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Implications of coupled degradation mechanisms of cement based materials exposed to cold climates

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ABSTRACT

Designing durable concrete structures is becoming increasingly important with emphasis being placed on understanding implications on coupled degradation mechanism in order to more accurately estimate service life. This study focuses on the coupled effect of low temperatures on chloride binding capacity, ion-binder interactions and the effect on service life estimates using Life 365 software. In addition to a control (100% general use (GU) cement) paste, three supplementary cementitious materials (slag (GGBFS), metakaolin (MK), and silica fume (SF)), two water to binder ratios (0.3 and 0.5), and several isothermal and thermal cycling curing regimes ranging from 23 °C to –15 °C were tested. Outcomes of this research indicate that: (i) service life estimates vary markedly (as low as 2 years up to 40 years) when coupled effects occur, namely, freeze-thaw temperatures and chloride ingress. (ii) After curing for 56 days and being exposed to chlorides at 23 °C, the order of magnitude of chloride binding is GGBFS (23.78) > GU (21.57) > MK (16.16) > SF (6.14) for 0.5M free chloride concentration but the trend is the same for 1.0 and 2.0 M. The dominance of the GGBFS samples was attributed to the importance of chemical binding related to the aluminate content of the mix components, resulting in the formation of increased amounts of Friedel's salt especially at higher free chloride concentrations of 1.0 M, 2.0 M and 3.0 M. (iii) Thermal cycling over a four month duration revealed that the value of binding capacity changed depending on the exposure temperature. In particular, service life estimates using Life-365 software revealed that chloride binding capacities determined at 23 °C may not be conservative when estimating service life in colder climates. For example, considering concrete containing 40%GGBFS, the ratio of the service life estimate based on 23 °C/0 °C exposure is 0.44.

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1. Introduction

Concrete is the most commonly used building material and its significance to basic infrastructure is immeasurable. Today, many concrete structures are designed with service lives of over 80 years. Concrete infrastructure from bridges to nuclear containment buildings, are undergoing ageing degradation and at the same time, agencies and owners have limited funds for repair and replacement. Designers must evaluate the concrete's resistance to the ingress of moisture and ions in order to control age related degradation. Concrete's ability to resist damage due to outdoor exposure to freeze-thaw cycles, de-icer salts, and sulfates, to name a few, are typically evaluated by accelerated laboratory tests specified in standards such as ASTM, CSA, and RILEM etc. These tests are valuable experiments which can discriminate between concrete mix designs which perform relatively well or relatively poorly. However, key challenges associated with interpretation of the material performance

are: (i) the evaluated durability does not necessarily correlate to the material damage that occurs in the field with age, (ii) the test specifications do not typically account for more than one degradation mechanism acting simultaneously, and (iii) conservative models of field performance of concrete need to account for implications of coupled degradation mechanisms. All structures degrade over time. Processes responsible for such degradation can be physical, chemical or a combination of both. One process of particular importance is related to the corrosion of reinforcing steel as a result of chloride ingress, moisture and oxygen. This can have serious implications on structural integrity and service life of reinforced concrete structures.

Chlorides supplied by the environment in the way of sea water, deicing salts, or chlorides in soil are referred to as external chlorides. Once external chlorides are in contact with concrete they can potentially enter and travel through the pore network as a result of transport mechanisms including sorption, diffusion, and permeation. Transport properties are

Abbreviations: C/S, CaO/SiO₂; C-S-H, calcium silicate hydrate; GGBFS, ground granulated blast furnace slag; GU, general use cement; MK, metakaolin; SCM, supplementary cementing material; SF, silica fume; w/b, water to binder ratio.

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influenced by chloride binding which involves chlorides being physically or chemically bound by the cementitious matrix which affects the rate of chloride ingress by way of delaying the onset of corrosion. Although the topic has been studied, in the literature, there is little consensus on the effect of temperatures variations on the chloride binding capacity. Researchers including Zibara [32], Larsen [12], and Hussain and Rasheeduzzafar [8] showed a decrease in chloride binding with an increase in temperature. Hussain & Rasheeduzzafar claimed that the decrease in chloride binding was due to the decomposition of Friedel's salt at higher temperatures, which in their case was 70 °C. Other researchers however, claimed that temperature only has an effect on the diffusion rate, with no effect on the amount of chloride being bound [18]. Arya et al. [3] determined that there was increased binding when the temperature was raised from 20 °C to 38 °C. Although the temperature range is relatively small, the increase in binding can be attributed to a faster rate of reaction at the increased temperatures [3]. Zibara [32] conditioned samples at 7 °C, 23 °C and 38 °C and showed a decrease in chloride binding with an increase in temperature. This trend was observed up to a free chloride concentration of 0.75 M after which the trend reversed, where the samples conditioned at higher temperatures exhibited greater binding. However, it was noted that the results at 38 °C were inconclusive [32]. Few studies have measured chloride binding isotherms at cold temperatures, particularly below 0 °C. Part of the complexity is because the physical microstructure as well as the chloride concentration plays an important role in determining the freezing point depression which is directly linked to the degradation mechanism [23]. For example, at a 0.1 M (0.58% NaCl) concentration, the freezing point is depressed by less than 1 °C. However, at relatively higher NaCl concentration such as 3.0 M (17% NaCl), the freezing point is depressed by approximately 12 °C.

This paper is focused on current research related to coupled degradation mechanisms which are of relevance and on the forefront of aging concrete infrastructure in Canada as well as cold climates worldwide. Service life of reinforced concrete is typically related to the diffusion of chloride ions into concrete, however, in reality, the diffusivity need reflect transient bulk and surface properties of concrete subjected to freezing and thawing, and salt scaling. The key objectives of this study are:

- i) To investigate the effect of temperatures between –15 °C and 23 °C on chloride binding of cement paste containing general use cement (GU), along with various types and amounts of supplementary cementing materials (SCMs). In this study, ground granulated blast furnace slag (GGBFS), metakaolin (MK), and silica fume (SF) are used.
- ii) Investigate the effect of temperature cycling on chloride binding, ion-binder interactions, chloride binding capacity on service life estimation.

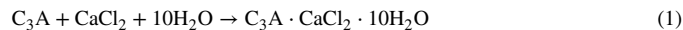
The significance of this research is to contribute to the understanding the coupled effects of chloride and temperature exposure on the chloride binding mechanism in particular related to reinforced concrete structures in geographic regions that experience winter conditions. The study of chloride-binder interactions at temperatures below 23 °C is not well reported in published literature but is critical in order to yield more accurate service life estimates of reinforced concrete structures exposed to chloride in the form of deicing salts. Furthermore, chloride binding capacity more representative of actual exposure conditions is critical to improve service life estimates of concrete structures, allowing the effective allocation of resources for repair or replacement activities.

2. Background

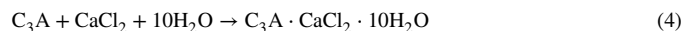
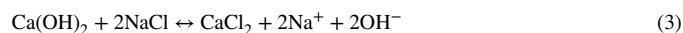
2.1. Chloride binding

Chloride binding processes may be classified as either chemical binding or physical binding. The formation of calcium chloroaluminate, better known as Friedel's salt, is a commonly recognized chemical bind-

ing mechanism where chlorides react with the tricalcium aluminate (C₃A) content of the cement. This reaction is shown in Eq. (1) in which the bound chloride is calcium chloride [25]. The ferrite phase, in the form C₄AF forms a binding product known as calcium chloroferrite [32] shown in Eq. (2) with the aluminate phase formed (C₃AH₆) being able to take part in binding reactions [11]. These two phases are thought to be the most active in chemical binding with the majority of binding being dependent on the C₃A content [30].



Chemical binding involves the creation of either calcium chloroaluminate hydrate C₃A·CaCl₂·10H₂O, better known as Friedel's salt, calcium chloroferrite, C₃F·CaCl₂·10H₂O, which is a variation of Friedel's salt that contains iron instead of alumina, and Kuzel's salt, which forms when ettringite, 6CaO·Al₂O₃·3SO₃·32H₂O, or monosulphate, 4CaO·Al₂O₃·SO₃·12H₂O, react in the presence of chlorides [9,30]. The formation of one of more of these products can be attributed to the reaction of the chlorides with the different phases present in concrete with Kuzel's salt being transformed into Friedel's salt at high concentrations of chlorides [30]. Highlighted in Eqs. (3) and (4) are the mechanisms involved proposed by Suryavanshi et al. [25] and Zibara [32] related to the formation of Friedel's salt and calcium chloroferrite. Based on their findings, the formation of Friedel's salt is dependent on the amounts of C₃A and C₄AF present in the cement.



Another theory was proposed where the increase in OH⁻ ions observed was attributed to the replacement of OH⁻ ions in monosulphate (4CaO·Al₂O₃·SO₃·12H₂O) hydrates by Cl⁻ ions. Eq. (5) highlights the ion-exchange reaction taking place between the OH⁻ ions and Cl⁻ ions in the interlayers of the AFm hydrates as was described by Suryavanshi et al., [25].



Physical binding consists of the adsorption of Cl⁻ ions directly to the calcium silicate hydrate (C-S-H) sheets through Van der Waals forces, with other studies investigating such theories as the electrical double layer theory where the adsorption of chloride ions is necessary to achieve electro-neutrality. Ramachandran [24] determined that the chloride was either present on hydrated calcium silicates in a chemisorbed layer, within the C-S-H interlayer spaces, or intimately bound within the C-S-H lattice. These results were attributed to the C-S-H sheets having a positive charge which would encourage adsorption of the Cl⁻ ions. Supporting this view was another study by Tang and Nilsson [26] where it was found that the bound chloride content was closely related to the C-S-H gel content regardless of the water to binder (w/b) ratio. As a result of these theories, the capacity of the C-S-H layers in adsorbing chlorides is dependent on the surface area of the sheets themselves [32].

2.2. Supplementary cementing materials

Advantages associated with the use of Supplementary Cementitious Materials (SCMs) in concrete which range from economic benefits, environmental benefits as well as the potential for improved durability resistance. SCMs are used as cement replacement materials and are in some cases industrial by-products. As a result, their incorporation into concrete is economically and environmentally advantageous [10]. Slag is a by-product of the iron industry, which after being water cooled and crushed, forms a material commonly referred to as GGBFS. SF is the by-product of the silicon metal industry formed when high-purity quartz is reacted with coal in an electric arc furnace. The SF formed in this process is comprised of 94% - 98% silicon dioxide, SiO₂. MK is a different

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