

INFLUENCE OF EXPOSURE ENVIRONMENTS ON THE CARBONATION RESISTANCE OF CONCRETE STRUCTURES

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Abstract

There is agreement that the greenhouse gases, in term of CO₂ emissions, are the main contributor to global warming. Increasing atmospheric CO₂ emissions are likely to increase the average of maximum temperature and reduce the relative humidity, RH. These changes in CO₂ concentration, temperature and RH have considerable impacts on the durability of concrete structures, in particular, the carbonation rate, chloride penetration, and corrosion rate.

This study aims to investigate the potential impact of temperature, relative humidity and permeation properties on the carbonation resistance or depth of carbonation (DoC). The study programme involves casting samples with different water-cement ratio (w/c), 0.4, 0.5 and 0.6. These samples exposed to different temperatures of 25, 35 and 45°C and three levels of humidity 65, 75 and 85 %. Carbonation depth was determined using an accelerated environment test programme based experimentally on the [BS EN13295:2004](#) by phenolphthalein indicator. The results have indicated that (i): The depth of carbonation (DoC) increased with the increase in water-cement ratio under different exposure environments. (ii): There is a considerable influence of relative humidity and relative increase in temperature on the carbonation depth in concrete structures. Finally, the permeation properties for all mixes used of concrete, porosity and gas permeability has a significant impact of DoC.

Keywords: Carbonation, Phenolphthalein indicator, temperature, and Relative humidity.

1. INTRODUCTION

Carbonation is the chemical reaction between carbonic acid (H₂CO₃), resulting from a combination of atmospheric carbon dioxide (CO₂) with water, and calcium ion (Ca²⁺) from dissolution of hydrated cement products such as calcium hydroxide Ca(OH)₂, calcium silicate hydrates (C-S-H) and calcium aluminates hydrates (C-A-H). The calcium carbonate (CaCO₃) is formed from this reaction [1, 2]. The carbonation resistance of concrete structures is controlled by their capacity to delay ion and fluid transport inside concrete. The transport properties of fluid into concrete are represented through permeability and cracks. The permeability is influenced by variation of the material composition of concrete, such as quantity of cement, water cement ratio w/c, curing period, type of cement and type and dosage of chemical admixtures [1, 3]. The carbonation is a function of the pore system of the hardened cement paste due to the diffusion of CO₂ in concrete structures [1]. The second significant

factor affecting transport properties is cracks. The cracks in concrete structures are predictable in reinforced concrete members due to the weakness of tension capacity in concrete. Carbonation rates in the vicinity of the cracks are considerably higher due to the relatively faster penetration of CO₂ into the crack followed by orthogonal outward diffusion into the uncracked concrete surrounding the crack [4]. On the other hand, the exposure environments, carbon dioxide, temperature, and relative humidity, have a considerable impact on the carbonation rate of concrete structures [5]. Chi et al. [6] observed a linear correlation between the increase in the CO₂ concentration and the increase in the rate of carbonation. Roy et al. [7] concluded that there is a significant increase in carbonation rate when relative humidity, RH, ranges between 52-75%. However, there is a reduction in it as the relative humidity increases up to 84%. There is again an increase in carbonation depth; which begins at 84% RH upwardly until 92%. The maximum rate of carbonation in different mixes used in his study was found for a relative humidity level of 92%. It seems that the causes of this abnormal behaviour at the higher humidity level were unknown at that time. Russell et al. [8] demonstrated that the peak of the carbonation rate is reached at a relative humidity range of 55-65%. There are high variations in carbonation rate in the previous two studies [8, 9] for the same w/c ratio 0.55 and 0.7. Roy et al. [7] found the carbonation rates are 10.89 and 15.5(mm/y^{0.5}) respectively, while Russell et al. [9] got the carbonation rates are 32.3 and 79 (mm/y^{0.5}).

Finally, Drouet et al. [9] tested two hardened cement pastes using 100% CO₂ at different temperature and relative humidity (RH) levels and concluded that the impact of temperature on carbonation rate is dependent on the types of cement. It was also concluded that the decrease in RH causes an increase in carbonation rate. The changes in the mineralogy of cement products due to carbonation is not the same in high concentration CO₂ environment (10 - 100%). The results in a microstructure are not like those corresponding to natural carbonation at 0.03% of CO₂ [10].

Even though there is a chemical explanation about the temperature dependence of the diffusion coefficient based on an Arrhenius Equation [5,11,12], investigations on the effect of temperature on DoC scarce in the literature.

Consequently, the exposure environments, permeability, and cracks in concrete have influenced the diffusivity of aggressive species (CO₂) affecting durability. Therefore, it is important to study and consider how the exposure environments, the permeability and the presence of cracks may affect the rate of carbonation in concrete structures. Finally, the present paper examines the effect of external factors, temperature and RH, and internal parameters, porosity and cracks width on the depth of carbonation (DoC) what extent can this concrete resist the carbon dioxide attack.

2. EXPERIMENTAL WORK

2.1 Materials

Portland Limestone cement (CEM II/A-LL 32,5R) with a specific gravity of 3.05 was used in this study. Chemical and physical properties of the cement satisfy the BS EN 197-part 1: 2011. Natural sand was used as fine aggregates, and coarse aggregate was crushed gravel with a size range of 5-14 mm. The grain size analysis, chloride and sulphate content satisfy BS 882:1983. Deformed or ribbed steel (8mm) are used to reinforce the concrete prisms (to achieve the crack in samples).

2.2 Concrete Mixes Design

In order to achieve various properties of concrete, different water to cement ratios were used. Building Research Establishment method was employed to design the mixes used in this study. The mix proportions (water, cement, sand and gravel) were (205:513:653:980) kg/m³, (205:410:711:1023) kg/m³ and (205:350:711:1041) kg/m³ of concrete for mixes M 0.4 , M 0.5 and M 0.6 respectively. The mixing method is important to obtain the required workability and homogeneity of the concrete mix by mechanical mixing. The flexural method was used to induce the cracks, reinforced concrete prisms were used by fixing reinforcement in moulds with concrete cover 2 cm. Samples were cast in two layers. Each layer was vibrated using an electrical vibrating device to achieve the homogenous concrete and avoid the segregation of concrete. The specimens were demoulded and cured in a sink filled up with tap water until the time of testing or exposure to CO₂ environment condition at the age of 28 days.

2.3 Methodology

The main objective of the study is to investigate the effect of exposure environments, the permeability and crack width on the carbonation in concrete structures. Cracked concrete prisms were designed and exposed to these parameters of carbonation environment, temperature, and relative humidity. The flexural method was employed to induce the cracks in concrete samples. Four different crack width ranges, (0, 0.05-0.15mm, 0.15-0.25mm and 0.25-0.35mm) were applied to 100*100*500 mm concrete prisms. The crack width was measured by microscope meter with accuracy 0.01 mm. In this study, the one face of the specimens were exposed to accelerated environment conditions in a CO₂ incubator under the different scenarios. Three relative humidity level (65, 75 and 85) and three temperature degree (25, 35 and 45) have been separately used with 5% CO₂ for all series.

All samples were exposed to accelerated environment conditions in a CO₂ incubator with 5% concentration for 8 weeks. This concentration has the same mineral –change in the microstructure of cement due to carbonation to the natural condition [7]. After exposure, the samples were split by compression machine into two parts. The parts were sprayed by phenolphthalein solution with one gram of phenolphthalein powder and dissolved into a solution of 70 ml and 30 ml of ethanol and deionized water respectively (CEN/TS 12390-10:2007) to measure (DoC).

2.4 Concrete Samples Tests

For all specimens, compressive strength, porosity, gas permeability, and carbonation depth were measured. The porosity, gas permeability, and compressive strength test were performed in accordance with ASTM C642:2013, RILEM TC-116-PCD-1999 and BS EN 12390, part 3:2000 respectively by using 100 mm cube specimens. The test procedure is given in CEN/TS 12390-10:2007 was used to determine the DoC by phenolphthalein indicator using 100 × 100 × 500 mm prisms.

3. RESULTS AND DISCUSSION OF TESTS

In this section, the results of the experimental works are presented and discussed. These results and their discussions are mainly focused on three aspects; the impact of temperature and relative humidity with different w/c ratio and crack width on DoC.

3.1 Effect of w/c ratio and crack width on Gas permeability

Gas permeability test was used to evaluate the microstructure and permeation properties of concrete (e.g., permeability, sorptivity, and diffusivity) [13, 14]. The values of gas permeability coefficient or Darcy's factor, K for the various types of concrete are shown in Table 1. The K' increased with the increase w/c ratio and crack width, as presented in Table 3. The results are indicated that the K value increases by increasing the w/c ratio for sound and cracked concrete. This increase of K values for concrete results from the increase in the volume of connected voids in the microstructure of concrete or the porosity of concrete due to w/c ratio [15]. On the other hand, cracked samples can increase permeation properties that lead to increase gas permeability coefficient as well.

Table 1: Effect of crack width and types of mixes on gas permeability

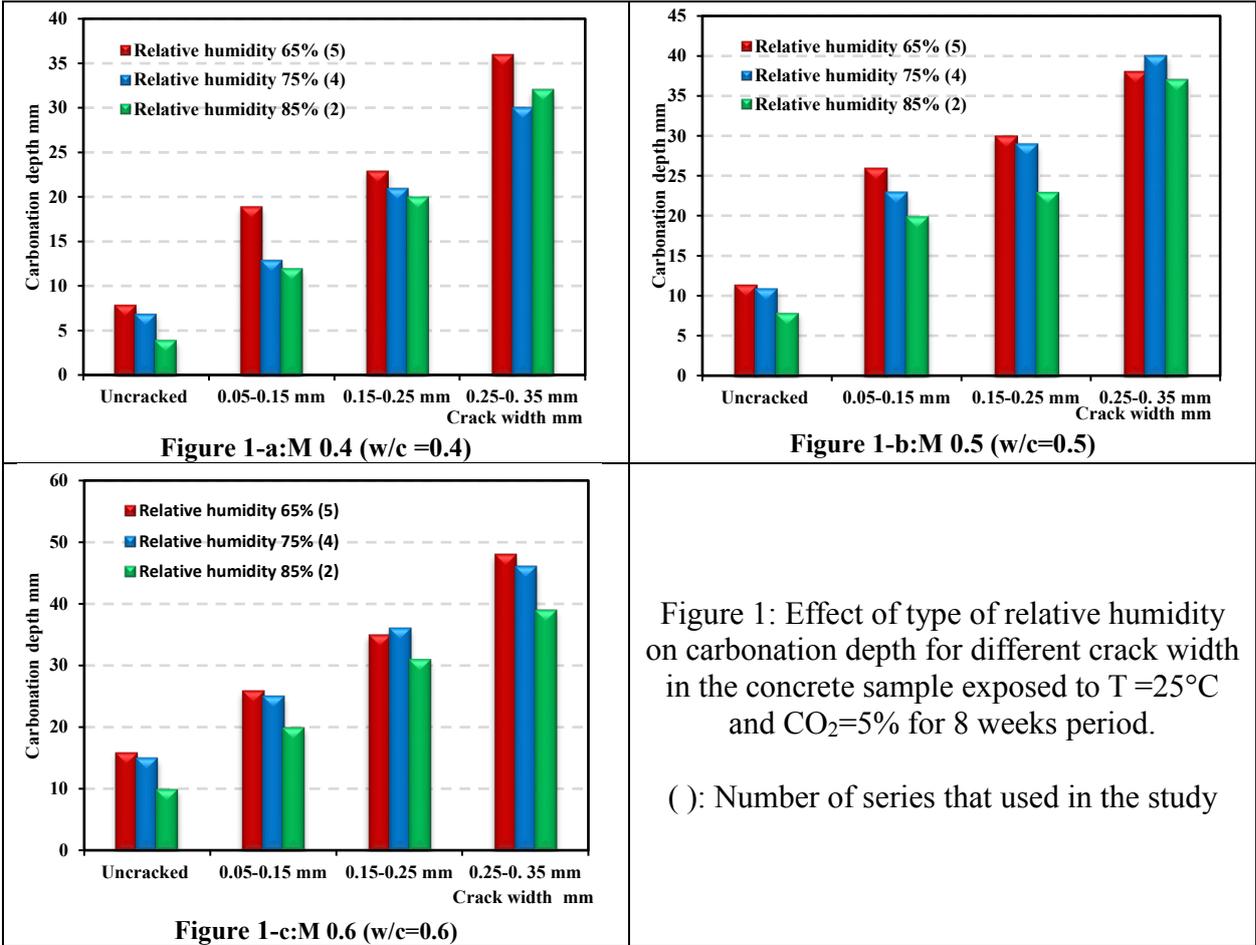
Crack width mm	Gas permeability coefficient, K, *10 ⁻¹⁷ , m ²		
	M 0.4	M 0.5	M 0.6
Un-cracked	6.1	7.1	10.6
0.05- 0.15	10.3	12.7	14.8
0.15 - 0.25	12.4	14.2	16.4
0.25- 0.35	12.6	14.7	16.7

3.2 Effect of relative humidity on Carbonation Depth in Concrete

The depth of carbonation (DoC) is a measure of diffusivity of carbon dioxide in concrete and reduction of alkalinity of pore water solution in concrete. The phenolphthalein solution was used to indicate DoC (CEN/TS 12390-10:2007). It is sprayed on concrete. In non-carbonated parts of concrete, the colour of these parts ranges from purple to red, due to concrete is still highly alkaline pH is greater than 9. While, the carbonated parts of concrete, the alkalinity is reduced, no colouration happened due to converting the Ca (OH)₂ to calcium carbonate by reacting with carbonate ion (CO₃⁺²). These samples have been exposed to three targets of relative humidity, 65 %, 75% and 85% with accelerated CO₂ environment 5% and temperature 25°C. The DoCs of all cases with different cracks width over the 8-week period are illustrated in Figure 1 (a, b and c) and presented samples are shown in Figure 2(a, b and c). The overall trend in these figures is that of decreasing DoC with increasing RH. However, the DoC does not appear to change at a constant rate over the range of RH investigated. For almost all of the samples with different cracks width and w/c, the maximum carbonation depth occurred at 65% target RH. On the other hand, there are two cases unlike the general trend due to other factors might have influenced the DoC at this RH values such as crack width. Also, the results indicated the (DoC) in concrete increases with increasing w/c ratio with different RH and crack width, the percentage increases in (DoC) for samples (sound and cracked) with respect to the control concrete samples (M 0.4 mixes) are presented in Table 2.

Also, the increase of w/c ratio leads also to increase the porosity in concrete and decrease compressive strength of concrete. According to Roy et al. [8], the rate of carbonation is inversely proportional to the strength. The porosity and compressive strength of concrete have a significant impact on carbonation depth.

The results show the RH affects the DoC, the effect is not the same for all the mixes investigated. The effect of w/c ratio on DoC is the highest at 65% RH and is the least at 85% RH. This can be attributed to the fact that the pores are free of moisture at low RH, i.e. the impact of open porosity is gotten at the low RH. Overall the tendency of declining DoC with increasing RH within this range is like that which could be expected. However, what the results show is that there is a wide range of variability depending on the mix proportions and the quality of the concrete [8,9].



The increase of (DoC) for concrete mixes with increasing the w/c ratio and crack width. That is a result the rise the porosity of concrete and volume of permeable voids which helps CO_2 to penetrate and react with water to form carbonate acid. The latter reacts with $\text{Ca}(\text{OH})_2$ (aq) to occur the carbonation [4,15] as shown in Figure 3. Furthermore, the fundamental influence controlling carbonation rate is the diffusivity of the hardened cement paste, which is a function of the pore system of the hardened cement paste during the period when the diffusion of CO_2 takes place [1].

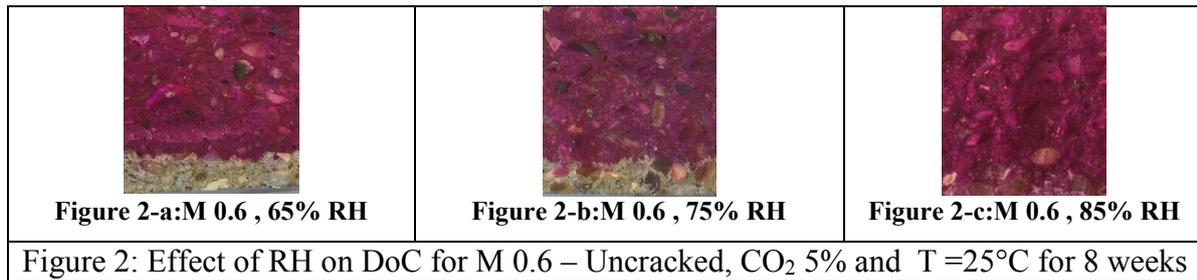
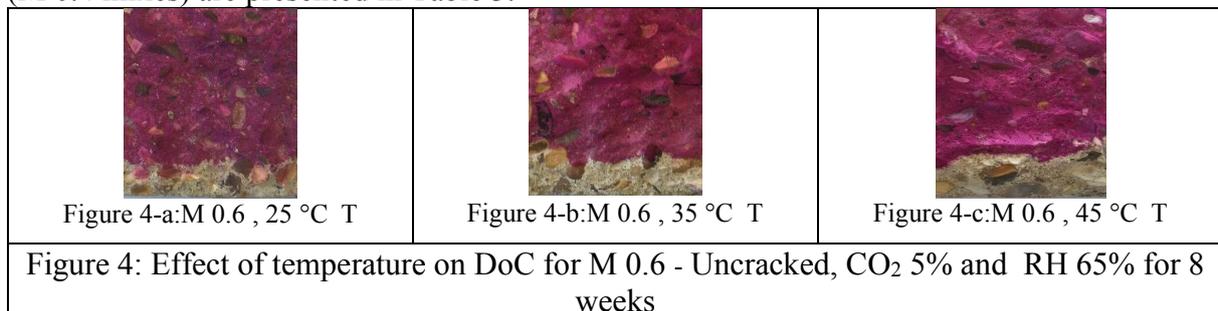


Table 2: Increase percentage in DoC with increase w/c ratio

Crack width mm	DoC of (M 0.5/ M 0.4) %			DoC of (M 0.6/ M 0.4) %		
	65% RH	75% RH	85% RH	65% RH	75% RH	85% RH
0	44	57	100	100	114	150
0.05- 0.15	37	77	67	37	92	67
0.15 - 0.25	30	38	15	52	71	55
0.25- 0.35	6	33	16	33	53	22

3.3 Effect of Temperature on Carbonation Depth in Concrete

The effect of the temperature on the DoC for M0.4, M0.5 and M0.6 for CO₂ 5% and RH 65% for 8 weeks, are presented in Figure 4. The samples were exposed to three different temperatures, 25°C, 35°C, and 45°C. Samples exposed to these conditions showed that the (DoC) increases with the temperature. Also, it was noted that when the samples were exposed to accelerated CO₂ condition at 45°C, the higher the w/c ratio in the samples' mix design, the less resistance of carbonation was found as shown in Figure 5 (a, b and c). Exposing at elevated temperatures altered the mineral composition of cement products due to carbonation and increased its depth in samples, but performance some of the cracked samples were not consistent with the un-cracked samples results. The results also show that the DoC in concrete increases with increasing w/c ratio with different temperatures and crack width, the percentage increases in DoC for samples (sound and cracked) with respect to the control concrete samples (M 0.4 mixes) are presented in Table 3.



The effect of accelerating CO₂ condition with increase temperature encourages the diffusion of carbonate ion into concrete, due to amplified molecular activity and an increase in the reaction rate between carbonate ion CO₃⁻² and Ca²⁺ resulting from dissolve of H₂CO₃ and Ca (OH)₂ respectively in the pore water of concrete and forms CaCO₃ [11,12].

The main reason for the increase of DoC for concrete mixes with increasing the w/c ratio and crack width is the same reason as mentioned in the previous section. On the other hand, the increase of crack width was an important factor of CO₂ penetration, is deeper and reacting with Ca(OH)₂ to form carbonation compound, CaCO₃.

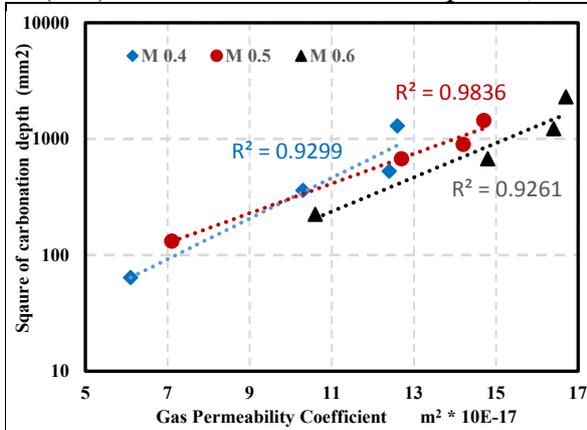


Figure 3: DoC in 5% CO₂, 65% RH and 25 °C as a function of concrete gas permeability

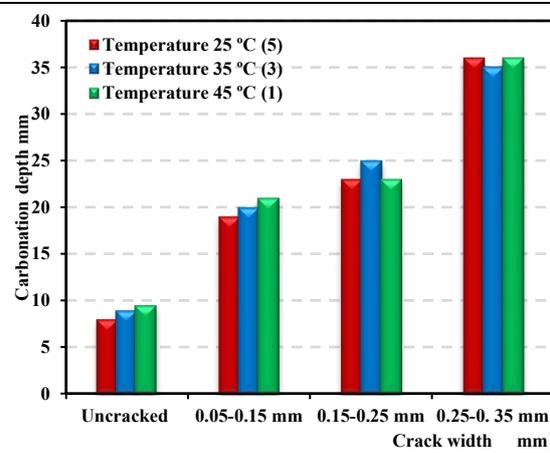


Figure 5- a:M 0.4

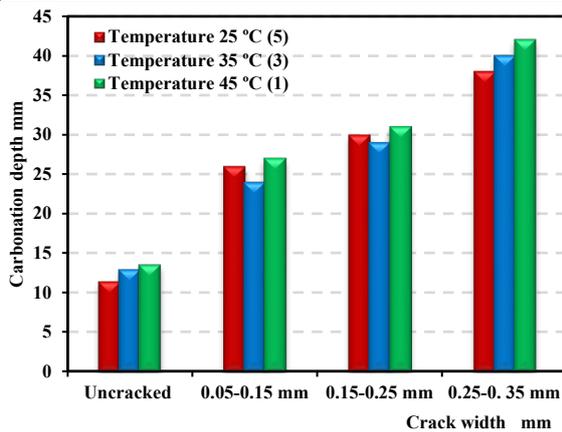


Figure 5-b:M 0.5

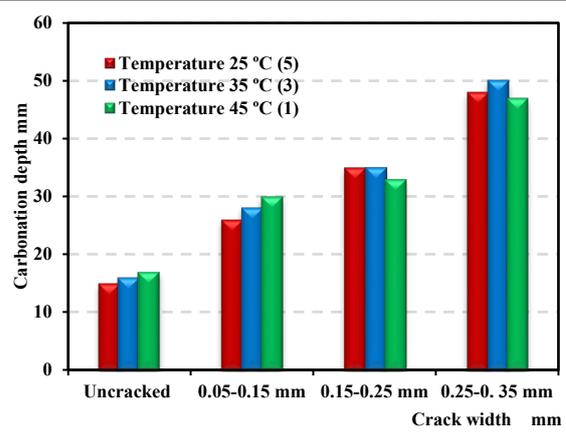


Figure 5-c: M 0.6

Figure 5: Effect of type of temperature on carbonation depth for different crack width in the concrete sample exposed to RH =65% and CO₂=5% for 8 weeks period

Table 3: Increase percentage in DoC with increase w/c ratio with temperature (T)

Crack width mm	DoC of (M 0.5/ M 0.4) %			DoC of (M 0.6/ M 0.4) %		
	T = 25 °C	T = 35 °C	T = 45 °C	T = 25 °C	T = 35 °C	T = 45 °C
0	44	44	42	88	78	79
0.05- 0.15	37	20	29	37	40	43
0.15 - 0.25	30	16	35	52	40	43
0.25- 0.35	6	14	17	33	43	31

3. CONCLUSION

This study has considered the influence of exposure environment and properties of concrete on the DoC in concrete structures. The effect of three level of RH, temperatures and w/c ratio on

DoC have been investigated by accelerated CO₂ environment condition. The following conclusions can be drawn from the results; -

- For samples exposed to an accelerated CO₂ environment, increase in RH led to a reduction in the DoC.
- The increment in the temperature increased the DoC due to relatively fast penetration of CO₂ into the concrete and an increase in the reaction rate with cement products, which led to the formation of the carbonation compound CaCO₃.
- The DoCs in concrete samples are influenced by material behaviours such as the permeation properties (permeability and gas permeability) and compressive strength due to the impact of diffusivity of CO₂.
- The crack width significantly increased the DoC for mixes with different RH and temperature.

REFERENCES

- [1] Neville, A.M., ' Properties of concrete', Five and Truly Final Edn, (Pearson Education Limited, London,2011).
- [2] Tarun R. and Rakesh, K., 'Global warming and cement-based materials', (UWM Center for By-Products Utilization, Milwaukee, Wisconsin USA,2010).
- [3] AL-Khaiat, H., Haque, M. N., and Fattuhi, N., 'Concrete Carbonation in Arid Climate'. 29th Conference on our world in concrete & structure. Singapore, 2004.
- [4] AL-Ameeri, A., Rafiq, M., Tsioulou, O. 'Influence of cracks on the carbonation resistance of concrete structures', Proceedings of a Sixth International Conference on the Durability of Concrete Structures, Leeds, July 2018(University of Leeds) 358-367.
- [5] Dyer, A., 'Concrete durability', (Taylor& Francis Group, LLC, New York,2014).
- [6] Chi, J. M., Huang, R.and Yang, C. C., 'Effect of Carbonation on Mechanical Properties and Durability of Concrete Using Accelerating Testing Method', *Journal of Marine Science and Technology*, 10(1) (2002)14-20.
- [7] Roy, S, Poh, K and Northwood, D., 'Durability of concrete accelerated carbonation and weathering studies', *Building and Environment*, 34 (5) (1999) 486-595.
- [8] Russell, D., Basheer, M., Rankin, G., and Long, A., Effect of relative humidity and air permeability on the prediction of the rate of carbonation of concrete. Proceedings of the Institution of Civil Engineers - Structures and Buildings, 146 (3) (2001) 319-326.
- [9] Drouet, E., Poyet, S., Le Bescop, P., Torrenti, J.M. and Bourbon, X., 'Carbonation of hardened cement pastes: Influence of temperature'. *Cement and Concrete Research*, 115(2019) 445-459.
- [10] Castellote, M., Lorenzo Fernandez, L., Andrade, C., Alonso, C., 'Chemical changes and phase analysis of OPC pastes carbonated at different CO₂ concentrations'. *Materials and Structures*, Vol. 42(2009) 515–525.
- [11] Yoon, I., Copuroglu, O.and Park K., ' Effect of global climatic change on carbonation progress of concrete', *Atmospheric Environment*, 41(34) (2007)7274–7285.
- [12] Talukdar, S., Banthia, N., and Grace J., 'Carbonation in concrete infrastructure in the context of global climate change: Part 1 – Experimental results and model development', *Cement and Concrete Composites*, 34(8)(2012)924– 930.
- [13] Yang, K., Basheer, P.A.M., Magee, B., Bai, Y. and Long, A.E., Repeatability and reliability of new air and water permeability tests for assessing the durability of high-performance concrete. *Journal of Materials in Civil Engineering*, 27(12), (2015) 04015057.
- [14] Hilsdorf, H. and Kropp, J., Performance criteria for concrete durability. 2014, CRC Press.
- [15] RILEM TC 116-PCD-1999'measurement of the gas permeability of concrete - Cembureau Method', *Materials and Structures/Matériaux et Constructions*, Vol. 32, (April 1999) 174-179.