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ACCELERATED CARBONATION OF ORDINARY PORTLAND CEMENT PASTE AND ITS EFFECTS ON MICROSTRUCTURE AND TRANSPORT PROPERTIES

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Abstract

Coupling of carbonation and chlorides ingress mechanisms is very common in concrete under certain exposure conditions such as coastal environments. The aggravation/ mitigation of corrosion by the existence of carbonation lies on the fact that microstructural changes due to carbonation result in changes on the transport properties of the material. In this study we investigate and quantify evolving transport properties of ordinary Portland cement paste, such as porosity, tortuosity and intrinsic permeability. Dual X-ray micro computed tomography (micro CT) is used for the quantification of porosity. Furthermore Dynamic Vapour Sorption (DVS) measurements are carried out to resolve water retention and relative permeability curves. The authors expect to provide insights into the mechanisms of accelerated carbonation in both types of cement paste, as well as data for input and validation of numerical and analytical models on this degradation phenomenon.
Keywords: transport properties cement, dual X-ray microcomputer tomography, DVS, carbonation, blended cement

1. INTRODUCTION

Concrete is the most common used material for construction all over the world [1]. However, the microstructure of concrete, which determines most of its properties such as strength and durability, does not remain constant over time. In fact, the microstructure of concrete still develops during its service life; however, deterioration due either to external or to internal factors can alter the microstructure properties as well, and consequently its durability. According to ACI Committee 201 [1], the durability of concrete is defined as its ability to withstand the deterioration process caused by its interaction with the environment.

Generally, all degradation mechanisms in concrete involve three main factors [2]. First of all, the presence of moisture, which is capable of caring aggressive ions within the concrete

and moreover needed for various internal concrete degradation processes themselves. Secondly, the rate of degradation largely depends on the transport properties of concrete, which in turn is strictly related to the pore structure of the hydrated cement paste such as porosity and connectivity of the pore network; lastly, the presence of aggressive ions: air, in fact contains carbon dioxide and oxygen, and the latter is required for the corrosion of the reinforcing steel. All these parameters are also capable of evolving with time, making the durability of concrete challenging to predict [3].

Nowadays, durability characteristics of construction materials are continuously gaining more importance, also due to its high economic impact on society. The repair and maintenance costs of existing structures require about 40% of the total resources of the construction industry and keep growing every year [4]. Carbonation is one of the degradation mechanisms responsible for the corrosion of reinforcing steel, and the latter is one of the major causes of concrete damage. It was reported by the U.S. Department of Transportation that 90 billion dollars were spent in 1991 for repairing damage caused by corrosion of reinforcing steel [4]. In summary, carbonation of the hardened Portland cement paste reduces the pH of the pore solution from approximately 13.5 to a value of about 9 [5], leading to removal of the steel passivation layer which covers and protect the steel rebar from corrosion. In addition, carbonation of Portland cement pastes alters its microstructure with consequent changes in transport properties which in turns may result in the initiation of other degradation phenomena.

Although carbonation in concrete has been widely studied experimentally, there is still the need for models that can correctly predict its evolution in real structures. For the development of such predictive models complete sets of data are needed. Unfortunately, the foci of existing experimental studies are not aligned with the need formulated earlier.

This study investigated the effects of the carbonation on the microstructure and transport properties of ordinary Portland cement pastes, and its evolution after carbonation. The main aim is to provide experimental data for further research as well as for modelling purposes.

2. METHODOLOGY

Ordinary Portland cement pastes were prepared with 0.6 water-binder ratio and CEM I 52.5R from ENCI, the Netherlands. The pastes were cast in polypropylene cylindrical moulds and sealed with plastic film and lids and rotated for 24 hours around their generatrix at 5-7 rpm as used in [6] to prevent bleeding. Subsequently they were stored at laboratory temperature (20°C) to cure. At age 90 days the samples where demoulded and cut into 1 cm side prims for preconditioning at the same temperature and 65% RH in order to reach the optimal internal condition for accelerated carbonation as suggested in [7].

Prior to the accelerated carbonation exposure, dual micro Computed X-ray Tomography (dual CT) as described in a previous paper from the authors [8], Thermogravimetric Analysis (TGA) and Dynamic Vapor Sorption (DVS), as described in a previous paper from the authors [9], were performed in order to assess porosity, hydrated phases composition, and water vapour sorption isotherms respectively. Moreover, by means of the water vapour sorption isotherms, the unsaturated permeability coefficient and capillary pressure curve were obtained. All the properties assessed directly or indirectly at this stage describe the noncarbonated state. Successively carbonation of the samples was done under accelerated condition at laboratory temperature (20°C) and 65% RH at a CO_2 concentration of 5%. All

the properties measured prior to carbonation are repeated after 22 days of accelerated exposure. At each carbonation step, the samples were taken out from the carbonation chamber and analysed as follows: dual scan to obtain the spatial distribution and magnitude of porosity, TGA for quantification of portlandite (CH) and carbonates content, and DVS to obtain water vapour sorption isotherms. Moreover, both TGA and DVS were performed at different depths of the sample from the exposed surface, namely 3, 6, 9 mm.

3. RESULTS AND DISCUSSIONS

The parameters of the water-retention and relative permeability curves were obtained by fitting the Van Genuchten and Mualem formulations [10, 11] (equation 1 and 2) with the data obtained from the DVS. Furthermore, from dual CT scans total and spatial distribution of porosity were resolved. These values are listed in Table 1 for the studied cement paste before and after 22 days of accelerated carbonation.

$$p_c(S_r) = a\left(S_r^{-1/m} - 1\right)^{1-m} \quad (1)$$

$$k_{rl}(S_r) = \sqrt{S_r}\left(1 - (1 - S_r^{1/m})^{m}\right)^2 \quad (2)$$

Table 1: Summary of measured properties before and after 22 days of accelerated carbonation

Property	Uncarbonated	Carbonated
Porosity [%]	50.1	41.3
Parameter ‘a’ from Van Genuchten [-]	73.3	66.47
Parameter ‘m’ from water-retention and relative permeability curves [-]	0.617	0.6

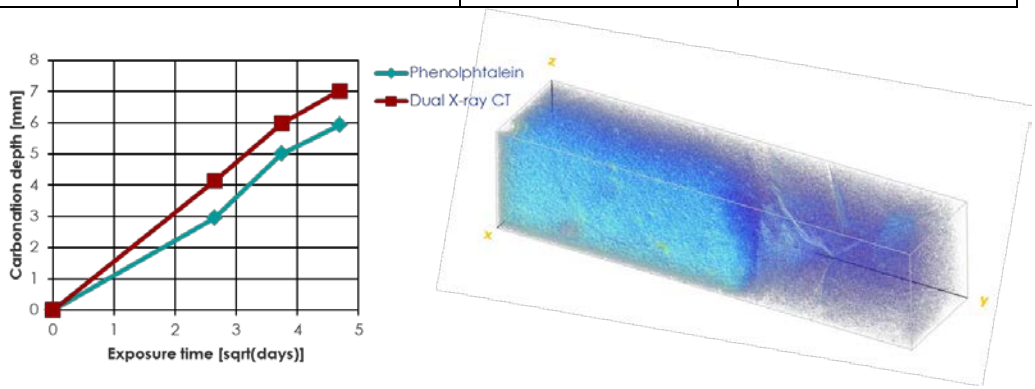

Figure 1: Carbonation depth as measured from dual CT in comparison to phenolftalein indicator.

It can be evinced from Table 1 that the dual CT methodology can quantify decreases in total porosity after carbonation, as well as locally, as shown in Figure 1. In the latter, it is

observed that there exists an underestimation of the carbonation depth, maybe due to the changes in pore structure at the carbonation front where pH has not been yet modified, similar findings were shown in [6] through TGA results and shown also in the TGA results shown in Figure 2. Regarding relative permeability, parameter m seems to change very literally which confirms what has been previously shown in literature for pastes with different water-to-cement ratio, regarding the independence of relative permeability from pore structure [12]. On the other hand, the parameter of the water-retention curve named 'a' decreases in value as found from DVS results.

Figure 2 shows the portlandite and carbonate contents at different depths from exposed surface in a 22 days carbonated paste.

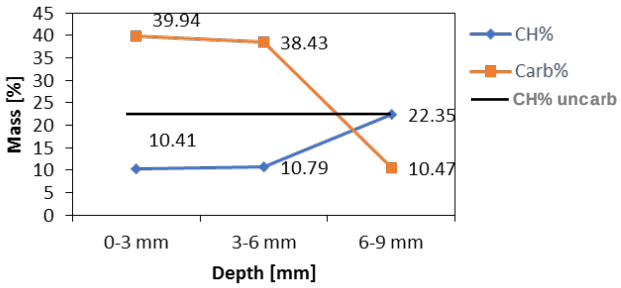

Figure 2: Portlandite and carbonate contents at different depths from exposed surface after 22 days.

It can be observed that after 22 days the carbonated area shows a nearly constant CH content approximately at 10.6%, whereas carbonates are not constant in the same area. This is an indication of the ongoing carbonation of the CSH gel after depletion of CH available for carbonation.

4. CONCLUSIONS

The following conclusions can be drawn out of the study presented herein:

- Dual CT scans can successfully be used to monitor local changes in porosity in cement paste samples.
- After accelerated carbonation the relative permeability function does not change but the water-retention curve parameter related to pore structure does decrease.
- Depletion of portlandite by carbonation was complete after 15-22 days of accelerated carbonation. Nevertheless, carbonates content are seen to vary within the carbonate region which indicates ongoing carbonation of CSH gel.

ACKNOWLEDGEMENTS

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