

Modeling of Corrosion; Steel, Concrete and Environment

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Editorial

In this editorial, an overview of modeling of corrosion, previous research, related theories and discussions step by step at first for steel, then for steel reinforced concrete and then for steel reinforced concrete under the effect of environment has been presented. In this editorial, material and environmental modeling has been carried out simultaneously for concrete exposed to a variety of environmental actions with varying concrete compositions and dimensions.

By definition corrosion is the chemical or electrochemical reaction between a material, usually metal, and its environment that produces a deterioration of materials and its properties [1]. Due to the phenomenon of passivity, it is not uncommon for the current to change by six orders of magnitude during a corrosion experiment [2]. An electrochemical reaction under kinetic control obeys the Tafel equation [3]. For the range of corrosion phenomena that can be studied with electrochemistry, the ability to measure very low corrosion rates, and the speed with which these measurements can be conducted, an electrochemical corrosion measurement system has become a standard item in the modern corrosion laboratory [4,5]. For a simple activation controlled metal dissolution (anodic) process with the rate of the reverse metal ion reduction, reaction is considered to be small and the current density is given by Tafel's Law [3]. It is assumed that the surface concentration of the cathodic reactant did not vary with potential. This is approximately true if the cathodic current density is much less than the limiting mass transfer controlled current density [6]. For a cathodic reaction, neglecting the reverse oxidation reaction, the limiting diffusion current density is given by Fick's 1st Law [7] with a surface concentration of zero.

After reviewing principles which govern modeling of metallic corrosion in general, it is valuable to describe about various states of steel corrosion modeling in concrete, related theories and physical phenomenon. In general, good quality concrete of appropriate mix proportion, compacting, and curing provides an excellent protective environment for steel. The physical protection is afforded by the cover concrete acting as a physical barrier to the access of aggressive species. Chemical protection is provided by concrete's high alkalinity solution within the pore structure of cement paste matrix due to the presence of sodium and potassium oxides in the cement, as well as calcium hydroxide produced in the hydration reactions of cement components [8,9].

Although there are many types of corrosion such as uniform, galvanic, crevice, pitting, environmentally induced, hydrogen damage, intergranular, dealloying, erosion corrosion, only uniform and pitting corrosion relating to reinforcing steel corrosion are discussed here. Uniform corrosion occurs when the corrosive environment is the same to all parts of metal surface. This can be considered as depassivation of protective film of steel due to carbonation which penetration of carbonation front is relatively the same. In contrast, pitting corrosion is localized corrosion which passivity protective film is locally broken down by presence of chloride ion or other halogen ions accelerated at high temperature. Although uniform corrosion consumes a larger amount of steel, localized corrosion is more difficult to predict and monitor and causes rapidly failure because of corrosion concentration at certain points [10-16].

The mechanism of corrosion of steel reinforcement in concrete could be resembled to an electro-chemical cell formation. This phenomena has same ground reaction mechanisms and surrounding

environment in principle. Similar to the electrochemical cell, moist concrete acts as an electrolyte, a medium for the movement of charge carriers/ions. Hydroxyl ions move from cathode to anode within the moist concrete and electrons move towards cathode from anode within the steel itself. This causes iron to corrode at the anode site by for formation of various oxides and hydro-oxides. Therefore, corrosion of steel in concrete consists of movement of electrons similar to the movement of electrons in case of electric current flow in the cell. This involves chemical reactions at anode and cathode sites in concrete on the surface of steel bar depending on environment surrounding the steel bar as well as the concrete. Thus the modeling of corrosion in RC structures has to be electrochemical in nature [17-19].

A simple model of electrochemical reactions associated with corrosion deterioration of steel within concrete follows. Oxidization of iron (Fe^{++}) molecules naturally occurs immediately after the bar is manufactured and exposed to the atmosphere, and will continue so long as sufficient oxygen and moisture are available to react with the steel. Upon exposure to the high pH environment of concrete, a passive layer of oxidation product forms on the encapsulated steel surface. This passivation process is actually a form of corrosion. However, in the moist, high pH environment of concrete, the reaction occurs at an ever-decreasing rate. In the absence of aggressive ions, oxidation nearly ceases after a sufficient passive layer has formed. The passive layer normally protects the reinforcement from spontaneous corrosion in a moist, oxygen-rich environment such as concrete. However, chloride ions (Cl^-) that diffuse to the steel surface can disrupt the passive layer and induce corrosion. Generally, metal atoms pass into solution as positively charged ions at the anodic site and liberated electrons flow through the metal to cathodic sites where dissolved oxygen is available to consume them. For example, chloride ions react with iron compounds in the passive layer to create an iron chloride complex ($FeCl_2$), which subsequently reacts with hydroxide (OH^-) from the surrounding concrete to form hydrated iron oxide compounds. This is commonly known as the anodic reaction. Simultaneously, at an alternate location on the steel surface, oxygen (O_2) reacts with water (H_2O) and electrons released by the anodic reaction to form hydroxide. This is referred to as the cathodic reaction. Together, the anodic reaction and the cathodic reaction form a corrosion cell [10-19].

As discussed above, the process of corrosion of steel in concrete is generally considered as being electrochemical in nature, and the basic mechanisms of electrochemical theory of corrosion developed for liquid electrolytes apply for modeling here [20-21]. However, electrochemical corrosion modeling of iron in concrete has a number of characteristic differences that are determined by the peculiarities of electrochemical

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processes in the complex “concrete electrolyte”, the properties of which are affected by the micro structure of the concrete and service exposure conditions. Concrete is a highly heterogeneous and complex structure, which contains a heterogeneous distribution of different types and amounts of solid phases, pores, and micro cracks (Mehta P.K, 1993). In addition, the structure of concrete is also subject to change with time, environmental humidity and temperature. A large amount of water can exist in the hydrated cement paste, depending on the environmental humidity, porosity and pore size distribution of the hydrated cement paste. The presence and transport of the corrosion reactants (water, oxygen and various ions), the corrosion products, and the passage of the ionic current necessary to support corrosion are normally confined to the cement paste phase – cement paste pore solution and the structure and distribution of pores.

The composition of the pore solution is the decisive factor in determining whether embedded steel will be passivated or whether it will actively corrode. Researchers on the influence of a number of factors on the composition of pore solution [21-23] have shown that the presence of sodium and potassium oxides in the cement, as well as calcium hydroxide produced in the hydration reactions of cement components can give the pore solution of ordinary portland cement a pH of about 13 while the pH of blended cement is somewhat lower. The range of high pH values of typical concrete is within the pH domain in which insoluble oxides of iron are thermodynamically stable to maintain a passive film on steel surface. Unfortunately, the pH value of concrete can be reduced by carbonation and by leaching.

Generally the best measure of corrosivity of concrete is its conductivity, usually expressed as the reciprocal, the resistivity. A low concrete resistivity indicates a high corrosive activity. The resistivity of the concrete is mainly determined by the salt content in the pore water, degree of saturation and temperature. The resistivity of the concrete can vary widely for different conditions [24-27]. Gjorv [25] has reported resistivity values of 7,000 and 6,000,000W cm for 100 % and 20 % saturated concrete, respectively, which are possible in natural environments. High water/cement ratio, chloride bearing, saturated concrete provide the lower resistivity, while low water/cement ratio, well cured, dry concrete provides the highest. There is an absence of complete correlation between electrical resistivity and concrete corrosivity.

In this editorial, corrosion modeling of steel in concrete is based on electrochemical processes. The corroding model system consists of an anode in which steel is corroded, a cathode, an electrical conductor, and an electrolyte (concrete pore solution). The potential difference between anode and cathode is the driving electrical force for steel corrosion. The modeling process is divided into primary electrochemical processes and secondary processes. Since the volume of rust products is much higher (about 4 to 6 times) than that of the iron, the formation of rust products will lead to cracking and spalling of the cover concrete when expansive stress exceeds the tensile strength of the concrete, and reduction of steel reinforcing cross section may lead to structure failure.

Reinforcement corrosion in concrete is essentially an electrochemical process, where at the anode iron is oxidized to iron ions that pass into solution, and at the cathode oxygen is reduced to hydroxyl ions. The anode and the cathode form a short circuited corrosion cell formed within the concrete, with the flow of electrons in the steel and of ions in the concrete pore solution [28]. Depending on various factors such as the pH value of the pore solution, the availability of oxygen and moisture, the content of aggressive agents etc., the corrosion products possess complex stoichiometry and may expand by about four to six times the original volume of iron corroded. Two corrosion products

are generally thought to be dominant — ferrous hydroxide $\text{Fe}(\text{OH})_2$ and hydrated red rust $\text{Fe}(\text{OH})_3$ [29-32]. Faraday's law forms the foundation for all existing reinforcement corrosion models that assume a constant rate of iron consumption and rust generation. To understand the mechanical effects of rust expansion, a corrosion-induced rust expansion model is illustrated [33].

Water-cement ratio in the original mix plays a decisive role in determining the pore structure of the hardened cement past. The excess water which cannot be consumed in the hydration process is the direct cause of the high degree of porosity in ordinary cement. This high degree of porosity causes to increase the penetration rate of the aggressive ions inside concrete and subsequently the depassivation. As the water-cement ratio increases oxygen permeability also increases. Also increase in the water-cement ratio increases the carbonation depth. Ohno et al., [34] reported that rust area increases as the water-cement ratio increases. The narrower and more twisted the pores of the hardened cement pastes the more the transport is obstructed and this reduces the corrosion rate. Miyazato [35] reported that in case of Cl^- induced corrosion, the decrease of the water-cement ratio made the macro cell corrosion activated. On the other hand, increasing the water-cement ratio made the microcell corrosion activated. The progress of steel corrosion in concrete is easily influenced by pH in pore solution surrounding steel. Evans [36] reported that the corrosion rate increased as the pH of concrete decreased in the condition of pH lower than 10. Montemor et al., reviewed the steel corrosion monitoring techniques including potential measurements, polarization resistance measurements, electrochemical impedance spectroscopy (EIS), and transient techniques. Half-cell potential measurement is one of the simplest methods to assess state of steel corrosion. ASTM C876 provides standard test method for half-cell potential measurement of uncoated rebar in concrete structure. Guideline for interpreting measured half-cell potential value is provided by ASTM C876. However, there are many factors should be considered before interpreting results such as condition of cover concrete, or type of rebar. Uomoto (2000) showed that measured half-cell potential values at concrete surface could be considered as actual value at steel surface, if cover depth was within 20 mm. Gu and Beaudoin [37] reported that interpretation criteria of ASTM C876 is most applicable for corrosion due to chloride. Stern et al., showed that for a simple corroding system, the polarization curve for a few milli volts around the corrosion potential obeys a quasi-linear relationship. The slope of this relationship is called “Polarization Resistance”. The value of B factor is determined from the particular electrochemical cell and generally ranges from 13 to 52 mV depending on the system. For measuring it, a potentiostat is necessary. Based on the theories of polarization resistance to obtain evidence of corrosion activity, two devices which are currently used for measuring the corrosion rate (corrosion current density) both in laboratory and in the field are K. CC. Lear's 3LP [38] and Gecor device [39].

AC impedance technique is the application to the working electrode maintained at its corrosion potential by potentiostat of a small amplitude (a few milli volts, DE peak to peak) sinusoidal voltage in an extensive frequency range [40-42]. The detail test procedures of preparing, cleaning, and evaluating corrosion test specimens are described in ASTM-G1. The corrosion model in this editorial was firstly adopted from the previous research [43]. In the model a general scheme of micro-cell corrosion is introduced based on thermodynamic electro-chemistry. Tafel Diagram [3] is a useful tool for simulating the corrosion phenomenon. From the electric potential and the formation of passive layers, electric current that involves chemical reaction can be calculated so that the conservation law of electric charge should be satisfied in a local area. It is well documented [44-49] that the presence

of chloride ions in reinforced concrete can cause the steel to corrode if sufficient oxygen and moisture are present to sustain the reaction. The chloride-induced corrosion is the most prevalent and damaging cause of corrosion of steel in concrete. The mechanism of chloride induced corrosion of steel is not yet fully understood. It is generally believed [50-55] that the chloride ions become incorporated in the passive film, replacing some of the oxygen and increasing both its conductivity and its solubility. The threshold concentration of chloride ions to initiate corrosion is controversial, because it is dependent on so many factors including quality of concrete (W/C ratio, mixture proportions, type of cement), relative humidity and temperature of the concrete, the pH of the pore solution and sulfated content [56-59].

The concrete capillary-pore system filled with water and air acts as the corrosive electrolyte. Apart from the pore structure, porosity and pore size distribution of the concrete, water content in the concrete is dependent on the local amount and time of rainfall, and external relative humidity and temperature. Certain water content in concrete is an essential requirement for corrosion to take place. As a rule, the presence of oxygen is also necessary for cathodic reaction process. If a pore volume of the concrete is only partly filled with water, transfer of oxygen will be easy to the steel surface by means of diffusion in the gas phase. Whereas, when a pore volume of the concrete is completely filled with water, then oxygen can reach the steel surface only by diffusion through the pore water, and the diffusivity of oxygen in water is about four powers lower than that in air [60].

Kobayashi [61] has reported that when moisture content of concrete is lowered from 80 % to 40%; the value of oxygen diffusion coefficient becomes approximately 15 times higher. Temperature of concrete can vary widely, depending on the geographic latitude and climatic conditions and also on the time of year and day. The temperature dependence of the kinetics of electrode processes and processes of diffusion that take place in the concrete can be a certain degree expressed by the Arrhenius equation [60]. The process of steel corrosion in concrete, the rate of which is usually determined by the kinetics of diffusion or by the kinetics of electrode process, will generally rise exponentially with a rise in temperature, in accordance with the Arrhenius equation. However, because of the complexity of the corrosion process in the concrete, there can be quite a significant deviation from this dependence [26,62]. For example, if a high temperature is associated with more rapid drying or lower oxygen solubility in the concrete moisture, the temperature dependence of corrosion in concrete will usually be quite different. Clear [59] suggested that the corrosion current density measured at given field temperature can be adjusted to another temperature using some formulation given in his publication. Usually, penetration of a particular substance such as chloride ions into concrete can be in two forms: capillary attraction and ionic diffusion, depending on the degree of saturation in the concrete [63,64].

The prediction of corrosion in RC structures due to environmental actions is complex and needs extensive research in this area. The methodology adopted by Hussain et al., [10-16] is based on a 3D finite element model developed by the research group. It is a computational program for the evaluation of various durability aspects of concrete related to material as well as environmental modeling such as concrete hardening (hydration), microstructure formation and several associated phenomenon, from casting of concrete to a period of several months or years. As such this tool can be utilized to study the effect of ingredient materials, environmental conditions as well as the size and shape of structure on the durability of concrete. The term durability considered here takes into account both the green concrete stage problems as well as matured concrete exposed to environment. This

tool can be used to analytically trace the evolution of microstructure, strength and temperature with time for any arbitrary initial and boundary conditions. Since the main simulation program is based upon finite-element methods, it could be applied to analyze real life concrete structures of any shape, size or configuration. Furthermore, dynamic coupling of several phenomena ensures that the effect of changing environmental conditions is easily integrated into the overall simulation scheme.

Summarizing all above in the model, a general methodology of micro-cell corrosion model is introduced based on thermodynamic electro-chemistry [43,12-16, 18,19]. The corrosion process is assumed to occur uniformly over the surface area of reinforcement in a reference finite element volume. Some issues still need to be taken into account such as formation of pits due to the localized attack of chlorides and corrosion due to the formation of macro-cell. The beauty of this model is that all the requisite parameters such as pH, temperature, partial pressure of oxygen, chloride concentration etc. are all obtained automatically from the connected sub-routines in the model for the computation of corrosion.

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