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Impact of organic carbon on hardened properties and durability of limestone cement concrete

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HIGHLIGHTS

- Total organic carbon (TOC) highly affects the properties of PLC concrete.
- The increase of TOC affects negatively the hydration process.
- Replacement of cement with 25% GLS decreases the corrosion rate of PLC concrete.
- Deterioration of PLC concrete in $MgSo_4$ solution increases with the increase of GLS.

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ABSTRACT

In this paper, the effect of organic carbon in ground limestone on the properties and behavior of Portland limestone cement (PLC) concrete in normal and aggressive medium was studied. Three different sources of limestone with total organic carbon of 0.94, 0.48 and 0.18% were considered, the cement replacement ratios were 0, 10, 15, 20 and 25% by weight and water to binder ratio was 0.55.

Mechanical properties, XRD and TGA experiments were conducted to evaluate the effect of the previous variables on hardened properties of concrete and hydration process of cement paste. Corrosion rate and polarization resistance of reinforcing PLC concrete specimens immersed in 5.0% sodium chloride solution for 12 months were measured using potentiodynamic polarization resistance technique. The sulfate resistance of PLC concrete specimens was also investigated. Concrete specimens exposed to wetting and drying cycles in 5% magnesium sulfate solution for 18 months were tested. SEMs of limestone cement paste after magnesium sulfate exposure were performed. The results emphasize the essential role of organic carbon on the properties and behavior of PLC concrete for both normal and aggressive medium especially at high ratios of cement replacement with limestone.

1. Introduction

The increase of the price of energy and the high amount of air pollutants released in cement manufacture make it necessary to search for sustainable solutions [1–3]. A lot of countries allow in their standards using Portland limestone cement to partially reduce the amount of clinker in cement. In 2012, ASTM C595 allows for Portland limestone cement containing 5%–15% limestone to cope with the needs of marketing and the environmental issues [2,4–7].

Limestone is a natural sedimentary rock composed of skeletal fragments of marine organisms. Limestone contains calcium carbonate ($CaCO_3$) as the principal constituent. Calcium carbonate occurs naturally in three crystal structure calcite, aragonite, and rarely vaterite [8]. During the geological formation of limestone layer, it may contaminate by soils or fragments which contain organic carbon. There is a lack of information about the effect of organic carbon of limestone powder on properties of PLC concrete. EN 197-1 identify a maximum total organic carbon (TOC) of 0.20% by weight for CEM II/A-LL and CEM II/B-LL, while, the maximum TOC for CEM II/A-L and CEM II/B-L has not to exceed 0.50% by weight [9,10].

In this paper, the effect of organic carbon on the hardened properties and durability of PLC concrete was investigated. Three sources of ground limestone of the same fineness and different contents of TOC (0.94, 0.48 and 0.18% by weight) were collected. Each source was blended by different ratios with ordinary Portland cement. Mechanical properties experiments were performed on concrete specimens to evaluate strengths developments, X-ray and TGA for cement paste was carried out to show the effect of TOC on the hydration process.

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Notation

PLC	Portland limestone cement	β_a	anodic Tafel slopes
GLS	ground limestone	β_c	cathodic Tafel slopes
C.R.	corrosion rate	I	electrical current
E_{corr}	corrosion potential	V	electrical potential
i_{corr}	corrosion current density	w/b	water/binder ratio
R_P	polarization resistance	S	source of limestone
B	Stern-Geary coefficient		

The behavior of PLC concrete with different sources of ground limestone exposed to 5% magnesium sulfate solution for 18 months were investigated through measuring concrete weight loss, compressive strength loss, expansion strain, visual inspection and microstructure observation. As well as, the corrosion rates of reinforcing concrete specimens immersed in 5% sodium chloride solution were measured by potentiodynamic polarization technique.

2. Experimental program

2.1. Materials

The materials used in this study were Portland cement, ground limestone (GLS), fine and coarse aggregates, reinforcing steel, super plasticizer. Portland cement (Type I) conforming to the ASTM C 150 standard. The chemical and physical properties of cement and limestone sources are presented in Tables 1 and 2. Siliceous sand and pink limestone aggregates with maximum size of 9.75 mm were used as a fine and coarse aggregates. The physical properties and the grading of aggregates are indicated in Table 3. The mechanical properties of reinforcing steel are listed in Table 4. Super plasticizer was added at the time of mixing, the dose of super plasticizer varied to achieve slump value of 8 ± 2 cm to all concrete mixes.

Table 1
Chemical composition and physical properties of Portland cement.

Chemical composition	Cement	Mineralogical composition (%)	(%)
Sio_2 (%)	19.9	C3S	58.22
Al_2O_3 (%)	5.1	C2S	16.48
Fe_2O_3 (%)	3.6	C3A	7.43
CaO (%)	62.6	C4AF	10.95
MgO (%)	2.7		
SO_3 (%)	2.9		
K_2O (%)	0.58		
Na_2O (%)	0.13		
LOI (%)	2.9		
Specific gravity	3.15		
Blaine surface area (m^2/kg)	340		

Table 2
Chemical composition and physical properties of ground limestone (GLS).

Chemical composition	GLS S 1	GLS S 2	GLS S 3
Sio_2 (%)	1.6	0.9	1.2
Al_2O_3 (%)	0.22	0.1	0.2
Fe_2O_3 (%)	0.19	0.1	0.13
CaO (%)	51.5	51.9	52.6
MgO (%)	1.23	1.03	1.13
SO_3 (%)	0.42	0.32	0.33
K_2O (%)	0.04	0.05	0.04
Na_2O (%)	0.18	0.11	0.12
LOI (%)	42.5	43.2	44.6
Specific gravity	2.72	2.7	2.71
Blaine surface area (m^2/kg)	347	340	344
Total organic carbon %	0. 94	0.48	0.18
Approximate $CaCO_3$ %	91.7	92.6	95.2
Color	Buff	Buff	White

2.2. Mix proportion and testing of specimens

Compressive strength tests were performed at 28 days on cubic specimens 150 * 150 * 150 mm, splitting tensile strength test was carried out at 28 days on 150 * 300 mm cylindrical concrete specimens according to ASTM C 496 – 96, and Static Modulus of Elasticity was conducted at 28 days on 150 * 300 mm cylindrical concrete specimens according to ASTM C 469.

The corrosion test was performed using specimens ∅ 75 * 150 mm concrete cylinders with embedded steel bars of 12 mm diameter. The steel bars were centered in the specimens so that the cover around the steel bar was 31 mm. The top of bars were painted with a double layer of epoxy resin to eliminate corrosion at free end. The specimens of corrosion test were immersed in 5% sodium chloride (NaCl) solution. The corrosion rate was measured using Gamry instrument Potentiostat/Galvanostat/ZRA through conducting potentiodynamic polarization technique. Test was performed using scanning potential of −250 mV through +250 mV, with a scan rate of 0.1 mV/s. The corrosion rate and polarization resistances testing methods were adapted from ASTM: G 59.

To evaluate the effect of magnesium sulfate on PLC concrete specimens, 15 concrete cubes 100 * 100 * 100 mm were prepared. After 28 days, SSD weights of cubes were measures, and five cubes were tested in compression. Then the specimens were monthly exposed to cycles of air drying and wetting in 5% magnesium sulfate solution for 18 months. Expansion strain tests were performed on concrete prisms of 75 * 75 * 285 mm according to ASTM C 1105. The initial length measurements were made after 28 days of casting, and then the specimens were continuously immersed in 5% magnesium sulfate solution for 18 months.

XRD, TGA, and SEM were performed on cement past specimens containing 0 and 10% ground limestone, all specimens were cured for 7 days followed by 21 days of air curing before testing. The SEM was performed on specimens after 18 months of 5% magnesium sulfate exposure at 20 °C. The mix proportions of one cubic meter of concrete mixes used in this research are listed in Table 5.

3. Test results and discussion

3.1. Effect of organic carbon on hardened properties of PLC concrete

3.1.1. Concrete compressive strength

Fig. 1 shows the compressive strength of PLC concrete made of three different sources of ground limestone, the cement replacement ratios were 0, 10, 15, 20, and 25%. From the figure, it is clear that S_3 achieves high improvement in compressive strength compared with S_1 and S_2, as an example, the use of 10% GLS ensures +3.0, −3.5, −10.0% increase in compressive strength for S_3 (TOC = 0.18%), S_2 (TOC = 0.48%) and S_1 (TOC = 0.18%) compared with the control specimen. The use of 20% cement replacement with S_3, S_2 and S_1 decreases the compressive strength by 7.0, 17.0 and 27.0% respectively. This shows that TOC has a considerable effect on concrete strength. TOC may delay or partially inhibit the hydration of cement through forming a thin coat around cement particles [7].

Compressive strength of PLC concretes with cement replacement of 0, 10, 15, 20, and 25% up to two years for GLS source (S_1) are presented in Fig. 2. The figure emphasizes that, the ratio between compressive strength at 2 years and 28 days compressive strength for PLC concrete is higher than that of the control specimen. As an example, the ratios of concrete compressive strength