Abstract: With the development of modern construction technology, Reinforced Concrete (RC) has become the most widely used composite construction material. Though RC possess many thriving properties, it has certain weaknesses as well. Corrosion observed in RC structures, which is basically the outcome of concrete carbonation and chloride attack is considered to be one such weakness. The objectives of this research are to evaluate the effectiveness of parameters that dominate the carbonation induced corrosion and to derive an equation for the coefficient carbonation (K). Hence, all the prominent factors affecting the carbonation process were identified. Then, the behaviour of each factor along with K was identified by conducting an Accelerated Carbonation Test (ACT). Following this, experimentally derived K values were used in deriving a statistical equation for predicting K of structures.

Keywords: Corrosion, reinforced concrete, coefficient of carbonation, accelerated carbonation test.

1. INTRODUCTION

Ever since the first industrial revolution, construction industry has undergone a rapid development. Following this construction boom, several construction materials were introduced to the industry. Amid the pile of construction materials, Reinforced Concrete (RC) is the most widely used composite construction material. High life expectancy of RC structures is one of the main reasons behind this. However, corrosion of embedded steel reinforcement in RC structures is one of the pioneering mechanisms leading to this rapid service life depletion. Generally, steel reinforcements embedded in concrete are protected against corrosion, by the high alkaline environment formed by the surrounding concrete. Ca(OH)₂ produced within RC structures due to cement hydration reactions, is considered to be one such prominent compound involved in this mechanism. Roy et al (1999) states that, so formed Ca(OH)₂ forms a passive protective iron oxide film around the steel reinforcement bars, leaving it in an environment at or above a pH value of 10.5. This pH environment acts as a chemical protection against reinforcement de-passivation (Pushpakumara, et al., 2013).

However, corrosion occurs when this passive protection is disturbed by the two processes, namely chloride ion ingress and concrete carbonation. When compared to chloride ingress, concrete carbonation is basically a slow deterioration process (Papadakis, et al., 1989). But, with the rapid industrialization occurring worldwide, contribution of concrete carbonation on service life depletion of RC structures has increased at an alarming rate. Basically, in this process alkalinity of the pore water solution decreases, forming an acidic environment which is favourable for corrosion of reinforcement steel. Once this carbonation depth is equal to the cover depth of the reinforcement bar, it causes to corrode the bar and then subsequently deteriorate the structure.

Concrete carbonation could be simply described as the physicochemical process in which atmospheric carbon dioxide gas reacts with alkaline constituents in concrete to form carbonates such that reducing the pH value of concrete samples. Main carbonation reactions involved in this process could be summarized as below,

\[
\begin{align*}
\text{Ca(OH)}_2 + \text{CO}_2 & \rightarrow \text{CaCO}_3 + \text{H}_2\text{O} \\
3\text{CaO} - 2\text{SiO}_2 - 3\text{H}_2\text{O} + 3\text{CO}_2 & \rightarrow 3\text{CaCO}_3 \cdot 2\text{SiO}_2 \cdot 3\text{H}_2\text{O}
\end{align*}
\]
In these reactions carbon dioxide gas is considered to be in the aqueous state. Apart from these, many subsidiary reactions, involving reactions of tricalcium silicate and dicalcium silicate with aqueous carbon dioxide gas too influence in the carbonation process (Peter, et al., 2008).

Even though concrete carbonation appears to be a simple chemical reaction between calcium hydroxide and atmospheric carbon dioxide gas, it is basically a broad process affected by many other environmental and physical factors of the concrete structures. Papadakis et al (1989), recognize these factors as pertinent material parameters and environmental conditions. There, authors refer composition of cement and concrete, porosity and pore size distribution as pertinent material parameters and atmospheric carbon dioxide gas concentration, humidity and temperature as environmental conditions. Similarly Jiang et al (2000), defines content of binder, water/binder ratio, degree of hydration, concentration of CO₂ gas and relative humidity (RH) as main factors affecting rate of concrete carbonation. Apart from these, influence of some other factors such as construction techniques, presence of external cracks, compressive strength and permeability of concrete structures have also been studied in past literature.

Considering the inevitable negative impact of carbonation on service life of RC structures, past researchers have developed numerous models to forecast the depth of concrete carbonation, which is the governing measure of carbonation induced service life depletion. In these models it has been revealed that, there exists a proportinal relationship between the depth of carbonation (xₖ) and the square root of time of exposure of the concrete element (tᵣ)(Sagues, et al. 1997). Later on, this relationship was expressed in the form of an equation as,

\[ xₖ = K(tᵣ)^{1/2} \]  

(1)

Where "K" is the coefficient of carbonation. This constant "K", which is a dimensionless coefficient, accounts for all the factors affecting the rate of concrete carbonation. So, it could be expressed as the combination effect of many environmental and physical factors affecting the rate of carbonation.

But in order to proceed with the derived equation, one must initially find out the carbonation coefficient of any concrete structure. So far, this has been mostly achieved through destructive experimental procedures. Moreover, few numerical models have also been developed. But it was evident that, most of them tend to omit certain prominent factors influencing the carbonation process. Besides, it is seen that, use of most of these models in the industry is quite challenging. As a result, service life calculation of RC structures based on carbonation depth has been discouraged. In order to address all these issues, there exists a need to develop a user friendly equation for the carbonation coefficient of RC structures. The main objective of this study is to develop an equation for calculation of the carbonation coefficient of RC structures.

2. METHODOLOGY

Under the present study carbonation of RC cylinders was experimentally studied by changing the influencing parameters. Among the parameters that govern the carbonation of RC structures, exposed RH and concentration of CO₂ gas were considered as the affecting environmental conditions, whereas concrete grade was considered as the material parameter affecting the rate of concrete carbonation. Accelerated Carbonation Test (ACT) was conducted to accelerate the carbonation of RC cylinders. Depth of the carbonation of all cylinders was measured after 75 days and carbonation coefficient was calculated, separately. Finally, an equation was developed to calculate the coefficient of carbonation of RC structures.

2.1. Accelerated Carbonation Test (ACT)

Experimental procedure of this study was focused on studying the variability of coefficient of carbonation (K) along with RH, exposed CO₂ gas concentration and concrete grade. Initiating the procedure, concrete cylinders (d=110 mm, height = 140 mm) were casted out of three concrete grades (C25, C30, C35), such that maintaining the w/c ratio to be 0.5. Eight concrete cylinders were casted from each grade. From these 24 cylinders, 18 were subjected to Accelerated Carbonation Test (ACT) while the remaining six were subjected to natural carbonation.
In this study, a new low cost accelerated testing procedure was developed to accommodate ACT. Apparatus comprised with six carbonation chambers which were mechanically filled with CO₂ gas. The CO₂ concentrations of 6% and 12% were maintained within test chambers, to achieve higher carbonation depths during the limited experimental period (Table 1). Relative humidity within the carbonation chambers were arranged to be maintained at 40%, 75% and 97% such that covering the full RH range (from 0% to 100%). Here the 75% RH range was achieved by adhering to the environmental RH range in the Rathmalana area, where the tests were carried out. Suitable desiccants for regulating RH in remaining RH ranges were identified during the RH tests which were conducted prior to ACT. Under these RH tests, adequacy of saturated salt solutions (Adequacy of MgCl₂ and K₂SO₄) and silica gel as desiccants was tested.

<table>
<thead>
<tr>
<th>Chamber number</th>
<th>Chamber conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>6% CO₂, 40% RH</td>
</tr>
<tr>
<td>2</td>
<td>6% CO₂, 75% RH</td>
</tr>
<tr>
<td>3</td>
<td>6% CO₂, 97% RH</td>
</tr>
<tr>
<td>4</td>
<td>12% CO₂, 40% RH</td>
</tr>
<tr>
<td>5</td>
<td>12% CO₂, 75% RH</td>
</tr>
<tr>
<td>6</td>
<td>12% CO₂, 97% RH</td>
</tr>
</tbody>
</table>

After 28 days of curing, concrete cylinders were oven dried for 24 hours, to remove any extra moisture from the specimens. Following this, in order to achieve unidirectional carbonation, specimens were covered by an adhesive tape, such that exposing only one surface to the atmosphere. Each chamber was then loaded with three concrete cylinders (one cylinder from each grade), such that leaving the remaining six cylinders to be naturally carbonated. Desiccants were placed within chambers, where necessary. Dehumidification of chambers was achieved with the use of silica gel (mass of 78 grams). Saturated K₂SO₄ solutions (made by dissolving 120 grams of K₂SO₄ in 200 milliliters of distilled water) were used in humidifying chambers. Hygrometers were placed inside each chamber with the motive of monitoring relative humidity and temperature values within chambers. Each chamber was then sealed and was subjected to a leakage test, assuring quality maintenance of the testing procedure. Chambers were then filled with predesigned CO₂ gas concentrations. In this procedure CO₂ gas was introduced to the chambers from the bottom inlet, whereas natural air was taken out through the top outlet (Error! Reference source not found.). This was designed in such a way, since carbon dioxide gas possess a high density than the natural air mixture.

![Figure 1 A Carbonation Chamber](image)

ACT was carried out for a period of 75 days. During this time period temperature and relative humidity values within the chambers were monitored twice a week by using the hygrometer readings. Even though the chambers with silica gel were expected to maintain a relative humidity value of about 40%, it was only observed for the first week of the experiment. During the rest of experimental procedure, those chambers were having an average relative humidity value of 92%. Apart from this unexpected behaviour of chambers having silica gel, relative humidity values inside all other chambers were of as expected.

At the end of 75 days, carbonation depth measurements of concrete specimens were recorded by using a 1% phenolphthalein solution. Initially the depth measurements were obtained by dipping the concrete specimens in the phenolphthalein solution. Later due to some practical errors, depth measurements were obtained by applying phenolphthalein solution on split halves of the specimen (Figure 1). Three
depth measurements were taken from each slice. Finally average value was taken as the representative carbonation depth measurement for each specimen. These depth measurements were then used in calculating K values (Eq. (1) was used in this calculation).

![Figure 1 Carbonation Depth Measurement](image)

**2.2. Derivation and validation of the equation**

Derivation of an equation for coefficient of carbonation (K) was one of the basic objectives of this research. For this purpose initially variation of K along with concrete grade, exposed relative humidity and carbon dioxide concentration was analyzed based on the results of the experimental procedure. With the identification of the linear variation of each factor along with K, principle of multiple linear regression was used in the process of equation derivation (Silva, et al., 2014). Exposed RH, exposed CO₂ gas concentration and concrete grade were considered to be the governing independent variables, whereas K was the dependent variable.

Initiating the derivation data set was tested for outliers. This was done since presence of outliers causes to produce a false inter relationship between the dependent variable and independent variables in a multiple linear regression analysis. Following this the data set was tested for its skewness and kurtosis. Data set was then analyzed to identify the presence of collinearity among variables. Succeeding these tests, data set was analyzed based on the principle of stepwise linear regression, to derive the equation for carbonation coefficient. Finally concluding the research methodology, attempts were made to validate the so derived equation using past research data.

**3. RESULTS AND DISCUSSION**

**3.1. Results of carbonation test**

Results of the carbonation test could be summarized under three sections based on the variability of K along with the three independent variables (i.e. relative humidity, concentration of CO₂ and concrete grade).

**3.1.1. Effect of RH**

Relative humidity (RH) of air plays a major role in controlling concrete pore water volume. Basically, concrete pore water is the medium through which atmospheric carbon dioxide gas reacts with carbonatable compounds in concrete. This addresses the necessity of pore water for the process of concrete carbonation. Although, presence of pore water is a must for concrete carbonation process, when excess amount of pore water is present, it blocks the concrete pore spaces retarding the carbon dioxide diffusion. Moreover, when the pore water percentage is low, carbonation process is hindered due to the lack of aqueous medium for carbonation reactions (Sagues, et al., 1997; Bertolini, 2008; Khunthongkeaw, et al., 2006; Roy, et al., 1999). In this study, although the full RH range was expected to be studied, experimental restrictions limited the studied RH range to the upper range (50% to 100%). In the studied RH range, both carbonation depths and K were following a negative linear relationship with the increase of RH.
3.1.2. Effect of exposed CO$_2$ concentration

Exposed CO$_2$ gas concentration is the most prominent environmental factor affecting the rate of concrete carbonation. Generally, increase of atmospheric carbon dioxide concentration accelerates the carbonation process (Khunthongkeaw, et al., 2006; Papadakis, et al., 1989). Similar variation was observed with the test results of the present study.

![Figure 2 Variation of Carbonation Depth Along with Concrete Grade Under Varying RH Values](image1)

![Figure 3 Variation of Carbonation Depth Along with CO$_2$ Gas Concentration for C25 Concrete Specimens Exposed 75% RH](image2)

Linear relationship of carbonation depth and K along with CO$_2$ gas concentration was revealed with the test results of both ACT and from controlled specimens exposed to natural environmental conditions (Figure 3). When the exposed CO$_2$ gas concentration of specimens subjected to ACT was multiplied by two, it nearly caused to double the resulting carbonation depth (e.g., carbonation depth value of grade 25 specimens at 6% CO$_2$ gas concentration is 9.7 mm while the carbonation depth at 12% CO$_2$ gas concentration is 18.4 mm, which is almost twice the depth value obtained for 6% concentration). But, similar behaviour was not experienced between specimens subjected to CO$_2$ gas concentration of 0.04% (under natural environmental condition) and specimens subjected to CO$_2$ gas concentration of
6% or 12% (under accelerated conditions). This was basically due to the alteration between accelerated and natural carbonation phenomena.

CO₂ gas concentration in accelerated carbonation chambers are clearly higher than the same in natural conditions. As a result, number of interactions between CO₂ gas molecules and concrete surfaces are higher inside accelerated carbonation chambers. This causes for higher CO₂ gas diffusion rates in the accelerated testing process, rather than expected diffusion rates in the natural condition. As a result, time taken for penetration of carbonation front in to the concrete specimen is comparatively low, in the specimens subjected to ACT, where high CO₂ concentrations were present.

3.1.3. Effect of concrete grade

Under the experimental study it was identified that both K and the carbonation depth, follows a negative linear relationship with the concrete grade of the structure (Figure 4, similar variation was observed in specimens exposed to 12% CO₂ concentration). Increase of cement content along with increase of concrete grade, can be considered to as the main cause for this variation. As the concrete grade increases, carbonatable constituents (basically calcium hydroxide and calcium silicate hydrate) in the concrete mixture increases. As a result of that, high amount of CO₂ gas is required to carbonate these high concrete grades.

High degree of porosity observed in low concrete grades too affect in this cause. Basically, permeability of a concrete specimens is correlated with porosity of the structure. Therefore, lower concrete grades are comprised of high permeability, which allows for high rate of CO₂ gas diffusion into the concrete structure. Along with these high diffusion rates, low grades of concrete tend to carbonate more easily.

![Figure 4 Variation of K Along with Concrete Grade and RH for Concrete Specimens Exposed to 6% and 12% CO₂ Concentrations](image)

3.2. Derivation and validation of the equation

3.2.1. Derivation

Since, the experimental data set had no missing values, initially outliers within the data set were identified. Following this skewness and kurtosis analysis was performed. There it was identified that, both skewness and kurtosis values for the dataset remained in the expected range. Collinearity test, which succeeded above two tests, revealed that the collinearity among independent variables (i.e. concrete grade, exposed RH and exposed CO₂ gas concentration) of the analysis is insignificant. Following this, the data set was analysed under the multiple linear regression principle, to derive an equation for "K". A stepwise regression approach was followed here.

Table 2 shows how the three independent variables were ranked by their relevance during the equation derivation. Under the use of this method, it was seen how addition of each independent variable caused to increase the variation of the dependent variable.
Table 2 Summary of the model

<table>
<thead>
<tr>
<th>Model</th>
<th>R</th>
<th>R Square</th>
<th>Adjusted R Square</th>
<th>Std. Error of the Estimate</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.733a</td>
<td>0.537</td>
<td>0.504</td>
<td>0.21857</td>
</tr>
<tr>
<td>2</td>
<td>0.864b</td>
<td>0.747</td>
<td>0.708</td>
<td>0.16762</td>
</tr>
<tr>
<td>3</td>
<td>0.924c</td>
<td>0.853</td>
<td>0.817</td>
<td>0.13278</td>
</tr>
</tbody>
</table>

a. Predictors: (Constant), RH (Relative humidity)
b. Predictors: (Constant), RH (Relative humidity), G (Concrete grade)
c. Predictors: (Constant), RH (Relative humidity), G (Concrete grade), C (CO₂ concentration)
d. Dependent Variable: K (Coefficient of concrete carbonation)

Model summary shows that, 85.3% of the variation of the dependent variable (K) is explained by the variation of the three independent variables, while the remaining 14.7% is explained by the other independent variables which were not studied under this study.

Analysis of variance of the global model (Table 3) justified the strong relationship between dependent variable and independent variables with the aid of F value and significance value of the model. F value being greater than the critical value (which is 3.49) confirmed the involvement of each independent variable in predicting variations of the dependent variable (K). Furthermore, the significance value of the model (sig.) being less than 0.05 (allowed p-value) explained the fact that there exists a strong relationship between independent variables and the dependent variable.

Table 3 ANOVA table of the model

<table>
<thead>
<tr>
<th>Model</th>
<th>Sum of Squares</th>
<th>Df</th>
<th>Mean Square</th>
<th>F</th>
<th>Sig.</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>Regression</td>
<td>1.232</td>
<td>3</td>
<td>0.411</td>
<td>23.288</td>
</tr>
<tr>
<td></td>
<td>Residual</td>
<td>0.212</td>
<td>12</td>
<td>0.018</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Total</td>
<td>1.443</td>
<td>15</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Finally, based on the regression analysis, an equation for carbonation coefficient variations in the relative humidity range of 50% to 100% was derived as follows.

\[ K = 0.034C - 0.028RH - 0.037G +3.872 \]  

(2)

Where K is the coefficient of concrete carbonation (mm/days²), C is the CO₂ concentration (as a percentage), RH is the Relative humidity (as a percentage) and G is the concrete grade of the structure/element.

3.2.2. Validation

Main limitation of the validation process was unavailability of past research data. Even though, several researches have been conducted in the field of concrete carbonation, very few researchers had considered about identical controlled experimental conditions. Therefore, use of such data in validating the equation derived under the present study was contradictory.

In the validation process it was identified that, the carbonation depths generated through the equation were slightly higher than the actual experimental carbonation depths obtained in past studies. Basically, the main reason for this deviation could be the change of experimental conditions maintained in this study, with the experimental conditions maintained in previous researches (relative humidity changes, pressure changes, temperature changes etc.). Similar results were observed, in the attempt of
calculating carbonation coefficients for specimens kept under natural environmental conditions in both past researches and the current study. This could be identified as an outcome of the changes between natural and accelerated carbonation processes.

4. CONCLUSIONS

Under this analysis exposed relative humidity (RH), exposed CO\textsubscript{2} concentration and concrete grade were identified as the governing factors of the rate of concrete carbonation Accelerated Carbonation Test (ACT) was conducted to identify the behaviour of each factor along with the carbonation process. As per the test results of the ACT it was identified that both K and carbonation depth shows a negative linear variation with increasing RH values (in the studied range from 50% to 100%) and increasing concrete grades, while showing a positive linear variation with increasing CO\textsubscript{2} gas concentrations. Then, considering the so identified linear relationships, an equation for the coefficient of carbonation was developed using the principle of multiple linear regression. Under this regression analysis it was identified that, 85.3% of the variation of carbonation coefficient (K) of a concrete structure is been explained by exposed RH, exposed CO\textsubscript{2} concentration and concrete grade used in the construction process. This simply depicts that the derived equation is statistically significant enough to predict the carbonation coefficients of both existing and proposed concrete structures.

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6. REFERENCES


