done to determine the mechanisms of protection of steel in the concrete environment.

It is known that the film formed at the metal surface is relatively unstable, and the passivity provided is temporary. For steel embedded in concrete, the two known means of depassivation are:

- a reduction in concrete alkalinity and
- attack by aggressive ions.

It is reported that when the pH of the surrounding environment is higher than 11.5, the passive film will remain intact and will provide protection to the steel against corrosion [25]. Once the pH falls below this level, the film becomes unstable and the protection of the steel will be lost. Carbonation of concrete is the most common mechanism for the loss of alkalinity in concrete. In addition, aggressive ions, such as chloride, can dissolve or penetrate the passive film at any pH. Most often, this occurs when chlorides diffuse through the concrete and attack the passive barrier. The two most common means of depassivation, carbonation and chloride ion attack, are presented in the following sections.

EFFECT OF CARBONATION

Decaying organic matter often imparts significant concentrations of dissolved carbon dioxide (CO_2) into mineral, groundwater, and seawater [25]. The solution of this gas in water, carbonic acid (H_2CO_3), reacts with the cement paste — a process termed "carbonation". Carbonation of calcium hydroxide ($Ca(OH)_2$) present in the hydrated cement paste is represented by the reactions below:

$$Ca(OH)_2 + H_2CO_3 \rightarrow CaCO_3 + 2H_2O \quad (1)$$

$$CaCO_3 + CO_2 + H_2O \leftrightarrow Ca(HCO_3)_2 \quad (2)$$

From these equations, it can been seen that calcium hydroxide in the hydrated cement paste is consumed through reaction with carbon dioxide or carbonic acid. This results in a gradual loss of buffering capacity which is essential for maintaining alkalinity in concrete. As stated previously, the passive film becomes unstable and no longer provides protection to iron at a pH below 11.5 which implies that carbonation can result in depassivation of reinforcing steel.

The kinetics of the carbonation reaction in concrete have been widely studied, and a parabolic model is generally accepted to describe the depth of carbonation expected over a period of time. According to this model, the depth of carbonation can be predicted as follows:

$$x = k\, t^{1/2} \quad (3)$$

In this equation, x is the thickness of the carbonation layer, t is exposure time, and k is a constant dependent on the permeability of the concrete cover. In good quality concrete, the rate of carbonation is typically on the order of 1mm/year, indicating that carbonation should not present a problem during the design service life of a reinforced concrete structure which has 50mm or more concrete cover over the reinforcement [14].

Carbonation and the resulting loss in alkalinity typically begins at the concrete surface and progresses in the form of a "carbonation front" toward the center of the concrete section. Whereas the carbonated concrete will have pH of less than 8, the uncarbonated concrete maintains its high alkalinity. Thus, the uncarbonated concrete continues to provide a protective cover for the embedded steel. In permeable concrete, concrete with low cover depth, or in concrete containing microcracks connected to surface cracks, the carbonation front may reach the reinforcing steel and thus leads to the onset of corrosion [26]. Therefore, adequate cover depth and a concrete mixture with low permeability and low potential for microcracking is crucial for the protection of embedded steel from corrosion.

Typically, corrosion induced by carbonation is a uniform attack. That is, the presence of very localized corrosion – or pitting – is notably absent. It is important to remember that depassivation of the steel by carbonation is not sufficient to cause corrosion. Water and oxygen must also be present at the cathode site. Because concrete structures are often subjected to cycles of drying and rewetting, an interesting interaction of carbonation and corrosion is found. Venuat [27] has shown that carbon dioxide diffusion is most rapid in concrete at intermediate humidity contents (50-80% relative humidity). When the concrete is completely dry, the carbonation reaction is very slow, and when the concrete is completely saturated, the diffusion of carbon dioxide is very slow. When the concrete is semi-dry, carbonation occurs. Upon wetting, the cathodic reaction of the corrosion process is enhanced. Thus, alternating cycles of drying and wetting provide the most aggressive environment for the carbonation-induced corrosion [16].

EFFECT OF CHLORIDE IONS

As compared to corrosion resulting from concrete carbonation, corrosion of reinforcing steel caused by chloride ion ingress is much more prevalent. Chloride ions can be introduced into the concrete during its manufacture. This can occur when seawater or water with a high chloride concentration is used as mixing water, when chloride-contaminated coarse or fine aggregates are used, or when chloride-containing admixtures such as calcium chloride are used. Chloride ions present in the surrounding environment may penetrate the concrete. Deicing salts, seawater, and chloride-contaminated soils are the primary sources of external chloride. When concrete is dry, chlorides can penetrate several millimeters in a few hours by the capillary draw of salt water into the concrete [16]. When concrete is partly or fully saturated, chloride ions penetrate by diffusion through the pore solution. Typical diffusion rate for a fully saturated cement paste are on the order of 10^{-8} cm^2/s [28].

Different models exist to describe the ingress of fluids and ions, including chlorides, in concrete. For steady-state flow, Darcy's law relates the rate of flow dq/dt to the hydraulic pressure gradient dh/dl by:

$$dq/dt = K\,F\,(dh/dl) \qquad (4)$$

with the permeability coefficient *K* and the cross section exposed to flow *F*: For the diffusion of gases, Fick's laws are typically applied. For the steady state, Fick's first law

$$m = -D\,(\delta c/\delta x) \tag{5}$$

applies, and for the unsteady state, Fick's second law

$$\delta c/\delta t = D\,(\delta^2 c/\delta x^2) \tag{6}$$

applies, where *c* is concentration and *D* is the diffusion coefficient. Recent research by Chatterji [29] has shown that Fick's second law, in particular, is not necessarily applicable to concrete as it assumes that the material is non-ionic and completely saturated. Based upon field experience with concrete structures exposed to chloride environments such as seawater, Sandberg [30] asserted that difficulties in the application of Fick's laws may arise from a pore-blocking effect that results when hydrated cement paste interacts with seawater. Much research has been conducted in recent years to predict the time required for migration of chlorides through concrete. In most cases, some form of Fick's second law has been used to calculate the penetration depth over long periods of time, and, as discussed above, the predicted behavior may be unreliable for use in service life design of reinforced concrete structures.

While it has been generally agreed that chloride ions act as catalysts for the loss of the protection offered by the passive film, the exact mechanisms of this process are not well understood. Two theories appear most often in the literature: the adsorption theory and the oxide film theory (note that the same nomenclature is used to the describe the mechanisms for passivation). According to the adsorption theory, the chloride ions replace oxygen atoms held within the passive film [16]. This causes differences in electrochemical potential across the film, and the film becomes unstable. The high reaction rate of steel and chloride, in areas where chloride ions have replaced oxygen, is thought to explain the occurrence of pitting on the steel surface which is typical of chloride-induced corrosion [23]. According to the oxide film theory, the passive film contains inherent defects and pores. By a selective dissolution of more reactive components of the passive film, chloride ions penetrate the film at these sites more readily than other anions present in the pore solution. The theory suggests that these localized attacks by chloride ions are responsible for pitting corrosion.

Both theories suggest that attack of the passive film by chloride ions is a localized phenomenon. This form of attack causes microgalvanic cells to form on the reinforcing steel as shown in Figure 2. In regions where the depassivation has occurred, iron will be lost by oxidation. The areas that remain protected by the passive film will become cathodic and, thus, sites of oxygen reduction. These processes are described more fully in a subsequent section detailing the chemical reactions involved in the corrosion process.

Because of the damaging effects of chloride in reinforced concrete, much research has been performed to determine a threshold chloride ion content below which corrosion will not occur in concrete. The ions exist in concrete in two forms — bound and free. Only the free chloride ions, those dissolved in the pore fluid, participate in the corrosion process, and as a result, it is the free chloride concentration, not the total chloride content, that is critical when determining threshold levels at the steel concrete interface. It is well-known that the chloride-binding capacity of a concrete depends on the cement composition.

Since chloride ions react with calcium aluminate (C_3A) present in the cement paste to form Friedel's salt ($C_3A \cdot CaCl_2 \cdot 15H_2O$), concrete made from cement with a high C_3A content will have a greater potential for binding chloride ions. Hussain *et al.* [31] demonstrated the beneficial effects of such cement. By raising the C_3A content of cement from 2.43% to 14%, with all other factors remaining constant, the chloride threshold was increased by a factor of 2.85. Kayyali and Haque [32] found that the use of superplasticizing admixtures increased the levels of free chlorides in concrete, but that the addition of fly ash resulted in an increase in chloride binding capacity. Hussain *et al.* reported moderate increases in the chloride threshold value with higher levels of concrete alkalinity. Also, it was found that the presence of sulfates either moderately increased or decreased the threshold value depending on the cement composition.

Hausmann [18] was the first to suggest a relationship between chloride content, concrete alkalinity, and the onset of corrosion. Subsequently, much research has been conducted to establish a Cl^-/OH threshold value for corrosion initiation. As shown in Table 1, a range of values are reported in the literature. For some time, the Cl^-/OH ratio of 0.6 reported by Hausmann was accepted by many, but it should be noted that the study that produced this value was conducted in an alkaline solution meant to simulate concrete pore fluid, not within concrete itself. Recent research suggests that the source of chloride contamination has a significant influence on the threshold values. Diamond [33] reported a threshold of 0.3 when the chloride is introduced into the concrete as an admixture or in the mixing water. Lambert *et al.* [34] reported a much higher threshold ratio, 3.0, when the chloride is introduced from an external source. It is important to recognize that the threshold Cl^-/OH ratio is not easily defined and that no value has been accepted as a unique limit for all concrete mixes. Threshold values are highly dependent on such parameters as the water/cement ratio, cement content, cement surface area, cement composition, ambient temperature, and alkalinity. As a result, a well-defined threshold relationship universally applicable to all concrete mixtures remains elusive. Consequently, service life predictions based on certain assumed threshold values are not reliable.

ROLE OF MINERAL SCALES

While it is generally accepted that the presence of a passive film on the steel surface provides some corrosion protection, some researchers, believe that an additional phenomenon may offer further protection. Many authors consider the pore fluid to be the environment of exposure in concrete. As a result of this assumption, much reinforced concrete corrosion research has been conducted in aqueous solutions that are meant to simulate the concrete pore fluid. Consequently, the corrosion behavior of steel in concrete has been extrapolated from its behavior in aqueous solutions of similar pH. This approach neglects the effect of the interfacial steel-cement paste zone which consists of hydrated cement paste minerals.

Borgard *et al.* [11] postulate that mineral scales, consisting of precipitates of cementitious compounds, form on metal surfaces in concrete and act in conjunction with the metal oxide passive film to protect reinforcing steel. The authors suggest that this phenomenon has been ignored in much of the literature as a consequence of the use of pore solution models, which do not account for the presence of the hydrated cement paste minerals, in corrosion studies. Page [35] has also proposed that passivation may result from a lime-rich layer of hydration products at the interface between steel and concrete.

Borgard *et al.* state that calcium carbonate scales are used for protection of oil well production tubing even in the presence of chloride concentrations much greater than in seawater. They assert that the formation of high calcium scales on steel embedded in concrete is likely since portland cement has a high calcium content. The authors reason that corrosion in concrete may be prevented by a similar mechanism and that corrosion will occur only after the calcium scales have been removed.

It should be recognized that chemical attack on the calcium hydroxide scale by carbon dioxide, chloride, or sulfate ions can result in eventual removal of the scale. However, the process of removal can provide additional protection to the passive film on the steel surface because of the dissociation of the calcium hydroxide by reaction with such species as chloride ions and carbon dioxide. Such reactions produce free hydroxyl ions which act to buffer the alkalinity of the pore solution. If this reaction takes place in the vicinity of the steel, passivity can be maintained by ensuring that the pH remains above 11.5. However, once the calcium hydroxide scale has been depleted, further ingress of chloride ions or carbon dioxide damage the passive film.

CORROSION CHEMISTRY

As stated previously, corrosion is an electrochemical process involving the transfer of ions. In reinforced concrete, electrons are produced at the anodic site by the oxidation of the steel. That is, metal ions from the parent metal go into solution at the anode, and the loss of metal indicates that this is the site of corrosion. The rate of steel corrosion in concrete is dependent on the rate of the anodic reaction:

$$Fe \rightarrow Fe^{++} + 2e^- \tag{7}$$

Electrons released at the anodic site are consumed at the cathodic site. Typically, at the cathode, either oxygen or hydrogen is reduced. The thermodynamic driving force for the reduction of oxygen is much greater than that for hydrogen. As a result, the oxygen reaction is more likely to occur, and the reduction of hydrogen in concrete corrosion is rarely a factor. The cathodic reactions are:

$$2H_2O + O_2 + 4e^- \rightarrow 4OH^- \tag{8}$$

$$2H^+ + 2e^- \rightarrow H_2 \tag{9}$$

Ferrous ions (Fe^{++}) produced in the anodic reaction (eq. 7) combine with the hydroxyl ions produced in the cathodic reaction (eq. 8) to form the corrosion product ferrous hydroxide ($Fe(OH)_2$). The corrosion rate can be increased by the presence of other ions in the concrete. According to Hime and Erlin [36], the reaction of ferrous ions and hydroxyl ions in the presence of chloride ions can also result in the formation of the corrosion product $Fe(OH)_2$ (eqs. 10-12). The reaction represented in equation 16 demonstrates that chloride ions are produced by this process, increasing the corrosion rate of the steel reinforcing.

$$Fe^{++} + 6Cl^- \leftrightarrow FeCl_6^{-4} \qquad (10)$$

$$Fe^{+3} + 6Cl^- \leftrightarrow FeCl_6^{-3} \qquad (11)$$

$$FeCl_6^{-3} + 2OH^- \leftrightarrow Fe(OH)_2 + 6Cl^- \qquad (12)$$

The process of corrosion for steel reinforcement bars embedded in concrete is illustrated in Figure 2. The diagram shows that the electrons are released at the anodic site and travel through the steel to the cathodic site, where they are consumed. The reduction of oxygen at the cathode produces hydroxyl ions (OH^-). The production of hydrogen gas, caused by the reduction of hydrogen, is less likely, but possible. Equations 7 and 8 demonstrate that the corrosion process may be limited by the rate of diffusion of the hydroxyl ions, as well as by the availability of oxygen (O_2), water (H_2O), or other aggressive ions (Cl^- in Fig. 2 and eq. 10-12).

Glasser and Sagoe-Crensil [37] have examined a 27 year old reinforced concrete post to determine the morphology and characteristics of the corrosion product (i.e. rust) present. In the specimen studied, the corrosion product was found to be composed of two layers: an inner, highly crystalline and dense region of Fe_3O_4 and γ-Fe_2O_3 and an outer more porous region of primarily α-FeOOH (goethite). The inner layer was found to have preferential orientation related to that of the substrate metal microstructure that resulted in epitaxial growth of the inner layer corrosion product. Wang and Monteiro [39] confirmed that the corrosion product tends to form a continuous band poorly bound to the reinforcing steel and that this band is composed of multiple layers. It has been suggested that this structure results from variations in oxygen availability and pore solution conductivity across the region of corrosion.

MECHANISMS OF EXPANSION AND CRACKING OF CONCRETE

The solid-state transformation of iron to the ferrous hydroxide corrosion product resulting in a volumetric expansion is generally assumed to be the mechanism by which concrete expands and cracks by the corrosion of embedded steel. However, the corrosion chemistry shows that the corrosion of iron to ferrous hydroxide occurs as a result of a through-solution process. That is, iron present in steel must oxidize to soluble ferrous ions. The ferrous ions combine, in solution, with hydroxyl ions generated at the cathode to produce ferrous hydroxide. Consequently, Mehta [39] and Figg [40] postulate that poorly crystalline or gel-like ferrous hydroxide tends to swell by water adsorption and that the hydraulic pressure generated is responsible for expansion and cracking.

Recently, Wang and Monteiro [38] studied the mechanisms by which corrosion of embedded steel reinforcement undermines the strength of concrete structures. Once the depassivation of the steel occurs and corrosion begins, they found that oxidation products form as a band around the steel. Loss of reinforcing steel by corrosion leads directly to loss in the strength of the reinforced concrete member. While the presence of water is necessary for the corrosion of reinforcing steel, Wang and Monteiro found that cycles of relative wet and dry conditions at the concrete and steel interface lead to weakening of the corroding reinforced concrete member. When wet, adsorbed water will force corrosion product particles further apart, resulting in expansion and cracking of the surrounding concrete. Upon drying, the corrosion product will tend to shrink and crack. In this way, cycles of wetting and drying will cause the steel and concrete to debond at the interface. In addition, Wang and Monteiro found that corrosion products diffuse away from the interface into the microcracks and pores of the surrounding concrete. The researchers reason that 'plugging' of existing voids in the concrete by diffused corrosion product will decrease the probability of crack arrest, effectively embrittling the concrete and weakening the reinforced member. Loss of steel, debonding at the steel/concrete interface, and cracking and embrittlement of surrounding concrete is detrimental to the strength and integrity of reinforced concrete.

Mehta [41] has proposed a holistic model, Figure 3, which illustrates all the *physico-chemical changes occurring in the reinforcing steel and concrete* in response to all environmental effects including weathering and ingress of aggressive chloride ions and carbon dioxide. Most descriptions of the effects of corrosion of steel embedded in concrete center on the volume expansion associated with the corrosion products and the resulting cracking and debonding of the concrete. As discussed previously, corrosion of reinforcing steel damages the surrounding concrete as well as the steel. The mechanisms depicted in Figure 3 are holistic. That is, no part of the system is overlooked. The mechanisms that lead to the corrosion of steel also affect the surrounding concrete, and these relationships are considered in this model.

For instance, since C-S-H is the primary source of strength in the cement paste and since the stability of C-S-H is dependent on the concentration of hydroxyl ions in the pore fluid, a decrease in alkalinity has a detrimental effect on the strength and elastic modulus [41]. Free hydroxyl ions in the pore fluid can combine with other species, such as carbonate, sulfate, and chloride. This type of reaction, which results in a decrease of the pore fluid alkalinity, causes loss of strength and elastic modulus in the concrete. The mechanism of expansion and cracking of concrete shown here takes into consideration the combined effect of weakened microstructure of the cement paste and the development of hydraulic pressure in the pores of the water-saturated system. Thus, Figure 3 demonstrates an integration of all environmental effects on *both* components of the reinforced concrete system.

CONCLUSIONS

Upon critical review of the state-of-the-art understanding of expansion and cracking of reinforced concrete due to corrosion of the embedded steel, several areas of uncertainty or ambiguity emerge. These are identified as:

- Mechanisms of steel passivation in the concrete environment.
- The stoichiometry of the passive film on steel and the mechanisms of its formation.
- The relative significance of the passive film and mineral scales in providing corrosion protection to steel.
- The composition of the mineral scales formed in the vicinity of the steel and the mechanism of corrosion protection.
- Applicability of Fick's second law for the prediction of chloride diffusion in concrete.
- Mechanisms of depassivation of reinforcing steel by chloride ions and carbonation.
- The threshold Cl^-/OH ratio for corrosion to occur.
- Composition, morphology, and mechanisms of expansion of the steel corrosion products.
- Detrimental effects of aggressive species on the material properties of concrete itself.

The study of corrosion is limited to some extent by the inherent difficulties associated with observing the corrosion of steel within concrete. However, some of the gaps in our understanding of the steel corrosion process in concrete seem to stem from the reductionist or fragmentary approach taken by researchers on this subject. Corrosion engineers and material scientists tend to study the corrosion of steel in alkaline aqueous solutions and, as a result, neglect the possible effects of the cement paste microstructure on the corrosion process and changes in the microstructure during corrosion. For instance, the effect of chloride attack and carbonation on the surface chemistry of steel is considered, whereas their detrimental effect on the strength and elastic modulus of the concrete is often completely overlooked in our corrosion damage models. Widespread misapplication of Pourbaix diagrams for prediction of passivity of steel embedded in concrete is an example of the narrow approach applied to the study of corrosion of steel embedded in concrete. Only when researchers take a holistic approach for the study of combined response of both the reinforcing steel and the surrounding concrete to all environmental effects will these ambiguities be resolved. To address the deficiencies and advance the state-of-the-art in concrete science, we obviously need a paradigm shift in our approach to corrosion research.

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