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Time-dependence of chloride diffusion for concrete containing metakaolin

Hossam S. Al-alaily, Assem A.A. Hassan*

Faculty of Engineering and Applied Science, Memorial University of Newfoundland, St. John's, Newfoundland and Labrador, Canada A1B 3X5

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ABSTRACT

Chloride diffusion coefficient depends on many variables including concrete quality, environmental conditions, and time. In this investigation, the concrete quality and time were studied while maintaining the environmental conditions constant. Fifty-three concrete mixtures were tested based on a refined statistical analysis. Enhanced response surface method (RSM) was used to present the most significant factors affecting the chloride diffusion at different ages. The tested mixtures contained various water-to-binder (W/B) (ratios 0.3–0.5), metakaolin (MK) replacement (0–25%), and total binder content (350–600 kg/m^3). Bulk diffusion test was adopted for two years to determine the time-dependent coefficient m of chloride diffusion for all mixtures based on the error function solution to Fick's law. This coefficient was calculated based on two different bulk diffusion test methods: total and average methods. Design charts were developed to facilitate the optimization of mixture proportions for designers/engineers. The investigation also included some experimental relationships between the rapid chloride permeability test (RCPT), chloride diffusion coefficient, and compressive strength results. The results showed that the values of the chloride diffusion indicated a general reduction from 28 days to 760 days of testing. As the percentage of MK or binder content increased or as the W/B ratio decrease, the chloride diffusion reduction coefficients, m_{total} and m_{avr}, were found to increase. Based on the analysis of variance (ANOVA) from the statistical model, MK was found to be the most significant factor affecting the chloride diffusion at late ages (360 and 720 days), while the W/B ratio was the most significant factor affecting early ages of chloride diffusion (28 and 90 days). The developed models and design charts in this paper can be of special interest to designers/engineers by aiding prediction of service life of concrete containing MK.

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1. Introduction

Enhancing protection and prediction of service life for reinforcing concrete structures strongly impacts economic and ecological sustainability. Rebar corrosion is one of the main problems affecting the durability of reinforced concrete structures. Service life time of reinforced concrete structures can be drastically reduced if the chloride ions near the rebar surface exceed the chloride threshold level in a short time [1]. Concrete cover is considered the first barrier for chloride ion penetration to reach the rebar surface. Chloride ions diffuse slowly through deeper cover thickness, thus improving the structure's service life. Once the chloride threshold is exceeded at the rebar surface, corrosion may start. The concrete cover protecting the rebars can be cracked due to the expansive nature of rust products. The cracking of concrete covers significantly increases the ingress of chlorides and other deleterious substances, causing strength reduction of the structural element and acceleration of the corrosion process [2,3]. Predicting the time of corrosion initiation is determined by the rate of chloride ingress (diffusion coefficient), environmental loading, and chloride threshold level [4–6]. The rate of chloride diffusion is affected by concrete quality due to the size and volume of pore structure, and by time due to the blockage and reduction of the capillary pore structure from the hydration process. Concrete is not a static material, as the concrete continues to hydrate with time, the volume of capillary pores decreases, which reduces the diffusion rate with time. The concrete quality depends mainly on binder content, water-to-binder (W/B) ratio, and any supplementary cementing material (SCM), where they are the primary controlling factors for concrete quality. On the other hand, aggregates in concrete generally have negligible permeability and consequently have an insignificant effect on the transport mechanism of chloride through concrete.

Metakaolin (MK) as a SCM in concrete is receiving a lot of attention recently. The use of MK as a SCM can considerably improve

* Corresponding author.
E-mail address: ahassan@mun.ca (A.A.A. Hassan).

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the performance of concrete structures by improving their durability properties [7–10]. Utilizing MK also improves mechanical properties such as compressive strength, flexural strength, and modulus of elasticity of concrete compared to concrete without any SCM [10–12]. The incorporation of MK in concrete mixtures not only enhances the durability properties and the mechanical properties of concrete, it also significantly reduces CO_2 emissions as it requires lower manufacturing temperatures compared to cement [13].

Diffusion is the prime chloride ingress mechanism, where chloride ions are transported from high concentration to lower concentration in a semi-finite medium (concrete matrix). The prediction and optimization of chloride ingress in reinforced concrete structures has become a crucial part in service life prediction and maintenance of new and existing structures [14–16]. Diffusion of chloride is modeled by using the error function solution to Fick's second law of diffusion (Eq. (1)) [17], which has been studied by numerous researchers [16,18–20].

$$\boldsymbol{C}(\boldsymbol{x}, \boldsymbol{t}) = \boldsymbol{C}_s\left(1 - \boldsymbol{erf}\frac{\boldsymbol{x}}{2\sqrt{\boldsymbol{Dt}}}\right) \tag{1}$$

where $C(x,t)=$ chloride concentration measured at depth x and exposure time t (mass %); $C_s=$ surface chloride concentration, (mass %); $x=$ depth below the exposed surface (to the middle of a layer) (m); $D_a=$ chloride diffusion coefficient (m^2/s); and $t=$ the exposure time (s).

The main weakness of Eq. (1) is the assumption of constant diffusion over time. As a consequence, a single value of the chloride diffusion coefficient can never be obtained from the measured concentration profiles, particularly if the measurements are performed over a long time period. This is because concrete is a hydraulic material that will continually hydrate if there is moisture available. The hydration process will cause refining of the pore structure and subsequently will lower the chloride diffusion coefficient with time [21,22].

Statistical design technique is an efficient tool for optimizing concrete mixtures. It provides statistical models, which help researchers understand the interactions between parameters that have been modeled and optimized [23]. Response surface method (RSM) design techniques are applied in numerous civil engineering fields [24–27]. The RSM is a compilation of mathematical and statistical techniques used for developing, refining, and optimizing processes, and it can be used to evaluate the relative significance of numerous factors even in complex interactions. RSM relies on ANOVA, a method that selects a few points out of the full factorial set that can represent efficient information about the response space.

Limited research has been undertaken that considers statistical analysis in developing models to predict and optimize the chloride diffusion and time dependent coefficient of concrete containing MK. In addition, most of the available studies relate only one factor in the prediction equation based on either the water-to-cement ratio (W/C ratio) or the SCM used. Moreover, most of the available studies utilized a traditional statistical analysis with minimum member points representing the response space. The main objectives of this investigation were divided into two stages: the first stage focused on developing enhanced prediction models for the chloride diffusion coefficient for 53 concrete mixtures at different ages (28, 90, 180, 360, and 760 days). The models incorporated the effects of the three experimental variables: total binder content, W/B ratio, and percentage of MK. The second stage of this investigation focused on predicting the time reduction coefficient (m) for chloride diffusion based on the three experimental variables. RSM was used in this investigation to present the most significant factors affecting chloride diffusion and chloride reduction coefficients in the 53 mixtures, and to develop prediction models for both stages. More points were used in the RSM to enhance the statistical analysis and to develop more precise models for predicting chloride diffusion in concrete containing MK. Design charts were also developed to aid designers/engineers to optimize mixture proportions. In addition, empirical relationship models were established based on the experimental results of chloride diffusion coefficient, chloride permeability test (RCPT), and compressive strength. Finally, a verification mixture was tested under compression strength, RCPT, and chloride diffusion to assess the performance of the prediction models.

2. Research significance

Although there are some available research papers studying chloride diffusion in concrete containing MK, there is limited research that covers the time-dependence effect of chloride diffusion in MK mixtures. The literature also lacks research on the combined effect of the total binder content, W/B ratio, and percentage of MK on the chloride diffusion at different ages; as well as modeling chloride diffusion using statistical analysis in MK mixtures. In this paper an enhanced statistical analysis with more points on the response surface is used in order to develop more precise models, which combine all factors in one equation. The models presented in this paper were developed with the aid of RSM based on an extensive experimental investigation designed to predict the chloride diffusion at different ages and chloride diffusion reduction coefficient in concrete mixtures containing MK. The developed models account for the effects of total binder content, W/B ratio, and percentage of MK. Moreover, unique empirical relationship models based on the results of chloride diffusion coefficient, RCPT, and compressive strength were also developed in this paper. The authors believe that the design charts developed in this investigation will help designers/engineers optimize their concrete mixtures and also strongly contribute to enhancing service life prediction for concrete containing MK.

3. Development of response surface method

In this study, three factors were considered in the development of the enhanced RSM: total binder content, W/B ratio, and the percentage of MK. The derived statistical models are valid for concrete mixtures made with total binder content ranging from 350 kg/m^3 (-1) to 600 kg/m^3 ($+1$), W/B ratios from 0.3 (-1) to 0.5 ($+1$), and percentage of MK from 0% (-1) to 25% ($+1$). The selected range for each of these variables was chosen based on literature and practical considerations. The developed RSM in this investigation considered extra points on the block surface than the common number of points used in standard RSM. In this study, a quadratic model was selected with 8 vertices points, 12 centers of edges, 6 constrain plane centroids, 8 axial check points, 18 interior points, and 1 overall centroid. These points give a total of 53 points to develop the optimization models compared to 15 points used in the standard RSM (see Fig. 1).

The coded values are expressed according to Eq. (2):

$$\boldsymbol{Coded\ \ Value} = \frac{\textbf{absolute value} - \textbf{central value}}{0.5 \times \textbf{range between maximum and minimum values}} \tag{2}$$

The significance of the variables and their connections were determined after compiling the ANOVA, which tests the probability values (Probability $> F$) and indicates that the factor is significant if its probability value is less than 0.05. Also, the factor

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