



Carbonation resistance of cements containing supplementary cementitious materials and its relation to various parameters of concrete



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HIGHLIGHTS

- Carbonation of cement containing supplementary cementitious materials was studied.
- Linear relation was observed between accelerated and natural carbonation coefficients.
- Reserve alkalinity was found to be the major factor governing rate of carbonation.
- No direct correlation was observed between carbonation depth and strength.
- Empirical relation is proposed to predict carbonation depth.

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ABSTRACT

In this study, the carbonation resistance of cements containing supplementary cementitious materials (SCMs) exposed to carbon dioxide concentrations of 1% and 3%, relative humidity of 40%, 60% and 80% and temperature of 27 °C and 45 °C at two different water to cement ratio is investigated. The carbonation performance of concrete samples placed in accelerated conditions is compared to that of concrete samples exposed to natural conditions. The comparison of carbonation coefficients of concrete exposed to different environmental conditions indicates that the rate of carbonation is primarily governed by the extent of clinker replacement, relative humidity and water to cement ratio. Concrete mixes with slag showed better carbonation resistance as compared to mixes containing other SCMs. No specific correlation can be observed between the carbonation coefficients and physical parameters of concrete (compressive strength and porosity), however, a reasonable correlation is observed between carbonation coefficients and chemical parameters (Total alkali content, reserve alkalinity and pH). An empirical equation is developed to estimate the carbonation depth for different cements for wide range of exposure conditions.

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1. Introduction

Corrosion of reinforcement is a major cause of deterioration of concrete structures. The concentration gradient between the concrete and the external environment leads to diffusion of carbon dioxide in concrete. Carbon dioxide reacts with the hydration phases of the cement resulting in a reduction of alkalinity of concrete. At reduced alkalinity, the passive oxide layer protecting reinforcement in concrete starts to disintegrate, making it prone to corrosion [1,2].

SCMs are used as clinker replacement, in order to reduce the carbon dioxide emissions and overall energy requirement of cement plants. SCMs replace part of clinker and aid in obtaining

a cement with properties better or similar to that of Ordinary Portland Cement (OPC). Fly ash and slag are the two most widely used SCMs apart from silica fume, metakaolin, rice husk ash and natural pozzolanas to produce blended cements. The rate of carbonation in concrete containing SCMs is generally higher than OPC due to lower calcium hydroxide content in them [3–8]. Branch et al. (2016) [9] studied the impact of alkalinity on carbonation by replacing OPC with different fly ashes by assessing the changes in microstructure and migration of its constituents. The rate and capacity of carbonation reaction was found to be inversely proportional to the alkalinity of the system. The rate of carbonation is generally slower in high alkali cement as the carbon dioxide binding capacity of the cement is more [10]. The lower alkalinity of blended cements can be compensated by the lower permeability of their cement pastes obtained from proper curing [11]. However,

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reduction in calcium hydroxide content dominates over the pore refinement [5].

The rate of carbonation in concrete exposed to the natural environment is slow because of low concentration of carbon dioxide in the atmosphere. The concentration of carbon dioxide in the environment varies from 0.03% in a rural environment to 0.1% in urban areas [12]. Accelerated carbonation tests having higher carbon dioxide concentrations are generally adopted to evaluate the performance of concrete structures under carbonation in shorter duration. Carbon dioxide concentrations between 0.03% and 100% have been used in previous studies to study the carbonation process [2,13–19].

The carbonation coefficient of concrete samples exposed to natural conditions can be determined from the accelerated carbonation tests on concrete samples. A square root relationship between time and carbonation depth has been reported [18,20–23]. Based on accelerated carbonation coefficient the carbonation depth in samples exposed to natural exposure conditions can be predicted using Eq. (1) [22,23].

$$\frac{K_{\text{acc}}}{K_{\text{nat}}} = \frac{\sqrt{C_{\text{acc}}}}{\sqrt{C_{\text{nat}}}} \quad (1)$$

where K_{acc} is the accelerated carbonation coefficient, K_{nat} is the natural carbonation coefficient, C_{acc} is the carbon dioxide concentration in accelerated test and C_{nat} is the carbon dioxide concentration in natural exposure condition. Several authors based on the above empirical equation have reported that the accelerated carbonation test carried out at 4% carbon dioxide concentration for one week is equivalent to exposure in the natural atmosphere for one year [11,20]. However, the above notion cannot be generalized as the rate of carbonation will vary with the type of concrete, preconditioning of samples before exposure to carbonation and environmental conditions.

The rate of carbonation in concrete is governed by numerous physical and chemical parameters of the concrete. The carbonation resistance of a particular type of cement can be correlated with a certain parameter, however, the correlation of the same parameter with rate of carbonation for different types of cement is generally weak. Therefore, identifying parameters that primarily govern the rate of carbonation in different types of cement are important. Also, accurate estimation of natural carbonation coefficients from accelerated carbonation test is vital to precisely predict the extent of carbonation in actual concrete structures.

2. Methodology and experiments

2.1. Materials

In this study, three types of binder systems were investigated for carbonation performance. Clinker, limestone and gypsum used in the study were procured from a cement plant based in Gujarat, India. Fly ash was procured from National Thermal Power Plant Corporation (NTPC), Dadri, India whereas processed slag was obtained from Goa, India. The purity of limestone and gypsum used was approximately 80%. The fly ash used can be classified as *Class F* fly ash according to ASTM C618 2015 [24] it contains at least 70% pozzolanic compounds (silica oxide + alumina oxide + iron oxide). Two clays with kaolinite content of approximately 60% and 80% were used in the study. Clay 1 was procured directly from a clay mine and was calcined in a rotary calciner whereas clay 2 was a commercially available metakaolin. River sand was used as fine aggregate whereas crushed stones were used as coarse aggregate. The fine aggregate used falls within the limits of zone II, described in Indian Standard IS 383 [25], approximately 90% and 50% particles were finer than 4.75 mm and 600 μm respectively. The chem-

ical composition of raw materials measured using X-Ray fluorescence (XRF) is given in Table 1.

Binary binders comprising of OPC/Fly Ash (B1) and OPC/Slag (B2), ternary binders comprising of OPC/Calcined Clay/Limestone (T1 & T2) and OPC/Slag/Fly Ash (T3) and control blend OPC (U1) were studied. The proportion of the raw materials mixed to prepare different binders is given in Table 2. OPC was prepared by inter-grinding clinker and gypsum in a laboratory ball mill with charge to the material ratio of 3:1. T1 (Limestone calcined clay cement – LC³) was produced in an industrial trial production, details of which are given elsewhere [26]. All the remaining blends were produced by interblending the raw materials with OPC in a ball mill without charge.

2.2. Sample preparation and exposure conditions

Concrete specimens were cast at water to cement ratio of 0.35 and 0.45 according to the guidelines described in Indian Standard IS 456 [27] and IS 10262 [28]. The total water content in concrete was kept fixed at 162 kg/m³ for all the mixes. Coarse aggregates conforming to the nominal size of 10 and 20 mm in a proportion of 1.00:1.50 of the total weight of the coarse aggregate were used in preparing concrete. Coarse aggregates were used in saturated surface dry (SSD) condition whereas moisture correction was applied for fine aggregates. Polycarboxylic ether (PCE) based water reducing admixture was used to improve the workability of concrete having specific gravity of 1.10 and a solid content of 40% was used in this study. The dosage of admixture was adjusted to get a slump of 140 \pm 20 mm. The details of the mix proportion of concrete are provided in Table 3. Concrete was mixed in a pan mixer. Cube (150 \times 150 \times 150 mm³), cylinder (Diameter: 100 mm, Height: 200 mm) and prism (500 \times 100 \times 100 mm³) specimens were cast. Table vibrator was used to ensure proper compaction. Thereafter, the concrete specimens were left in the casting area maintained at 27 \pm 2 $^{\circ}\text{C}$ and were demolded 24 h after casting. After demolding, the specimens were cured in lime-saturated water for 120 days.

Cement paste samples were also prepared by mixing the raw material in the same proportion as given in Table 2 at a water to cement ratio of 0.35 and 0.45 in cylindrical plastic molds having a diameter of 20 mm and height of 100 mm. Cement paste samples were demolded 24 h after casting and were cured in lime-saturated water for 120 days.

At the end of the curing regime, standard epoxy adhesive was applied on the square bases of the prism specimens after mixing resin and hardener in equal proportion. Fig. 1 shows the epoxy coated prism samples. Subsequently, all the concrete specimens were left for preconditioning for 15 days at a temperature of 27 \pm 2 $^{\circ}\text{C}$ and relative humidity of 60 \pm 5%. The cylindrical concrete specimens were cut into discs of length 50 \pm 3 mm using a diamond saw cutter. Top 20 mm from the cylinder was discarded while slicing as it might not have representative composition of the concrete.

After the end of the preconditioning period, the concrete specimens were placed in different exposure conditions. In this study, carbon dioxide concentration, relative humidity and temperature were varied to understand the carbonation process. The different exposure conditions adopted are given in Table 4. The carbon dioxide concentrations of 1% and 3% were used as it has been reported that in these conditions the microstructure of specimens after carbonation is similar to that of specimens carbonated in natural conditions [13,29–31]. The specimens for accelerated carbonation tests were placed in an environment controlled carbonation chamber. Samples exposed to the natural environment were placed in two different sets of conditions. The first set of samples was sheltered in a box with louvers fixed on all four sides of the box at an

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