

On the Effects of Relative Humidity and CO₂ Concentration on Carbonation of Cement Pastes

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1 Introduction

Many environments to which concrete is exposed are highly aggressive due to various chemical components. In such environments, concrete is subjected to processes of chemical degradation, among which carbonation is one of the most frequently seen degradation processes. Though, the influence of saturation degree (or relative humidity - RH) of the specimen and CO₂ concentration on the carbonation of cementitious materials is still not comprehensively described with respect to carbonation rate/degree as well as alteration in microstructure and mineralogy. This work aims at thoroughly investigating how these two key parameters affect the carbonation under accelerated conditions. Furthermore, the effect of initial moisture state of the specimen on the carbonation rate is also demonstrated. For such purpose, a numerical model at continuum scale is developed to investigate the effects of RH and CO₂ concentration on the carbonation depth, phase changes in phases and porosity of hardened cement pastes due to carbonation under accelerated conditions.

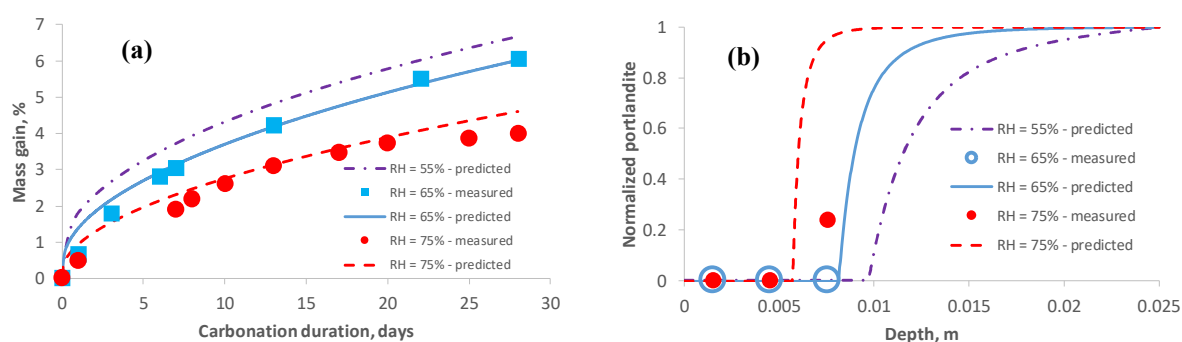


Figure 1. Effects of RH on mass gain (a) and change in portlandite content after 28 days of carbonation (b).

2 Model Development and Experimental Program

The proposed model is a one-dimensional reactive transport model coupling moisture – CO₂ – Ca transport, which is adapted from a previous work (Phung et al., 2016). The model enables to predict the carbonation degree, mass gain, portlandite content, C/S ratio and porosity change over time and space. The model only considers the carbonation of portlandite and C-S-H, which are the main hydrated phases in CEM I ordinary Portland cement (OPC) paste. Continuous hydration of the samples during carbonation is not taken into account as the experiments were

conducted on 28-day-old samples and the time for carbonation was quite limited. Accelerated carbonation experiments were performed on cement pastes at three w/c ratios of 0.4, 0.5 and 0.6. Type I OPC cement (CEM I 52.5 N) was used. The chemical and physical properties of the cement can be found in (Phung et al., 2019). Prior to the carbonation experiment, the samples were conditioned to different target internal relative humidity of 55%, 65% and 75% in a climate chamber. The accelerated carbonation tests were performed in 28 days at constant temperature of 20°C. Two CO₂ concentrations of 0.3% and 1% and three RH of 55%, 65% and 75% were investigated. After carbonation, a number of post-analysis methods including thermo-gravimetric analysis (TGA), mercury intrusion porosimetry (MIP) and phenolphthalein spraying were used to quantitatively and qualitatively analyse the carbonated samples.

A parametric study confirms that two key parameters, i.e. CO₂ concentration and RH, play an important role in accelerated carbonation of cement pastes. Variation of these parameters in a common testing range could significantly change the carbonation degree; however, their effects depend on the microstructure of the samples, which is controlled by the w/c ratios. The higher the w/c ratios, the larger the contribution of RH and CO₂ concentration as seen in **Figure 1**. The model helps to better interpret the experimental observations and highlights the underlying phenomena such as the formation of a gradual carbonation front due to carbonation of both portlandite and C-S-H (**Figure 2**), the changes of moisture state along the sample depth. Furthermore, the modelling results confirm the vital contribution of initial moisture state of the samples to carbonation process. Poor sample conditioning (i.e. too short time to lower the initial RH after curing to targeted testing RH) could dramatically reduce the carbonation efficiency.

3 Results and Discussion

Preliminary verification of modelling results with accelerated carbonation experiment gives a good agreement even though more experimental data is still required to validate and improve the model, especially to better capture changes in transport properties due to carbonation.

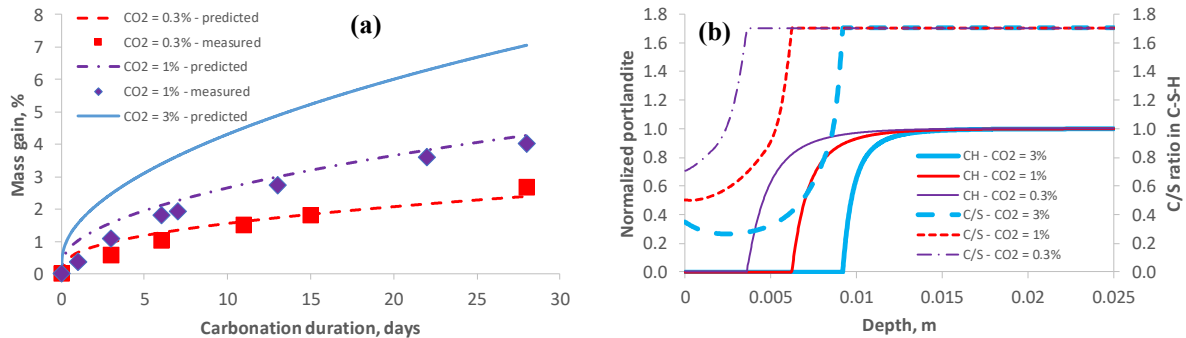


Figure 2. Effects of CO₂ concentration on mass gain (a) and changes in portlandite content and C/S ratio of C-S-H after 28-day carbonation (b) - samples with the same w/c ratio of 0.5 and carbonated under 65% RH.

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