

Research Article **Carbonation Coefficients from Concrete Made with High-Absorption Limestone Aggregate**

Eric I. Moreno

College of Engineering, Universidad Autonoma de Yucat ´ an, Avenida Industrias No Contaminantes s/n por Perif ´ erico Norte, M ´ erida, ´ YUC, CP 97110, Mexico

Correspondence should be addressed to Eric I. Moreno; emoreno@uady.mx

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Normal aggregates employed in concrete have absorption levels in the range of 0.2% to 4% for coarse aggregate and 0.2 to 2% for fine aggregate. However, some aggregates have absorption levels above these values. As the porosity of concrete is related to the porosity of both the cement paste and the aggregate and the carbonation rate is a function, among other things, of the porosity of the material, there is concern about the effect of this high porosity material in achieving good quality concrete from the durability point of view. Thus, the objective of this investigation was to study the carbonation rates of concrete specimens made with highabsorption limestone aggregate. Four different water/cement ratios were used, and cylindrical concrete specimens were exposed to accelerated carbonation. High porosity values were obtained for concrete specimens beyond the expected limits for durable concrete. However, carbonation coefficients related to normal quality concrete were obtained for the lowest water/cement ratio employed suggesting that durable concrete may be obtained with this material despite the high porosity.

1. Introduction

Concrete is the most used material for infrastructure development. However, durability of these structures is one of the main concerns, as the expected service life is approaching more than a hundred years. Among the known durability issues for concrete, corrosion of reinforcing steel is the main problem. In concrete structures, reinforcing steel is protected due to the high alkaline environment provided by the pore solution ($pH > 12.5$ units). However, it may corrode if the protection is lost. The protection may be lost due to chloride attack or due to the neutralization of the concrete pore solution surrounding the reinforcing steel. This neutralization of the pore solution is also called concrete carbonation. Concrete carbonation is the result of the reaction of hydrated cement compounds with atmospheric $CO₂$. Carbonation reduces the pH of the concrete pore solution (pH \approx 8 units), developing uniform corrosion in the reinforcing steel. In places with tropical climate, atmospheric conditions may encourage the $CO₂$ aggressiveness [1]. However, the carbonation rate depends on several factors [2] such as the type and amount of cement, porosity of the material [3], and amount of pozzolanic additions [4].

The porosity of concrete is related to the porosity of both the cement paste and the aggregate and the proper compaction during casting. The porosity of the cement paste is related to the water/cement ratio and the degree of hydration. From the durability point of view, it is accepted that the porosity of the cement paste is the one that controls the porosity of concrete. According to the literature [5], concrete's porosity above 15% is an indication of bad quality and below 10% is an indication of good quality.

Normal aggregates used in concrete have absorption levels in the range of 0.2% to 4% for coarse aggregate and 0.2 to 2% for fine aggregate [6]. However, some limestone aggregates have absorption levels above these values [7]. And due to the high-absorption characteristics of the aggregate, concrete made with this material has porosity values above 15%, thus suggesting bad quality concrete from the durability point of view.

One way to prove if this material is suitable for durable concrete structures is by determining the carbonation coefficient of the material. However, as concrete carbonation

proceeds at a very low rate, the use of an accelerated carbonation chamber is required.

Therefore, the objective of this investigation was to study the carbonation rates of concrete specimens made with highabsorption limestone aggregate.

2. Methodology

2.1. Specimens and Materials. Four water/cement (w/c) ratios were used (0.80, 0.70, 0.62, and 0.55). The cementitious material was Ordinary Portland Cement with no mineral additions. Crushed limestone of high absorption was used as coarse and fine aggregate (Table 1). Mixtures were designed according to the ACI specifications [8], and casting was made following the ASTM standards (Table 2) [9].

Concrete specimens were cylindrical in shape of different size: 300 mm by 150 mm for compressive strength tests, 200 mm by 100 mm for porosity tests, and 150 mm by 75 mm for carbonation tests. After casting, the curing period consisted of 28 days of water immersion. Once cured, the specimens were allowed to condition at lab environment prior to undergoing accelerated testing. The conditioning period was 80 days. Following conditioning, the specimens were placed in a 4% CO₂ environment in order to accelerate the carbonation process for ∼100 days, except for mixture 0.55 that was exposed for 176 days.

2.2. Carbonation Depth Measurements. The progress of the carbonation depth was determined by spraying a 1% phenolphthalein solution on top of a freshly broken concrete surface following the RILEM criteria [10]. For each determination, two specimens were removed from the carbonation chamber per w/c. After that, an ∼50 mm slice was split from each specimen using a chisel and a hammer. After applying the acid-base indicator, the measurements were performed using a digital caliper. Eight measurements were taken in the more suitable half cylinder as the cuts were irregular. Each measurement was corrected from radial measurements in a cylinder into carbonation depth measurements that would be obtained in a semi-infinite plane [3]. Once corrected, the average was obtained for each cylinder, and the result was averaged with the result from the other cylinder, obtaining an average from each pair of cylinders. After taking the carbonation measurements, the remaining portion of the concrete cylinders was placed back in the chamber.

In addition, a first carbonation depth measurement was performed using the external control specimens.This test was made after the conditioning period was finished and prior to placing the specimens in the carbonation chamber in order to determine the progress of the carbonation front during the conditioning period. In turn, the first carbonation depth measurement was used to correct the carbonation coefficient during the accelerated carbonation period.

3. Results

Table 3 shows the results from the compressive strength, porosity tests, and fresh concrete slump. The compressive

TABLE 1: Materials properties.

Material	Property	Value	
	Maximum size	3/4	
	Specific weight	2.4	
Coarse aggregate	Lose unit weight	$1,180 \text{ kg/m}^3$	
	Compact unit weight	1,290 kg/ m^3	
	Absorption	5.5 w/o	
	Specific weight	2.44	
Fine aggregate	Fineness modulus	2.6	
	Absorption	4.7 w/o	

TABLE 2: Mixture design (kg).

strength results were above the nominal values, particularly for 0.80 w/c mixture. The porosity values were high, however, in the expected range for the type of high-absorption aggregate used and with little difference among the different mixtures [7]. Concrete slump was in the expected range of 10 cm \pm 2; however, 0.55 w/c mixture had the smallest slump, and 0.62 w/c mixture had the highest.

Table 4 shows the results from the carbonation depth measurements. As the diameter was 75 mm, the specimens were fully carbonated when the carbonation depth was 37.5 mm. Due to the fact that mixture 0.55 was a better mixture than the others, carbonation depth was not determined at 51 days but at 112 days, instead. By day 51, mixtures 0.7 and 0.8 were fully carbonated.

4. Discussion

From the data in Table 4, the average carbonation coefficients at 4% CO₂ were estimated. The data is presented in Table 5. Usually, carbonation is modeled as

$$
x = k\sqrt{t},\tag{1}
$$

where t is the time of exposure, x is the carbonation depth at time t , and k is the carbonation coefficient. However, when initial carbonation exists previous to accelerated carbonation, then, the carbonation coefficient at 4% CO₂ may be obtained from [3]

$$
k_{4\%} = \sqrt{\frac{(x^2 - x_0^2)}{t}},
$$
 (2)

where t is the time of accelerated exposure, x is the carbonation depth at time t, and x_0 is the initial carbonation

w/c ratio	Designed strength MPa	Compressive strength MPa	Volumetric weight kg/m ³	Air $\%$	Slump cm	Porosity %
0.80		18.0	2,120	2.8	10	23.4
0.70	20	21.7	2,120	2.8	10	23.1
0.62	25	26.1	2,120	2.9		24.1
0.55	30	30.9	2,160	2.9		22.9

Table 3: Concrete properties.

TABLE 4: Carbonation depth measurements and standard deviations (mm).

Mixture	Days of exposure at 4% CO ₂							
	15		37		51		112	
	\mathcal{X}	σ	\mathcal{X}	σ	\mathcal{X}	σ	\mathcal{X}	σ
0.80	15.7	1.0	37.5	$\overline{}$	—			
0.70	13.7	1.6	24.2	1.4	37.5			
0.62	10.8	1.3	20.2	1.3	23.1	1.0	n.d.	
0.55	7.8	1.2	14.6	1.4	n.d.		22.3	1.3

Table 5: Carbonation coefficients from accelerated and natural exposure $\text{(mm/y}^{1/2})$.

depth measured after the conditioning period. The average carbonation coefficient was obtained from

$$
k_{\text{avg}} = \frac{\sum k_i \cdot (x_i - x_0)}{(\sum x_i - i \cdot x_0)}.
$$
 (3)

Carbonation coefficient has been modeled using the CO_2 diffusion coefficient in concrete (D) , the CO₂ concentration (c) , and the concentration of hydrated calcium compounds (M) [3]:

$$
k = \sqrt{\frac{2Dc}{M}}.\tag{4}
$$

In the event of experiments with two different external $CO₂$ concentrations ($c₁$, $c₂$) using separate specimens of the same concrete mixture, the time to reach a given carbonation depth x_c in both specimens would be, for concentration c_1 ,

$$
t_1 = x_c^2 \cdot \frac{M}{2Dc_1} \tag{5}
$$

and, for c_2 ,

$$
t_2 = x_c^2 \cdot \frac{M}{2Dc_2};\tag{6}
$$

thus, solving for x_c^2 ,

$$
x_c^2 = \frac{t_1 2Dc_1}{M} = \frac{t_2 2Dc_2}{M};
$$
 (7)

solving for t_1 ,

$$
t_1 = t_2 \frac{c_2}{c_1};\tag{8}
$$

from (4) and (7),

$$
x_c^2 = t_1 k_1^2 = t_2 k_2^2; \tag{9}
$$

substituting (8) in (9),

$$
t_2 \frac{c_2}{c_1} k_1^2 = t_2 k_2^2; \tag{10}
$$

solving for
$$
k_1
$$
,

$$
k_1 = k_2 \sqrt{\frac{c_1}{c_2}},\tag{11}
$$

and then, the average carbonation coefficients obtained in (3) were converted to atmospheric carbonation coefficient (k_{atm}) using (11):

$$
k_{\rm atm} = k_{\rm avg} \sqrt{\frac{c_{\rm atm}}{c_{4\%}}},\tag{12}
$$

where c_{atm} and $c_{4\%}$ are the atmospheric and 4% CO₂ concentrations, respectively.

The atmospheric carbonation coefficients are also presented in Table 5. According to the literature [5], carbonation coefficients above $6 \text{ mm/year}^{1/2}$ are representative of low quality concrete and carbonation coefficients below 3 mm/year $^{1/2}$ are representative of high quality concrete. The carbonation coefficients were above $6 \text{ mm/year}^{1/2}$ for specimens from mixtures 0.80, 0.70, and 0.62, suggesting low quality concrete from the durability point of view but as expected when using the w/c ratios employed. However, carbonation coefficients related to normal quality concrete were obtained for the lowest w/c ratio employed, suggesting that durable concrete may be obtained with this material despite the high porosity.

Thus, despite the high porosity of the concrete mixtures, enough compressive strength was obtained and the carbonation coefficients were consistent with the compressive strength's quality of the material.

5. Conclusions

- (i) High porosity values for concrete, above 22%, were obtained when using high-absorption aggregates.
- (ii) Designed compressive strength was achieved up to 30 MPa despite the use of high-absorption aggregates.
- (iii) Carbonation coefficients related to normal quality concrete were obtained for concrete mixtures of 0.55 water/cement ratio when using high-absorption aggregates.

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