 5.44    | 9.11 | 6.82 |
| CLS100-20RHA | 77       | 6.92            | 14.95    | 21.16    | 4.06           | 5.49    | 9.50 | 6.88 |
Table 10. Properties of the mixtures with 8% HRM.

| Mixtures      | Slump (mm) | Air content (%) | CS (MPa) 7 days 28 days | FS (MPa) 7 days 28 days | Absorption % 7 days 28 days |
|---------------|------------|-----------------|-------------------------|-------------------------|-----------------------------|
| RF2-8HRM      | 97         | 1.56            | 24.12 36.12             | 5.37 7.48               | 5.18 3.70                   |
| CLS25-8HRM    | 87         | 3.80            | 21.10 31.66             | 5.10 6.81               | 7.43 5.39                   |
| CLS50-8HRM    | 87         | 3.91            | 18.71 28.10             | 4.61 6.25               | 8.61 5.74                   |
| CLS75-8HRM    | 84         | 4.45            | 16.13 24.11             | 4.41 5.73               | 9.02 5.62                   |
| CLS100-8HRM   | 80         | 4.31            | 15.51 28.44             | 4.01 5.80               | 9.11 6.44                   |

The mean value of slump for the three groups of mixtures was 84.25mm, 80.75mm, and 84.50mm, respectively as shown in table 11. It was found that using CLS caused a reduction in a slump. The reduction was up to 19% when CLS increased from 25% to 100% for the first group of mixtures. The author's explanation for this reduction is the existence of high porosity in the CLS compared to the natural gravel which caused absorbing more water. On the other hand, the reductions in slump were 13% and 8% for mixtures with 20% RHA, and 8% HRM respectively. The air content increased whenever CLS content increased, the mean value of air content in the mixtures incorporating 8% HRM was less than the other mixtures as shown in table 11. Also, it can be deduced that the air contents decreased by 11% and 30% for the mixtures with 20% RHA, and with 8% HRM respectively compared to the mixtures without SCM. These results showed that the mixtures with 8% HRM were more effective in decreasing the porosity as well as improving the mechanical properties of the hardened concrete.

Table 11. Mean values of properties for each group of mixtures.

| Mixtures     | Slump (mm) | Air content (%) | CS (MPa) 7 days 28 days | FS (MPa) 7 days 28 days | Absorption % 7 days 28 days |
|--------------|------------|-----------------|-------------------------|-------------------------|-----------------------------|
| CLS          | 84.25      | 5.95            | 15.52 23.11             | 3.54 5.06               | 9.36 8.01                   |
| CLS+20RHA    | 80.75      | 5.29            | 16.74 24.73             | 4.15 5.81               | 8.75 6.37                   |
| CLS+8HRM     | 84.50      | 4.11            | 17.86 28.07             | 4.53 6.14               | 8.54 5.74                   |

Figures 1 and 2 show that the compressive strength is decreasing when CLS content is increased, and in relation to the results described in table 11, it is obvious that the mean compressive strength for each of the three groups of mixtures at the age 7 days decreased by 22% up to 26%. On the other hand, at the age of 28 days, it decreased by 10% up to 29%. It was observed that in each one of the three groups of mixtures, the compressive strength at 28 days decreased by 28% for the mixtures without CLS, 29% for the mixtures with 20% RHA, and 10% for the mixtures with 8% HRM when CLS content increased from 25% to 100%. The reduction in the compressive strength for the mixtures with 8% HRM was less distinguished than the other mixtures. The author's explanation for this reduction is the existence of the pozzolanic activity of HRM compared to RHA.
As shown in figures 3 and 4, the FS of mixtures with 8%HRM had relatively better values compared to the others. The addition of 20% RHA and 8%HRM increased the FS by 17% and 27% respectively at the age of 7 days.

Figure 1. 7 days CS.

Figure 2. 28 days CS.

As shown in figures 3 and 4, the FS of mixtures with 8%HRM had relatively better values compared to the others. The addition of 20% RHA and 8%HRM increased the FS by 17% and 27% respectively at the age of 7 days.

Figure 3. 7 days FS.
The FS at 28 days decreased by 29% for the mixtures without CLS, 16% for the mixtures with 20% RHA, and 14% for the mixtures with 8% RHA when CLS content increased from 25% to 100%.

The reduction in each of CS and FS for the mixtures using crushed limestone aggregate compared to the ordinary concrete may be attributed to the fact that the crushed limestone aggregate has low crushing strength compared to gravel as indicated in table 4.

The water absorption at ages 7 days and 28 days is shown in figures 5 and 6 for all mixtures. It is evident that the mixtures with 8%HRM have the lowest water absorption compared to other mixtures. Water absorption decreased by 6% and 8% for the mixtures with 20% RHA and 8%HRM respectively at the age of 7 days. In this study, the progress of the compressive strength was monitored using traditional methods (compression machine), however, recent studies used embedded sensors to have real-time measurements for the properties of concrete [56]. Therefore, the authors recommend the application of the EM sensors (electromagnetic sensors) for monitoring the mechanical properties of the limestone-contain concrete samples because the EM sensors showed high efficiencies in many fields [57-59].
4. Conclusions
According to the results of this study, it can be concluded that the coarse crushed limestone caused to reduce each of the compressive strength and flexural strength in concrete. That reduction in compressive strength and flexural strength was up to 30%. That crushed limestone concrete can be used for concrete structures for which the compressive strength is < 25 MPa and the flexural strength is <5 MPa. That the addition of supplementary cementitious materials SCM improved the performance of mixtures incorporating crushed limestone, it also caused to improve each of compressive strength and flexural strength up to 21%. That HRM showed high activity compared to RHA. That the water absorption increased due to the addition of crushed limestone but it decreased by about 20% and 28% in the mixtures containing rice husk ash and high reactivity metakaolin respectively at age of 28 days. That the water demand increased when CLS increased due to the porosity of CLS compared to gravel resulting in less workable mixtures. Finally, the addition of SCM improved the performance of the fresh mixtures with respect to the slump.

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