Review

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Atmospheric corrosion prediction: a review

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Abstract: The atmospheric corrosion of metallic materials causes great economic loss every year worldwide. Thus, it is meaningful to predict the corrosion loss in different field environments. Generally, the corrosion prediction method includes three parts of work: the modelling of the corrosive environment, the calibration of the corrosion effects, and the establishment of the corrosion kinetics. This paper gives an overview of the existing methods as well as promising tools and technologies which can be used in corrosion prediction. The basic corrosion kinetic model is the power function model and it is accurate for short-term corrosion process. As for the long-term corrosion process, the general linear models are more appropriate as they consider the protective effect of the corrosion products. Most corrosion effect models correlate the environmental variables, which are characterized by the annual average value in most cases, with corrosion parameters by linear equations which is known as the dose-response function. Apart from these conventional methods, some mathematical and numerical methods are also appropriate for corrosion prediction. The corrosive environment can be described by statistical distributions, time-varying functions and even geographic information system (GIS), while the corrosion effect can be captured via response surface models and statistical learning methods.

Keywords: atmospheric corrosion; corrosion effect; corrosion kinetics; corrosion prediction; corrosion test; environmental factors.

1 Introduction

Atmospheric corrosion on metallic materials is very common and it causes a big amount of economic loss every year (Roberge et al. 2002). There are many types of atmospheric environments with different temperature, relative humidity, pollutants concentration, and other environmental conditions. When metal materials are exposed in various atmospheric environments, the kinetics law of the corrosion process is also different.

Multiple environmental factors can affect the atmospheric corrosion process with complex mechanisms (Leygraf et al. 2016; Simillion et al. 2014). However, the corrosion behavior and kinetic laws of many metal materials are not very clear and there are no rational models for corrosion prediction. In view of this, it is meaningful to study the influential factors, corrosion effects, and the kinetic laws of the corrosion process and predict corrosion loss in various atmospheric environments.

In the atmospheric environment, the corrosion process is influenced by multiple factors, including temperature, relative humidity, solar radiation, precipitation, wind, pollutants, and so on (Leygraf et al. 2016). These environmental factors are non-constant and change continuously over time. In different regions, the environmental conditions are in great difference. Some factors are dependent on each other such as temperature and relative humidity (Cai et al. 2018a). These environmental factors are also affected by a variety of factors in random behaviors.

The influence of different environmental factors on the atmospheric corrosion process is very complicated. Firstly, the influence of different environmental factors on the corrosion rate of metals is generally nonlinear (Cai et al. 2018a). For example, in previous studies (Shinohara et al. 2005; Wang et al. 2015), the corrosion rate of metals increases exponentially as the relative humidity increases. Secondly, there may be interactive effects between different environmental factors. For example, when different pollutants are present in the atmosphere at the same time, the corrosion behavior is influenced in complex ways depending on multiple factors (Castano et al. 2007).

The long-term atmospheric corrosion behavior develops along with corrosion time and varies under different environmental conditions. The corrosion rate exhibits a

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multi-stage law during the entire exposure period in the atmospheric environment. At the early stage, the corrosion rate is relatively high. As the corrosion products accumulate on the metal surface, the corrosion rate gradually slows down and tends to be constant (Barton 1976; Cui et al. 2015; ISO 2012b; Lv et al. 2015). At the same time, under different environmental conditions, the protective effect of corrosion products and the long-term corrosion kinetic law is quite different (Panchenko et al. 2014).

Therefore, it is of great theoretical and engineering value to build accurate atmospheric environmental models, corrosion effect models, and corrosion kinetic models. It is helpful for the establishment of corrosion prediction method for metal materials in various atmospheric environments. These models are reviewed in this paper as well as the field exposure and laboratory corrosion tests that produce the corrosion data for the calibration of these models.

2 Field exposure and laboratory corrosion tests

2.1 Field exposure corrosion tests

The field exposure corrosion test is the most commonly used method in atmospheric corrosion research because the results can truly reflect the corrosivity of the actual atmospheric environment (Hou and Liang 1999; Wallinder and Leygraf 2001). The American Society for Testing and Materials (ASTM) is dedicated to the study of corrosion in the atmospheric environment (Feliu et al. 1999) and has carried out series of corrosion tests on various materials. For example, in the ISO CORRAG program (Knotkova et al. 2010), 53 test sites in 13 countries around the world are selected and an 8-year field exposure corrosion test is conducted on carbon steel, zinc, copper, and aluminum. Corrosion loss is measured at the 1, 2, 4, and 8 years of the test. The temperature, relative humidity, sulfur dioxide concentration, chloride deposition rate, and other environmental factors are monitored and recorded during the entire exposure period. These data can be used to analyze the corrosion development along with corrosion time under different environmental conditions.

There are some other international corrosion test projects such as the UN/ECE, MICAT, OSD, RPF project which also obtained a large number of corrosion data in the natural atmospheric exposure conditions. The UN/ECE project (Tidblad et al. 1998, 2001) selected 39 test sites in 12 European countries, the United States and Canada. An 8-year field exposure corrosion test is conducted on weathering steel, zinc, aluminum, copper, limestone, sandstone, and other materials. The environmental data of different test sites were recorded, including temperature, relative humidity, sulfur dioxide concentration, ozone concentration, rainfall, hydrogen ion and chloride ion content in the rainwater.

The MICAT project (Morcillo 1995; Morcillo et al. 1998; Pintos et al. 2000; Rosales et al. 1999) aims to construct a map of metallic atmospheric corrosion in the Americas. Therefore 72 test sites in 14 countries in the Ibero-America are selected. Carbon steel, zinc, copper, and aluminum were subjected to a 4-year corrosion test and their corrosion weight loss was measured at the 1, 2, 3 and 4 years respectively. At the same time, environmental data are observed including temperature, relative humidity, dust fall, sulfate deposition rate, chloride deposition rate.

The OSD project (Abbott 2008) covers most of the US military bases around the world. Large amount of environmental data and corrosion data are obtained which can be used to evaluate the environmental severity index of different military bases. The materials studied in the project include different types of steel, copper, and aluminum alloy. The monitored environmental factors include relative humidity, rainfall, and chloride ion deposition rate. The exposure test time ranges from weeks to years, and the measured corrosion data include weight loss and pitting depth.

The RPF project (Panchenko and Shuvakhina 1983; Panchenko et al. 1985, 2017; Mikhailov et al. 2008) aims to study the corrosion of metals under severely cold environmental conditions. The 12 test sites involved in this project locate in the Far-Eastern regions of the USSR. The monitored environmental factors include temperature, relative humidity, and sulfur dioxide concentration. The corrosion loss rate of steel and zinc are measured. In addition, Russia has carried out research on the corrosion of metals in extremely cold environments in the Antarctic and sub-Arctic regions (Henriksen and Mikhailov 2002; Mikhailov et al. 2008).

These projects enriched the environmental and corrosion data and construct the corrosion map of different atmospheric environments around the world. However, the following problems may exist between these projects.

Firstly, the environmental factors monitored by each project are different. The difference makes it difficult to process the data uniformly. Secondly, as the chemical composition of the same material used in different projects may be different, there may be great differences in the corrosion behavior. Thirdly, as these international projects are carried out by different researchers in different countries and regions, the test procedure, standards, as well as data recording and reporting may be different (Panchenko and Marshakov 2017). In addition, the environmental corrosivity levels covered by each project are different. The ISO CORRAG, UN/ECE, and MICAT project covers a relatively narrow range of environmental corrosivity levels and the bias is skewed towards the lower end of the corrosivity distribution (Cai et al. 2018b). The OSD project represents a wider range of the severity conditions especially in some extremely corrosive environments. While the RPF project mainly focuses on the cold environment.

2.2 Laboratory corrosion tests

As the field exposure corrosion is time and expense consuming, laboratory corrosion test are carried out (Mendoza and Corvo 1999, 2000) to extrapolate the results to field exposure environmental conditions. The basic idea of the laboratory corrosion test is to capture the corrosion behavior in a shorter time by simulating or accelerating the actual environmental conditions. Generally, the existing laboratory corrosion test can be divided into two categories: one is based on the combination of environmental spectrum, and the other is based on the design of environmental factors.

2.2.1 Laboratory corrosion test based on combination of environmental spectrum

The basic idea of this test method is to divide the complex actual environment into several typical environmental sub-spectrums, such as salt spray spectrum, damp heat spectrum, cyclic wet-dry spectrum, gas erosion spectrum, and so on. Then these environmental sub-spectrums are appropriately combined to simulate the environment conditions actually experienced by the material. In each environmental sub-spectrum, the corrosion process is accelerated by increasing the magnitude of the environmental factor to shorten the test time. The salt spray test and the damp heat test consider the effects of salt deposition, temperature, and humidity on the corrosion process. The cyclic wet-dry test is used to simulate the process of electrolyte film formation and disappearance on the metal surface due to moisture condensation and evaporation. The gas erosion test is mainly used to evaluate the influence of sulfur dioxide, nitrogen dioxide, and other gas pollutants in the industrial atmosphere.

Mizuno et al. (2014) combines the humid stage, salt application stage, and the dry stage into a cyclic corrosion test procedure according to the SAE J2334 standard (2003) when study the corrosion resistance of automotive coated steel sheets. The proportion and environmental conditions (temperature, humidity, and salt solution concentration) of each sub-spectrum are different during the entire test period of 60 cycles. Qian et al. (2015) studied the relative corrosion resistance of ordinary carbon steel and four weathering steels through the cyclic dry-wet test. In a 1 h cycle, the pH of the immersion solution is kept at 4.4 at 45 °C for 12 min in the wet phase, and the surface temperature of the samples is kept less than 75 °C for 48 min in the dry phase. LeBozec et al. (2008) compared different test procedures according to six salt spray test standards and results shows that the corrosion performance of steel or zinc-coated steel is highly dependent on the testing conditions.

2.2.2 Laboratory corrosion test based on design of environmental factors

The basic idea of this method is to establish quantitative acceleration models between various environmental factors and the corrosion parameters. To achieve this, the actual environment is considered as the combination of several environmental factors which affect the corrosion process. The corrosion tests are conducted in several accelerated corrosion environments. In each test, the environmental condition is the combination of different levels of the environmental factors. Then quantitative acceleration models can be calibrated with the experimental results.

For example, when Wang et al. (2015) studied the effects of temperature and relative humidity on the atmospheric corrosion of zinc, the temperature was controlled at 285, 300, and 308 K with relative humidity at 53, 84, and 92%, respectively. And a log-linear model was established to correlate the measured corrosion current with temperature and relative humidity. Lin and Wang (2005) measured the corrosion weight loss of carbon steel under different chloride deposition rate (0/100/2500 $mg \cdot m^{-2} \cdot day^{-1}$) and temperature (288/298/308 K), and modified the Arrhenius model to establish an accelerated corrosion model for chloride deposition rate and temperature. LeBozec et al. (2004) obtained corrosion weight loss data of magnesium at different temperature (25/35 °C), relative humidity (75%/85%/95%), and chloride deposition (14/70/140/250 $\mu g\ m^{-2}$). The effect of the three environmental factors on the corrosion process was analyzed but no quantitative accelerated corrosion model was built.

At present, accelerated corrosion test cannot simply replace the atmospheric corrosion exposure test (Boelen et al. 1993; Damborenea and Conde 1995; Johansson and

Gullman 1995). Continuous improvements in the design of the test conditions and parameters are needed to have a good correlation with the actual field exposure corrosion test results (Cole 2017). The choice of the test methods should also base on the nature of the material and the actual atmospheric environment.

3 Corrosion influential factors and the effects

The atmospheric corrosion process of metals is simultaneously affected by multiple environmental factors (Kallias et al. 2016; Morcillo et al. 2013; Sabir and Ibrahim 2017), such as relative humidity (Esmaily et al. 2015; LeBozec et al. 2004; Samie et al. 2007; Wang et al. 2015), temperature (Kong et al. 2016; LeBozec et al. 2004; Samie et al. 2007; Wang et al. 2015), wetting time (Cole et al. 2011), rainfall (Corvo et al. 2005; Veleva et al. 2009), wind (Roberge et al. 2002), and pollutants (Castano et al. 2007; Esmaily et al. 2015; Feliu et al. 2003; Kim et al. 2004; Kreislova and Geiplova 2016; Qu et al. 2002; Tidblad et al. 2016).

3.1 Influence of relative humidity

The atmospheric corrosion rate is mainly determined by the electrochemical reaction in the electrolyte film on the metal surface. The electrolyte film forms when the ambient relative humidity is higher than the critical humidity level. Under this condition, the depolarization of oxygen can proceed smoothly and electrochemical corrosion happens with high corrosion rate. Below the critical relative humidity, the corrosion reaction nearly halts with no electrolyte film.

Relative humidity is the most important environmental factor which affects the atmospheric corrosion process. The effect of relative humidity on corrosion is complex. Multiple studies have indicated that the increase of relative humidity leads to the increase of corrosion rate (LeBozec et al. 2004; Samie et al. 2007; Shinohara et al. 2005) on clean surface without deposited salts. For example, the corrosion of magnesium alloys is accelerated significantly when the relative humidity increases from 75 to 95% (LeBozec et al. 2004). Studies have also shown that when the humidity increases from 53 to 92% (Wang et al. 2015), or from 40 to 90% (Shinohara et al. 2005), the corrosion current also increases following an approximate exponential relationship.

However, when the relative humidity is lower than a certain value, almost no corrosion occurs. This value is called the critical relative humidity (CRH). Studies have indicated nearly no corrosion occurs on carbon steel when the relative humidity is lower than 80–85% (Lapuerta et al. 2008; Nyrkova et al. 2013) when the surface is not contaminated by salts. This is because the corrosion reaction is an electrochemical reaction process, and the fundamental condition is that an electrolyte film must be formed on the metal surface to promote the cathodic and the anodic reactions.

Outdoor environments generally represent the most complex type of environment from an atmospheric corrosion point of view. In the actual outdoor environment, CRH is influenced by various factors, such as the type of the material, composition of the corrosion product, atmospheric pollutant concentration, and salt particle deposition (Van den Steen et al. 2016). Studies have shown that the CRH is about 70% for steel, zinc and nickel and about 76% for aluminum when the surface is clean. The CRH will be greatly reduced when the metal surface is covered by dust or corrosion products. Cole et al. (2004b) found that wetting occurs when surface RH exceeds the deliquescent RH (DRH) of the salts making up the contaminates and the DRH for different salts varies in a wide range from 35.0% for MgCl to 84.2% for Na_2SO_4.

Although the mechanism of the influence of relative humidity on the corrosion process is very complicated in the outdoor environment, there are some models that can be used to describe the acceleration effect or relative humidity under controlled laboratory conditions. In many applications, the Peck model (Escobar and Meeker, 2006) is used which is expressed as:

$$AF(RH) = \left(\frac{RH}{RH_0}\right)^A \tag{1}$$

where $AF(RH)$ is the acceleration factor, RH is the ambient relative humidity, RH_0 is the reference value of relative humidity, and A is a constant.

Klinger (2010) replaces RH with $RH/(1\text{-}RH)$, and the above model is changed to

$$AF(RH) = \left[\frac{RH(1-RH_0)}{RH_0(1-RH)}\right]^A \tag{2}$$

In the actual field environment, the relative humidity can be very close to one or even equal to one, and the above model will no longer be applicable. Therefore, the acceleration effect of relative humidity can also be described by the exponential model proposed by Vernon (1927) as

$$AF(RH) = \exp\{A \cdot (RH - RH_0)\} \tag{3}$$

In the actual engineering applications, the most suitable model can be selected from Eqs. (1)–(3) according to the fitting of the experimental data.

3.2 Influence of temperature

Temperature along with its change acts as another important factor affecting atmospheric corrosion. It is generally believed that when the relative humidity is lower than the CRH and no electrolyte film is formed, the influence of temperature on atmospheric corrosion is negligible. However, when the relative humidity is higher than CRH, the increase in temperature leads to the increase of the reaction activation energy and corrosion rate. Therefore, in the rainy season of tropical areas, the corrosion is quite serious. In addition, the fluctuation of temperature also has a great influence on atmospheric corrosion. The larger the temperature fluctuation, the easier for the condensation of the moisture to form the electrolyte film.

The influence of temperature on the corrosion process reflects in two ways: the direct influence on the corrosion rate, and the influence on the electrolyte film formation and evaporation.

Theoretically, atmospheric corrosion is an electrochemical process with cathodic and anodic reactions. The relationship between corrosion rate and ambient temperature can be described by the Arrhenius model. The increase of temperature results in the increase of the corrosion rate. For example, Lin and Wang (2005) conducted accelerated corrosion tests on three carbon steels and proposed a corrosion prediction model based on the Arrhenius model. The corrosion current of zinc also increases with increasing temperature (Wang et al. 2015). Esmaily et al. (2015) and LeBozec et al. (2004) conducted field corrosion tests on magnesium alloys and also found that temperature has a significant positive effect on increasing the corrosion rate.

Generally, under laboratory environment without considering the interactive effects between different factors, the effect of temperature on the corrosion process can be described by the Arrhenius model (Escobar and Meeker 2006), which is

$$AF(T) = e^{B\left(\frac{1}{T_0} - \frac{1}{T}\right)} \tag{4}$$

where $AF(T)$ is the acceleration factor, T is the ambient temperature (Kelvin), T_0 is the reference value of temperature. $B = Ea/K$ and Ea is the activation energy which can be estimated from the experimental data, K is the Boltzmann constant.

However, in the actual outdoor environment, the corrosion reaction can only happen when there is an electrolyte film on the metal surface. Therefore, as the condensation and evaporation of moisture is affected by the continuous changes of various environmental factors in the outdoor environment, the atmospheric corrosion reaction becomes a very complicated process. Generally, the metal surface undergoes wet-dry cycles every day (Cai et al. 2018a). The temperature decreases at night and the metal surface cools down, which is conducive for the condensation of moisture to form the electrolyte film and promote the corrosion reaction. During the daytime, as temperature increases the electrolyte film is easier to evaporate although the ambient absolute humidity also increases. Although there may be a positive effect due to the increased concentration of the electrolyte during the evaporation process (El-Mahdy and Kim 2004; Thee et al. 2014), the corrosion reaction will finally slow down or even stop after the electrolyte film disappears. But in some highly humid areas (tropical climates for example), as the humidity is very high and the metal surface is moist most of the time (Corvo et al. 2008), the increase of temperature can enhance the moisture concentration in the air and promote the corrosion process (Castano et al. 2010). In the winter, when the temperature drops below the freezing point, the electrolyte film freezes. The oxygen cannot reach the metal surface and the corrosion reaction hardly happens.

Moreover, in the outdoor environment, the influence of temperature is more complicated in the presence of other atmospheric environmental factors. For example, Lindstrom et al. (2000) found that when the concentration of carbon dioxide in the atmosphere is high, the change of temperature has little effect on the corrosion rate of zinc. For another example, when the nitric acid content in the atmosphere is high, the corrosion rate of copper is also independent on temperature (Samie et al. 2007). In addition, Cao et al. (2018) found that the effect of temperature on the corrosion process of aluminum alloys is more complicated in the marine atmospheric environment. When the temperature increases, the solubility of oxygen decreases and the corrosion product is more compact, the corrosion reaction slowed down as a consequence. However, temperature increase also leads to the increase of diffusion rate of oxygen and chloride ions, which accelerates the corrosion reaction. Therefore, when the outdoor environmental condition is more complicated, a specific analysis of the possible interactive effects between these environmental factors is required.

3.3 Influence of pollutants

There are many kinds of atmospheric pollutants in the atmosphere, including sulfur dioxide, nitrogen dioxide, hydrogen sulfide and dust. Among these pollutants, sulfur dioxide is one of the factors that has been most studied. The sulfur dioxide in the atmosphere is mainly contributed by the combustion of fossil fuel.

Generally, when the sulfur dioxide concentration is high, the increase of the SO_2 concentration in the air leads to the increase of the corrosion rate (Klinesmith et al. 2007; Qu et al. 2002). Kim et al. (2004) found that the corrosion rate of copper and steel exposed in the field atmospheric environment is basically proportional to the concentration of sulfur dioxide, and the mathematical relationship can usually be captured by a power function (Klinesmith et al. 2007; Mikhailov et al. 2004; Tidblad et al. 2002). Walter (1991) conducted a simulated atmospheric corrosion test in the laboratory and found that the dissolved sulfur dioxide in the water will be oxidized to sulfate ions which can accelerate the corrosion reaction. Cao et al. (2013) also found that sulfur dioxide can accelerate the corrosion process of carbon steel, and summarized the acceleration mechanism as the following points: (a) the cathodic reaction is more active because the solubility of sulfur dioxide is much higher than oxygen (about 1300 times); (b) the presence of sulfate leads to the decrease of CRH and the time-of-wetness of the metal surface is prolonged; (c) sulfur dioxide can act as a catalyst to promote the dissolution of the iron atoms. In addition, sulfur dioxide also has a great influence on the long-term corrosion kinetic. The obtained results from a exposure project for more than 45 years (Kreislova and Knotkova 2017) confirm the significant dependence of atmospheric corrosion on SO_2 air pollution. And both the short- and long-term atmospheric corrosion rate of carbon steel depends on SO_2 pollution level much more than zinc and copper. When the SO_2 concentration is low or the concentrations of other pollutants are also high, the effect of SO_2 would be more complicated as other factors may become the controlling factor and interactive effects becomes significant (Castano et al. 2007).

Chloride is another important atmospheric pollutant that affects the corrosion process. The deposition of chloride on metal surfaces can significantly improve the corrosion rate of various metals, including carbon steel (Lin and Wang 2005; Yang et al. 2017), zinc (Qu et al. 2002), aluminum alloy (Wang et al. 2018), magnesium alloy (Esmaily et al. 2015; Lin and Wang 2005; LeBozec et al. 2004), and so on. The effect of chloride on atmospheric corrosion rate is mainly reflected in the following aspects. Firstly, it is easy for chloride ions to adsorb on the metal surface because chloride ion has small hydration energy. Chloride ions will help to cause breakdown and create imperfections of the protective film, which promotes the corrosion reaction (Ambat et al. 2000; Zhao et al. 2008). On the other hand, the deposited chloride salt makes metal surface strongly hygroscopic (Van den Steen et al. 2017) and it is easier to form the electrolyte film and increase the wetting time. At the same time, the conductivity of the electrolyte film increases and the corrosion reaction is accelerated. Mathematically, the acceleration effect of chloride on corrosion rate are usually described by a power function (Klinesmith et al. 2007; Mikhailov et al. 2004; Tidblad et al. 2002) or a quadratic function (Lin and Wang 2005; Qu et al. 2002). In addition, chloride may also influence the corrosion kinetics as well as corrosion product. Ma et al. (2009) reported that the low carbon steel exposed to marine environment follows the equation $C = At_1^{B_1-B_2}t^{B_1}$ $(t \geq t_1)$ which significantly deviate from the well-known power equation. The turning point will move onwards as the amount of chloride increases. In marine site with high amount of chloride deposition, β-FeOOH is produced as for chloride accelerative effect, while the main corrosion products are γ-FeOOH and α-FeOOH in exposure station with low or no chloride.

Chloride mainly comes from the ocean and deicing (ISO, 2012a).The deposition rate of chloride from the ocean is affected by wind speed and wind direction (Cole et al. 2003b; Li and Hihara 2014; Roberge et al. 2002), distance from the coast (Cole et al. 2003b; Cole et al. 2003a; Guerra et al. 2019), rainfall (Cole et al. 2004a), temperature and humidity (Castaneda et al. 2018; Cole et al. 2003b), terrain (Cole et al. 2003c; Cole et al. 2004a) and other factors (Cole et al. 2011). The influence of chloride has a significant relationship with the distance from the sea (Abbott 2008; Feliu et al. 1999). As the distance from the sea increases, the deposition rate of chloride decreases rapidly (Cole et al. 2003b) and its impact on corrosion is rapidly weakened. Stronger wind can promote both the production and transportation of sea salt aerosols (Cole et al. 2003b). Airborne salinity generally increases with increasing wind velocity, and the effect becomes very significant when a critical velocity is exceeded. A range of critical wind velocity values (3–7.1 m/s) has been reported in the literature, but no consensus has been reached (Morcillo et al. 2000; Piazzola and Despiau 1997; Spiel and De Leeuw 1996). Meira et al. (2007) recommended 3 m/s as the critical wind velocity for a significant increase in airborne salinity. The effect of humidity on the salt concentration is also very significant, particularly at the higher humidity. Significant decrease in transportation is observed when the surface relative humidity is increased from 50 to 70% mainly due to the increase of aerosols mass and decrease of the vertical dispersion (Cole et al. 2003b). The effect of sheltering by headlands and cliffs is evident at coastal areas (Cole et al. 2003a) and the transport of aerosol is influenced by the roughness of landforms and buildings (grass, forest, buildings, and so on) (Cole et al. 2003b,c, 2004a).

The dose-response function proposed by Mikhailov et al. (2004) and Tidblad et al. (2002) from the results of the ISO CORRAG project can be used without considering the influence of other atmospheric pollutants and the interaction between different pollutants. The accelerated corrosion model for sulfur dioxide and chloride are

$$AF(S) = \left(\frac{S}{S_0}\right)^E \tag{5}$$

$$AF(Cl) = \left(\frac{Cl}{Cl_0}\right)^G \tag{6}$$

where $AF(S)$ and $AF(Cl)$ are the acceleration factors for sulfur dioxide and chloride, respectively. S_0 and Cl_0 are the reference values. E and G are constants which can be estimated from experimental data.

Acidified aerosol is another important pollutant which has the potential to make a significant contribution to the atmospheric corrosion process (Cole et al. 2009). Acidified aerosol mainly exists in industrial and marine atmosphere. Aerosol transportation and deposition are size dependent, with aerosol deposition increasing with aerosol size and decreasing with transportation distance. While gas absorption may occur relatively close to the source, acidified aerosols can be transported over some distance from the original gaseous source. The aerosols are characterized by low pH, fine size (typically 1–100 mm) and high dissolved ionic salt content. The deposition of aerosols is primarily controlled by wind turbulence and is a function of turbulence intensity, wind speed, object shape, and aerosol size (Cole and Paterson 2004).

The types, size and distribution of aerosols can influence the initiation and propagation of the corrosion process in more complex ways (Li and Hihara 2014; Risteen et al. 2014). Take zinc as an example, the presence of acidified aerosols may lead to enhanced corrosion by disrupting any protective oxide films and subsequently establishing electrochemical cells, relatively uniform oxide layers are formed, which may be dissolved by typical aerosol with pH values of 1–4 and slightly acidified rain (<5) (Azmat et al. 2011) and thus may not constitute as an effective barrier against corrosion. In addition, the susceptibility of zinc to chloride containing environments is found may in part be associated with the effect of acidified marine aerosols. A multiscale modeling approach is proposed to build a map of airborne salinity and zinc corrosion rates in Australia (Cole et al. 2011), which is useful in material selection, maintenance programs, and corrosion resistant materials development. Additional research is required both to directly determine the corrosion caused by marine aerosols and to define the geographic spread of such aerosols.

3.4 Influence of other factors

As the sulfur dioxide concentration in some regions has been decreasing in the recent decades, the influence sulfur dioxide is more likely to be correlated with other factors like nitrogen dioxide (Castano et al. 2007). Different concentrations of SO_2 and NO_2 may lead to the change of the corrosion mechanism. The combined effect of SO_2 and NO_2 on the corrosion process may be insignificant (Kucera 2003; Tidblad 1991), inhibitive (Henrikksen and Rode 1986; Takazawa 1985) or dependent on many factors, including relative humidity (Arroyave and Morcillo 1996; Ericsson and Johansson 1986), sulfur dioxide concentration (Feliu et al. 2003; Castano et al. 2007), and the type of material (Castano et al. 2007).

The effect of dust on corrosion acts in different ways depending on the type of the dust. Some dust particles are soluble and corrosive (salt containing particles). They become corrosive media and accelerate the corrosion rate when dissolved in the electrolyte film (Lin and Zhang 2004). Some dust particles are non-corrosive and non-soluble (such as carbon particles), but it can adsorb corrosive particles and promote corrosion reaction. Some dust particles are neither corrosive nor adsorptive (such as soil particles), but gaps are formed when they fall on the metal surface and trigger localized corrosion (Wang et al. 2019). The particle size may also be an important factor that influences the initiation and expansion of the corrosion process. Small particles are more likely to induce the initial corrosion while the final corrosion area cannot expand too much. In contrast, the corrosion area influenced by large particles is able to intensively expand although the initial corrosion rate is low (Wang et al. 2019).

The effect of rainfall on atmospheric corrosion can be both positive and negative at the same time. On one hand, rainfall prolongs the wetting time but may also help to wash away the dissolvable corrosion product on the metal surface (Azmat et al. 2011). On the other hand, the rain can wash away the pollutants (Abbott 2008; Cole and Paterson 2007) if drops on the surface pass a critical size. Cole and Paterson (2007) have found that the average rain required to initiate drop movement is 1.3 mm and the average rain required to clean a surface to 10 and 1% of the original pollutants is 1.5 and 3 mm respectively. As a consequence, hygroscopicity of the metal surface and the corrosiveness of the electrolyte film are reduced.

Apart from the environmental factors, the chemical composition (Cano et al. 2017; Diaz et al. 2018; Liao and Hotta 2015), microstructure (Liao and Hotta 2015), rust characterization (Kinugasa et al. 2016), and other factors of the tested material also have great impact on the corrosion resistance. Different materials exhibit different atmospheric corrosion behaviors and different sensitivity to the environment.

4 Modelling of the atmospheric environment

The atmospheric environment is composed of a variety of factors. As the environment changes continuously along with time and space with interactive and random effects, each factor cannot be characterized by only one or several parameters. Instead, quantitative models are needed to describe the fluctuation, dependency, randomness, and spatial difference of the actual environment. The field environment modelling methods mainly considers the following two topics: modeling the fluctuation along with time, and modeling the spatial difference at different locations.

4.1 Modeling the environmental fluctuation along with time

The field atmospheric environment changes along with time with both periodic and random components. For example, the temperature undergoes daily cycles from day to night and annual cycles from summer to winter. Various unknown factors may also influence the temperature in random patterns. Meanwhile, different factors may be dependent on each other. For example, the relative humidity is a function of the absolute humidity (water vapor content in the air) and temperature. Generally, it is inversely proportional to the temperature when the absolute humidity stays constant (Cai et al. 2018a).

In this section, the literature review will give a summary on the modeling method of the field environment. As the influence of the environment is a common problem in many research areas, the review is not restrained within the field of corrosion research. Generally, the current methods for modeling the field environmental fluctuation can be divided into three categories: the mean value method, the distribution method, and the time-varying function method.

4.1.1 The mean value method

The basic idea of this method is using the statistical mean value to represent the actual condition of each environmental factor. In the commonly used corrosion prediction models, such as the linear (Knotkova et al. 1995; Morcillo 1995) and log-linear (Knotkova et al. 1995, 2002; Panchenko and Marshakov 2017; Roberge et al. 2002) dose-response functions, environmental variables such as temperature, relative humidity, sulfur dioxide concentration, and chloride deposition rate are all described by the annual average values. In other studies considering the impact of the environment, the use of the mean value to describe the field environment is also very common. For example, some set the time-varying environment to be a worst-case steady-state ambient (Lam et al. 2007). Wang (2003) approximates the field operating environment as the combination of multiple discrete stages, and the environmental factors remain constant in each stage.

The mean value method is simple and convenient to be used. It has good prediction accuracy in the cases where the fluctuation of the environmental factors is small or the corrosion effects of the environment are approximately linear. However, if the corrosion effect is highly nonlinear and the environment is highly fluctuated, the mean value method will result in large prediction error.

4.1.2 The distribution method

This method considers the variation of the environmental factors and establishes a probability density function (PDF) for each environmental factor. The total effects of different environmental factors can be calculated through the integration of the corrosion effect within the entire distribution range of each environmental factor.

This method has been widely used in the studies considering the influence of the environment. For example, Meeker et al. (2009) and Hong and Meeker (2010a) built distribution models of product usage rates and estimated reliability by taking the usage frequency into the failure rate function. Monroe and Rong (2010) used the Normal distribution and the Beta distribution to fit the temperature and relative humidity respectively to predict the product reliability in a dynamic natural environment. Liu et al. (2013) used the Gamma distribution to fit the distribution of temperature during storage and estimated the product reliability under non-constant temperature conditions. Wang (2003) proposed a reliability assessment method for products used in the natural environment and describe the environmental factors with mean and variance of random distributions. Cai et al. (2018a, 2020a,b) built a dependent distribution model to describe the conditional PDF of relative humidity under different values of temperature to study the influence of different environmental factor on atmospheric corrosion in dynamic environment.

The distribution method considers the variation of the environmental factors. Using distributions to describe the environmental factors is closer to the actual situation, which improves the accuracy of the corrosion prediction model, especially when the corrosion effects of the environmental factors are nonlinear. However, most of the studies did not consider the correlation between different environmental factors. When multiple correlated environmental factors have influence on the corrosion process at the same time, the independent distribution model will result in great prediction error.

4.1.3 The time-varying function method

The time-varying function method describes the change of environmental factors along with time and extracts information directly from the observed environmental data. In this method, the periodicity and randomness of each factor can be captured and the dependency between different factors can also be considered by correlating each factor with time. For instance, Chan (2001, 2008) built a time-varying model for the sunlight and temperature by fitting the weather observation data with the ARMA time series method to estimate the reliability of coating in the dynamic field environment. Gebraeel and Pan (2008) established a time-varying environmental model to predict the change of products performance under actual use environmental conditions. Hong and Meeker (2010b, 2013) also studied the dynamic environmental data and the effects of environmental factors on product performance changes.

By adding corresponding items and controlling the model parameters, the time-varying method can capture different degrees of periodicity and randomness of the environment with high accuracy. Time *t* is the common variable in the models and can act as the bridge to capture the dependency between different environmental factors.

4.2 Modeling the spatial difference at different locations

The above methods only describe the changes of environmental factors at fixed locations along with time. However, in the actual conditions, the environmental conditions may change at different locations. For example, oceangoing vessels may experience both low and high levels of temperature and salinity in different seawaters. Therefore, it is necessary to study the spatial modeling method of the natural atmospheric environment. The spatial modeling of the environmental factors is based on the monitoring data at different sites. As these sites are geographically discrete, two categories of methods are built. One is the cluster analysis method and the other is the interpolation method.

The cluster analysis method divides the environmental factors of different regions into several categories or several levels and then evaluates the environmental severity. For example, Austin et al. (2012) proposed an improved hierarchical cluster method to validate the distinct air pollutant mixtures. Wang et al. (2013) proposed two spatial-temporal clustering algorithms and applied them to analyze the ozone pollution which achieved good results. With the continuous improvement of the cluster analysis method, the classification of environmental severity is more and more scientific and accurate. However, there are some problems when it is used for the corrosion prediction especially when the corrosion effects are highly nonlinear. Great prediction error may occur due to the discretization of the continuously distributed environmental parameters.

The interpolation method is a promising tool as it can obtain continuous results of various environmental factors within the entire distribution ranges. For example, Kilibarda et al. (2014) made predictions of the mean, maximum, and minimum temperatures using spatial-temporal regression-Kriging, and the average prediction accuracy is within ±2 °C. Aryaputera et al. (2015) used the spatial-temporal Kriging method to predict the short-term solar irradiance of the island of Hawaii, which improved the prediction accuracy compared to the conventional parametric models. Susanto et al. (2016) proposed a distribution-based distance weighting spatial interpolation method, which improves the interpolation accuracy compared to the ordinary Kriging, inverse distance weighting, and the triangular irregular network spatial interpolation techniques. With the continuous development and improvement of the interpolation method, the interpolation accuracy and efficiency are continuously improved and this method can be applied to the spatial modeling of various environmental factors.

There is another important method which does not entirely depend on the observation data and is constructed with physics-based models. The model mainly describes the relationship between the target factor and its' controlling factors. For example, Cole et al. (2004a) proposed a geographic information system (GIS) for predicting airborne salinity. The GIS is based on several mathematical models which links the aerosol production with wind frequency, direction, and speed, as well as aerosol transportation with distance, relative humidity, terrain, rainfall, and so on. With the help of the GIS model, the target factor can be predicted as long as the controlling factors are known. Cole et al. (2004a) built a salinity map and a corrosion rates map of zinc (Cole et al. 2011) of Australia. This is a promising method for environmental corrosivity classification and material corrosion resistance assessment.

5 Corrosion kinetic and corrosion prediction models

The corrosion data have several typical characteristics such as small sample size, large scatter, and influenced by multiple environmental factors. To construct the corrosion prediction model, two parts of work need to be done: build the corrosion development model over time (the corrosion kinetic model), and establish corrosion performance model in different environments (the corrosion effect model).

5.1 Atmospheric corrosion kinetic models

Due to the relatively slow corrosion rate in the field environment, long time exposure tests are required to obtain reliable long-term corrosion data. Sometimes the tests are longer than 20 years. Therefore, analyzing the existing corrosion data from the exposure test and establishing a corrosion kinetic model to predict the long-term corrosion development has become an important research topic. Existing corrosion kinetic models include the power function models (Fuente et al. 2007; Morcillo et al. 2013; Sun et al. 2009), the general linear models (Fuente et al. 2011; Ma et al. 2010; Morcillo et al. 1993), and the power-linear models (Panchenko and Marshakov 2016; Panchenko et al. 2014, 2017; Sabir and Ibrahim 2017).

5.1.1 The power function model

The atmospheric corrosion loss C over time t is usually described by the following power function

$$C = C_1 t^n \tag{7}$$

where C_1 and n are constant. C_1 represents the amount of corrosion in the first year. n is the exponent which characterizes the protective effect of corrosion products. The lower n is, the more protective the corrosion product layer on the metal surface.

Since the model contains only two parameters, it is widely used and the corrosion data of specific materials under specific environmental conditions can be accurately fitted. For example, using this model to fit the 8-year corrosion data at different testing sites in the ISO CORRAG project, the correlation coefficients are all above 0.95, which is similar to the results in the literature (Panchenko and Marshakov 2016; Benarie and Lipfert 1986). The power function model can also be well fitted for field exposure corrosion data up to 20 years (Fuente et al. 2011; Sun et al. 2009).

In the ISO 9224 standard (ISO 2012b), the values of exponent n are calculated from the exposure data of the ISO CORRAG program. For carbon steel, zinc, copper, and aluminum, the generalized n values are 0.523, 0.813, 0.667, and 0.728 respectively. Considering the uncertainty in the data, the conservative upper 95% confident limit of corrosion attack can be estimated using n values of 0.575, 0.873, 0.726, and 0.807.

5.1.2 The general linear model

During long-term corrosion process, as the corrosion products form continuously, the metal surface is protected from the corrosive medium (Fuente et al. 2011; Zhang et al. 2003). The corrosion rate gradually decreases and hence the exponent n becomes smaller. In view of this, the conventional power function model is no longer appropriate to be used for long-term corrosion prediction because a large error may be generated. Therefore, some researchers used a two-stage log-linear model (Fuente et al. 2011; Ma et al. 2010; Morcillo et al. 1993; Surnam et al. 2015) to extrapolate the short-term corrosion test data to predict the long-term corrosion development (Morcillo et al. 1993). In the two-stage log-linear model, the atmospheric corrosion kinetic process is approximated to an initial stage and a steady stage. The model is expressed as

$$\log C = \begin{cases} n_1 \log t & , t \le t_1 \\ n_1 \log t_1 + n_2 \log(t/t_1) & , t > t_1 \end{cases} \tag{8}$$

where n_1 and n_2 are the exponent of the initial stage and the stationary stage respectively. t_1 is the time when the initial stage ends. The values of n_1 and n_2 are significantly influenced by the type of atmosphere and the compactness of the rust layers (Fuente et al. 2011; Surnam et al. 2015). And the value of t_1 varies for different metals in different atmospheres.

In the ISO 9224 standard (ISO 2012b), the bi-linear model is employed to make the long-term corrosion prediction for carbon steel, zinc, copper, and aluminum. The model is expressed as

$$C = \begin{cases} r_{\mathrm{av}} \cdot t & , t \le 10 \\ r_{\mathrm{av}} \cdot 10 + r_{\mathrm{lin}} \cdot (t - 10) & , t > 10 \end{cases} \tag{9}$$

where r_{av} is the corrosion rate in the first 10 years. At the steady stage after 10 years, corrosion loss increases with the average rate r_{lin}. The upper limits of r_{av} and r_{lin} for carbon steel, zinc, and copper are given corresponding to each corrosivity category from C1 to CX according to the ISO 9223 standard (ISO 2012a).

5.1.3 The power-linear model

According to a large number of field exposure corrosion test data (Fuente et al. 2007, 2011; Panchenko 2013; Shaw and Anderson 2010; Sun et al. 2009), the actual atmospheric corrosion process can be divided into three stages. The first stage is the initial stage, which is the stage when the corrosion reaction begins to occur. The second stage is the transition stage and the relationship between the corrosion loss and exposure time can be described by a power function. The third stage is the steady stage, and the amount of corrosion increases linearly with time (Teng et al. 2015). Therefore, the corrosion process can be described by the following power-linear model

$$C = \begin{cases} C_1 t^n & , t \le t_s \\ C_{t_s} + \alpha (t - t_s) & , t \le t_s \end{cases} \tag{10}$$

where t_s is the time to reach the steady stage, C_{ts} is the cumulative amount of corrosion when the steady stage is reached, and α is the stationary corrosion rate. Compared to the power function model, this model can be used for both short-term and long-term corrosion prediction with higher accuracy.

It can be seen from Eq. (10) that the accuracy of the short-term prediction depends mainly on parameters C_1 and n, while the long-term prediction depends mainly on the stabilization time t_s and the stationary corrosion rate α. Parameters C_1 and n can be obtained by regression analysis with satisfied goodness of fitting at specific locations. However, it is difficult to accurately estimate parameters t_s and α. The reason is that the effects of the environmental factors on the long-term corrosion process are rather complicated under different environmental conditions.

The stabilization time t_s differs a lot for different metals in various types of atmosphere (Díaz et al. 2012; Fuente et al. 2011; Melchers 2008). For example, t_s for low alloy steel (Díaz et al. 2012) ranges between 4 and 8 years corresponding to corrosivity categories C2–C5 according to ISO 9223 standard. In the ISO 9224 standard, the stabilization time t_s is set to be 20 years. In most cases, the steady stage can be reached within 8 years. The value of the stabilization time t_s can be calculated with $t_s = 1/(0.9^{(1/n-1)}-1)$ according to Morcillo's criterion (Morcillo et al. 2013). When n increases from 0.1 to 0.9, the stabilization time t_s decreases from 8.1 to 0.5 years (Cai et al. 2018b). This result coincides with the ranges reported in the literature (Díaz et al. 2012).

For the stationary corrosion rate α, Panchenko and Marshakov (2016) set t_s as 6 years and estimated α with the following equation

$$\alpha = \frac{C_8 - C_6}{2} \tag{11}$$

$$\alpha = \left.\frac{\mathrm{d}C}{\mathrm{d}t}\right|_{t=t_s} = C_1 \cdot n \cdot t_s^{n-1} \tag{12}$$

where C_6 and C_8 are the amount of corrosion after 6 and 8 years of exposure which can be calculated with Eq. (7). Eq. (11) regard the average corrosion rate from the 6th year to the 8th year as the stationary corrosion rate. In Eq. (12), the instantaneous corrosion rate at the 6th year is considered as the stationary corrosion rate. The difference between the α values obtained with Eq. (11) and Eq. (12) is insignificant (Panchenko and Marshakov 2016).

5.2 Corrosion prediction in various atmospheric environment

The atmospheric corrosion process is influenced by many environmental factors, such as temperature, humidity, pollutants, and so on. However, the above corrosion kinetic models are only valid at specific locations. When the environmental condition changes, the model is no longer applicable. Therefore, it is important to establish quantitative relationship which captures the influence of these environmental factors on the corrosion process to make corrosion prediction in different environments. The existing models include the dose-response function, the multifactor combination model, the response surface model, and the artificial neural network model.

5.2.1 Dose-response function

The dose-response function is the most widely used model for the atmospheric corrosion prediction. It predicts the corrosion amount according to the corrosivity of the atmosphere (including time, temperature, relative humidity, wetting time, pollutant concentration, and so on) (Tidblad 2012). The current dose-response equation is empirical formulas based on the regression analysis of the field exposure corrosion test results and the environmental parameters at the testing sites. Generally, the corrosion kinetics can be described by the power function shown in Eq. (7), and the parameters C_1 and n are affected by environmental factors (Dean and Reiser 2002).

Numerous studies have shown that environmental factors such as temperature, relative humidity, wetting time, SO_2 concentration, and Cl^- concentration have great impacts on the corrosion rate (Arroyave et al. 1995; Feliu et al. 1993a,b; Mendoza and Corvo 2000). These environmental factors are introduced into the dose-response function and the interaction between these environmental factors are also considered.

The basic form of the most commonly used dose-response function follows the simple linear (Knotkova et al. 1995; Morcillo 1995) or log-linear relation (Knotkova et al. 1995, 2002; Panchenko and Marshakov 2017; Roberge et al. 2002)

$$C = a + b \cdot SO_2 + c \cdot Cl + d \cdot TOW \tag{13}$$

$$\ln C = a + b \cdot SO_2 + c \cdot \ln Cl + d \cdot \ln TOW \tag{14}$$

where a, b, c, and d are constant, SO_2 is the concentration of sulfur dioxide (μg.m^{-3}), Cl is the chloride deposition rate (mg.m^{-2} day^{-1}), and TOW is the wetting time (h).

Different researchers have extended the basic models to various forms which have different definitions of the type, meaning, and description of the environmental variables. These dose-response functions are listed in Table 1. Here a recapitulative discussion of the difference between these models are made as the following.

(i) Different models include different environmental factors. Some models only include temperature, relative humidity, and sulfur dioxide concentrations while some also include factors such as rainfall and hydrogen ion content in rainwater; (ii) Different models choose different variables to describe the same environmental factor. For example, for the time-of-wetness, some models only

Table 1: Dose-response functions according to previous publications.

References	The model	Note
(Morcillo 1995; Knotkova et al. 1995)	$C = a + b \cdot SO_2 + c \cdot Cl + d \cdot TOW$	The basic linear model
(Knotkova et al. 1995, 2002; Panchenko and Marshakov 2017; Roberge et al. 2002)	$\ln C = a + b \cdot \ln SO_2 + c \cdot \ln Cl + d \cdot \ln TOW$	The basic log-linear model
(Mendoza and Corvo 1999)	$C = a + (b\tau_{5-25} + c\tau_{25-35} + dt_{\text{rain}})Cl + (e\tau_{5-25} + f\tau_{25-35} + gt_{\text{rain}})SO_2 + hR/t_{\text{rain}}$	τ_{5-25}: time of wetness at 5–25°C, h; τ_{25-35}: time of wetness at 25–35°C, h; t_{rain}: time of rainfall, h; Cl: Cl$^-$ deposition rate, mg·m^{-2}·day^{-1}; SO_2: sulfur compounds deposition rate, mg·m^{-2}·day^{-1}; R: amount of rainfall, mm.
(Feliu et al. 1993a)	$C = a_1 + a_2t_w + a_3RH + a_4D + a_5T + a_6Cl + a_7SO_2$	t_w: wetness time, annual fraction; RH: annual average relative humidity, %; D: raining days of a year, day; T: annual average temperature, °C; Cl: Cl$^-$ deposition rate, mg.m^{-2}.day^{-1}; SO_2: SO_2 deposition rate, mg.m^{-2}.day^{-1}.
(Tidblad et al. 2001)	$C = f_{\text{dry}}(T, RH, SO_2, O_3) \cdot t^k + f_{wet}(R, H^+) \cdot t^m$	f_{dry}: the dry deposition term; f_{wet}: the wet deposition term; O_3: O_3 concentration, μg.m^{-3}; H^+: H^+ concentration, mg.L^{-1}; t: corrosion time, year. R: amount of rainfall, mm.
(Kucera et al. 2007)	$C = 1.82 + (0.041R \cdot H^+ + 1.37N)t + (1.71 + 0.471SO_2^{0.22}e^{0.018RH}e^{f(T)})t$ $f(T) = \begin{cases} 0.062(T-10) & , T < 10^{\circ}\mathrm{C} \\ -0.021(T-10) & , T > 10^{\circ}\mathrm{C} \end{cases}$	This model is built for zinc. Models for carbon steel, copper, bronze, and limestone can be referred in Ref. (Kucera et al. 2007). SO_2: SO_2 concentration, μg.m^{-3}; R: amount of rainfall, mm.year^{-1}; H^+: H^+ concentration, mg.L^{-1}; N: HNO_3 concentration, μg.m^{-3}.
(Mikhailov et al. 2004; Tidblad et al. 2002; Tidblad 2012)	$C = f_{\text{dry}}(SO_2) + f_{\text{dry}}(Cl) + f_{\text{wet}}(H^+)$ $f_{\text{dry}}(SO_2) = ASO_2^Be^{c_1(RH)+g_1(T)}$ $f_{\text{dry}}(Cl) = DCl^Ee^{m_1(RH)+k_1(T)}$	$f_{\text{dry}}(SO_2)$: the effect of dry depositions of sulfur dioxide; $f_{\text{dry}}(Cl)$: the effect of dry depositions of chlorides; $f_{\text{wet}}(H^+)$: the effect of wet depositions of hydrogen ions (acid rains). Cannot be considered in the existing dose–response function due to the shortage of data.

contain one variable *TOW*, while some contain two variables τ_{5-25} and τ_{25-35}. As for rainfall, some models consider the amount of rainfall, while others consider the time of rainfall; (iii) Different models have different ideas for the construction of the models. Some models do not distinguish the source of corrosion and simply include all the environmental variables into the model at the same time, while others divide the source of corrosion into several parts according to the cause of corrosion; (iv) Different models cover different periods of corrosion data and different ranges of environmental corrosivity. Some models are based on short-term corrosion test within a narrow environmental corrosivity range, while other models cover a wider range of environmental severity levels on a global scale with a longer period of test time.

These dose response functions are all designed to describe the relationship between environmental factors and parameter C_1. The influence of environmental factors on parameter n is more complicated and difficult to describe with quantitative models. Currently, only limited number of literatures have explored the relationship between parameter n and the environmental factors (Cai et al. 2019; Dean and Reiser 2002; Panchenko et al. 2014; Panchenko and Marshakov 2016).

Dean and Reiser (2002) used a regression analysis method for the ISO CORRAG data to establish the relationship between exponent n and environmental variables including time-of-wetness (*TOW*), sulfur dioxide concentration (SO_2), and chloride deposition rate (Cl). The results indicate that for different metals, parameter n is affected by different environmental factors. For example, for carbon steel, parameter n is significantly affected by the *TOW*, while for zinc and aluminum, parameter n is hardly affected by any of the three environmental variables. For copper, parameter n is affected by both *TOW* and *Cl*.

Panchenko (2014, 2016) gives the relationship between n and the C_1 based on the analysis of the long-term corrosion data from the ISO CORRAG program, which is

$$n = A(BC_1 - G)^2 + D \tag{15}$$

where A, B, G, and D are constants. The values differ under different types of atmosphere and be referred in the literature (Panchenko and Marshakov 2016). When Eq. (15) is used, the first step is to determine the type of atmosphere according to the ISO-9223 standard based on the levels of sulfur dioxide concentration and chloride deposition rate. Then the value of n can be calculated with C_1 as input. Although the form of the model is general, the prediction accuracy is not high due to the limited sample size and scatter of the data. Cai et al. (2019) built a hierarchical linear model to predict the corrosion growth along with time in different environments of carbon steel, zinc, and copper of the ISO CORRAG program. The result shows that, statistically, only the exponent n of copper shows significant positive effects on sulfur dioxide. It is obvious that no consistent conclusions are drawn for now and much more work is needed.

5.2.2 Multi-factor combination model

The dose-response function directly correlates all the influential environmental factors with the corrosion parameters to describe the comprehensive effect of the environment. Many researchers also start from the effect of each environmental factors and establish the model by combining all the individual models together (Haynie and Upham 1974; Hakkarainen and Ylasaari 1980; Knotkova et al. 1984). These models have different structures. Some of them are not very rational and need to be improved to avoid erroneous effects that they yield (Klinesmith et al. 2007). At the same time, most of these models only include the effect of one or two environmental factors and corrosion time is not considered.

Based on these models, Klinesmith et al. (2007) constructed a model with a more rational structure including time-of-wetness (h), sulfur dioxide concentration (μg m^{-3}), chloride deposition rate (mg m^{-2} day^{-1}), and corrosion time (years), which is

$$K = At^B\left(\frac{TOW}{C}\right)^D\left(\frac{1+SO_2}{E}\right)^F\left(\frac{1+Cl}{G}\right)^H e^{J(T+T_0)} \tag{16}$$

where A, B, C, D, E, F, G, H, J, and T_0 are all constant coefficients and are calculated from the ISO CORRAG data. The results show that for carbon steel, copper, and zinc, the prediction ability is better, while for aluminum, the prediction error is large. The reason is that the corrosion of aluminum is mainly pitting corrosion. The corrosion process is slow and uneven, resulting in large scatter of the corrosion data.

With the same procedure, the multi-factor combination model can also be established with the commonly used acceleration models (Escobar and Meeker 2006) for different individual environmental factors. The Arrhenius model, the Eyring model (David and Montanari 1992), the Coffin-Manson model (Cui 2005), and the inverse power law model (Nelson 1975) are the most commonly used for different factors. Based on these models, some new models are proposed to describe the corrosion effects of various environmental factors.

For example, Nguyen et al. (2013) used the Arrhenius model to describe the effect of temperature on corrosion rate and a power function to describe the effect of relative humidity and salt deposition. The power function is employed by Cole et al. (2011) to predict the weight loss of zinc under different salt deposition rate. Corvo et al. (2005) also used a power function to describe the effect of chloride deposition rate and rainfall on weight loss in the marine atmosphere. Qu et al. (2002) used the exponential function and the quadratic polynomial to correlate temperature and salt deposition with weight gain of zinc. Revie (2008) described the influence of salinity on the immersion corrosion rate in seawater by using the truncated normal distribution probability density function (PDF). This model can also be used to describe the effect of temperature in the field environment which increases first and then decreases due to the interaction between different environmental factors.

These models can be used to establish the following types of comprehensive corrosion effect models: the combined model, the generalized Eyring model, and the generalized log-linear model.

The basic assumption of the combined model is that the acceleration effects of different environmental factors are independent. It is simplified compared to the actual situation without considering the interactive effect between different factors. For example, Soares et al. (2005) proposed a correction factor model for marine corrosion. The model considered the seawater temperature T, dissolved oxygen DO, salinity S, pH, and water velocity V based on a large number of actual corrosion data. All the correction factors are combined together and the actual correction coefficient K is

$$K = f(T)f(DO)f(S)f(pH)f(V) \tag{17}$$

For atmospheric corrosion, considering the environmental factors of relative humidity RH, temperature T, sulfur dioxide concentration SO_2, chloride deposition rate Cl, the combined corrosion model can also be established by the above method. This has been done by Soares et al. (2009) to study the influence of RH, T, and Cl on corrosion in marine atmospheric environment. And the corrosion depth of ship structures in different trading routes is predicted.

The generalized Eyring model also doesn't consider the interactive effects between various environmental factors. The model follows

$$\xi = \alpha \cdot \exp\left(\frac{-\beta}{T}\right) \cdot \exp(\gamma \cdot RH) \cdot \exp(\eta \cdot SO_2) \cdot \exp(\lambda \cdot Cl) \tag{18}$$

The difference between the generalized log-linear model and the generalized Eyring model is the form of the temperature acceleration model. The generalized log-linear model is a simple logarithmic model which follows

$$\xi = \alpha \cdot \exp(\beta \cdot T + \gamma \cdot RH + \eta \cdot SO_2 + \lambda \cdot Cl) \tag{19}$$

When it is necessary to consider the interactive effect between different environmental factors, an interaction term can be introduced into the generalized log-linear model as a new variable. For example, Ouache and Kabir (2016) constructed a generalized log-linear model including temperature, relative humidity, wind speed, and their interaction terms as variables to evaluate the structural failure probability.

5.2.3 Response surface model

Actually, the influence of environmental factors on the corrosion process is very complicated considering the interaction between different environmental factors and the nonlinearity of the corrosion effects. In view of this, the commonly used power function and general linear models are no longer representative for the actual corrosion effect. The response surface function method can be used to solve such problem which is good at dealing with the nonlinear and interactive effects.

The response surface methodology is commonly used to establish the mathematical model between the response and the influential factors of an unknown system or process. It is an effective method in structural reliability analysis under the condition of implicit limit state functions (Gomes and Awruch 2004). The essence of the response surface method is to approximate the relation between the response and the influential factor to first-order or second-order model as the real function relationship of the complex system (Khuri and Mukhopadhyay 2010). It is especially useful when the system structure is complex with multiple influential factors and mechanisms and is difficult to obtain the explicit expression.

The basic response surface model (RSM) is to use the first-order or quadratic polynomial to approximate the real implicit limit state function. Some researchers also developed higher order RSM (Gavin and Yau 2008). But in most engineering applications, the quadratic model is the most widely used because of the following advantages: (i) the quadratic model can approximate multiple function forms with high accuracy; (ii) the parameters can be estimated by simple least squares method; (iii) there are a large number of successful applications of quadratic models to solve practical problems.

Let the influential variables to be $X = [x_1, x_2, \cdots, x_k]$, and the response variable to be y, then the general form of the quadratic response surface model is

$$y = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \sum_{j=1, i\neq j}^{k} \beta_{ij} x_i x_j + \sum_{i=1}^{k} \beta_{ii} x_i^2 + \varepsilon \tag{20}$$

where ε is the random item which represents the unexplained variance caused by various factors.

Take the atmospheric environmental influence factor $X = [TOW, T, RH, \cdots, SO_2, Cl]$ as the influential variables and the corrosion amount $y = C$ as the response variable, the RSM can be established. By applying the RSM, the interactive effect and nonlinearity of the atmospheric corrosion process can be better captured than conventional dose-response function models (Cai et al. 2018b). But there are only limited applications of RSM to corrosion problems and further explorations are needed.

5.2.4 Statistical learning model

As the great uncertainty in corrosion data and the limitations in dealing with nonlinear interactive effects of conventional regression method, the statistical learning methods are more widely employed in corrosion researches. As an effective tool to solve complex nonlinear problems, the artificial neural networks (ANN) have been used by many researchers in the study of atmospheric corrosion and obtained good prediction results (Diaz and Lopez 2007; Kenny et al. 2009; Vera et al. 2017; Wang et al. 2006). For example, Pintos et al. (2000) built an ANN model for the MICAT atmospheric exposure corrosion test data. The input variables are temperature, humidity, wetting time, dust reduction, sulfur dioxide concentration, and chloride deposition rate. The output variable is corrosion weight loss. The prediction results shows that the model has high goodness of fit and the residual is small in a reasonable distribution range (Diaz and Lopez 2007; Pintos et al. 2000). The ANN model can be applied to different environments as long as the model is properly adjusted (Diaz and Lopez 2007). Cai et al. (1999) constructed a 5-8-1 neural network. The input environmental variables are temperature, relative humidity, corrosion time, sulfur dioxide concentration, and chloride concentration. The output variable was the corrosion depth. The results showed that the neural network is possible to consider more than 70% of the changes in atmospheric corrosion. Sensitivity analysis shows that relative humidity and sulfur dioxide concentration are linearly related to atmospheric corrosion of carbon steel. For zinc, the corrosion rate decreases with increasing relative humidity and increases when sulfur dioxide concentration increases.

The advantage of the ANN method is the ability of dealing with interactive and nonlinear effects. This makes it to be widely used in the study of marine corrosion (Cai et al. 2020a,b), soil corrosion (Ding et al. 2019), corrosion fatigue (Cheng et al. 1999; Haque and Sudhakar 2001), pitting (Cavanaugh et al. 2010) and other processes which have multiple environmental influential factors with complex mechanisms. However, it is also worth to be noticed that the high accuracy of the neural network is based on the adequate training samples and correct adjustment of the network. In most cases, corrosion data is far from enough due to the high consumption of test time and expense. That is the reason why it is mostly used for some of the international corrosion test programs. In addition, as no explicit expression is obtained, the trained neural network is not recommended to be extrapolated to the environments out of the ranges covered by the training samples.

The support vector regression (SVR) method is another powerful tool in dealing with nonlinear interactive effects which is based on structural risk minimization principle. The basic idea of the SVR method is to find a set of optimal parameters which best fit the samples after being mapped into a higher dimension space from the input space via a mapping function. Wen et al. (2009) predicted the corrosion rate of 3C steel under different seawater environment with SVR models and found the prediction error is smaller than back-propagation neural networks (BPNN) models for the identical training and test dataset, and the generalization ability of SVR model is also superior to that of BPNN model. Applications of SVR in predicting the pitting corrosion behavior of stainless steel (Jimenez-Come et al. 2015, 2019) also obtained high prediction accuracy and it is useful to determine the critical factors that influence the corrosion process via sensitivity analysis (Jimenez-Come et al. 2019). Fang et al. (2008) hybrid genetic algorithms and SVR to forecast atmospheric corrosion of zinc and steel which provides better prediction capability than ANN models.

6 Discussion

6.1 Summary of the current literature

There have been numerous researches in exploring the mechanism of the atmospheric corrosion process. There have been comprehensive reviews (Roberge et al. 2002; Simillion et al. 2014) of the experimental techniques, corrosivity factors, computational aspects, as well as corrosion models from different viewpoints. From a mathematical viewpoint, the prediction of atmospheric corrosion should at least include the following three parts of work: (i) Build the atmospheric environment model which directly characterizes the main influential environmental factors in good accuracy; (ii) Establish the corrosion effect model which links the environmental factors with corrosion parameters; (iii) Obtain the atmospheric corrosion kinetic model which describes the development of corrosion over time. Based on the above three aspects, the environment model and the corrosion kinetic model are linked by the corrosion effect model, and finally the corrosion prediction model under different environmental conditions can be built. However, in the current researches, there are still the following aspects need to be further studied:

(1) The atmospheric environment model

For the quantitative description of the atmospheric environmental factors, the mean value method, the distribution method, and the time-varying function method have their own advantages and limitations. The mean value method is simple and convenient for calculation. It can be applied to the cases where the environmental factors are relatively stable and the corrosion effects can be approximated to a linear relationship. However, in the actual environment, the factors are dynamic and dispersive. It is obvious that only one parameter (the mean value) cannot fully include all the information of the environment and cannot reflect the nonlinearity of the corrosion effects. The distribution method describes the environmental factors with the probability density function (PDF) of statistical distributions, which contains more information of the environment. It can reflect the dispersion of environmental factors and the nonlinear corrosion effects. This makes it suitable for situations where the distribution parameters rarely change over a long period of time. However, the distribution method is no longer appropriate when the environmental factors change continuously and the distribution parameters change accordingly. The time-varying function method directly correlates environmental factors with time. It can include almost all the information of the environmental factors and is the most representative of the actual situation. But it needs to be improved to deal with the dependency between different environmental factors.

With respect to the spatial difference of the environmental factors, the cluster analysis method is not accurate due to the discretization of continuously distributed variables. The interpolation method is effective to give accurate calculations of the environmental factors based on the observation of neighboring samples. When the observed samples size is small, the interpolation results may be questionable. In view of this, a physics-based model is useful to predict the target variables with the knowledge of the controlling factors (for example, aerosol production and transportation as a function of wind velocity, distance from sea, relative humidity, terrain factor, and so on (Cole et al. 2003b,c, 2004a). Furthermore, a geographic information system (GIS) is powerful to map the distribution of the environmental factors, which can be a tool for the visualization of environment corrosivity as well as material corrosion resistance.

(2) The atmospheric corrosion effect model

To build the comprehensive corrosion effect model incorporating multiple environmental factors, one should first identify the main influential factors and describe them with proper parameters. Then select a rational form of the corrosion effect model and finally estimate the model parameters based on experimental data. However, in most cases, it is not practical to calibrate the model accurately due to the following reasons.

First, the influential factors for different metals may be different and complex interactions between these factors may possibly exist. For example, the interaction between nitrogen dioxide and sulfur dioxide depends on various factors (Castano et al. 2007), including relative humidity, metal types, and sulfur dioxide concentration. Therefore, it is necessary to determine the types of major environmental impact factors for different materials under different environmental conditions.

Second, there is still some limitations in the selection of environmental variables in most existing corrosion models. For example, time-of-wetting (*TOW*) is the commonly chosen variable to capture the combined effect of temperature and relative humidity on the corrosion process. However, *TOW* is a function of temperature and relative humidity and believed should be replaced by *T* and *RH* (Cai et al. 2018a; ISO 2012a), while there is also researches indicating that absolute relative (AH) maybe a better indicator to capture the combined effect of *T* and *RH* (Boswell-Koller and Rodriguez-Santiago 2019). In addition, the environment is usually parameterized with the annual average value. However, the actual environmental factors are continuously changing and are influenced by various random factors. In the cases where the corrosion effects are nonlinear, the employment of the average value will lead to the loss of important information of the environment and will cause the inaccuracy of the corrosion prediction results.

Moreover, the existing corrosion effect models are mostly linear or generalized linear relations, which cannot describe the complex comprehensive nonlinear effects of the actual environment. The prediction performance of the corrosion models is also limited due to the scatter of the environmental data and the corrosion data.

(3) The atmospheric corrosion kinetic model

In the corrosion kinetic model $C = C_1 t^n$, the exponent n can be used to characterize the protection ability of the corrosion product and acts as the key parameter of the long-term corrosion kinetics. The value of n is affected by various environmental factors in different atmospheric types (rural, urban, industrial, and marine) (Fuente et al. 2011). However, there is only limited works which have studied the relationship between n and the environmental factors and need to be further improvement (Cai et al. 2019; Dean and Reiser 2002; Panchenko and Marshakov 2016; Panchenko et al. 2014). The results show that the effects of the environmental factors are not or just partly statistically significant, which doesn't agree with the actual situation that the environment will inevitably lead to the difference of the composition and protection ability of the corrosion products. Two reasons are contributors to the unsatisfactory of the current model, that is, limited data samples, and improper analysis method.

In respect of the data, the environmental data and the corrosion data usually distributed in a relatively narrow range. Taking the ISO CORRAG data as an example, the corrosivity categories are mostly the C3 and C4 levels, while the low corrosivity levels C1 and C2 and the high corrosivity levels C5 and CX are rarely covered. Other corrosion test programs data may possibly cover the high and low corrosivity levels, but these data are from different test procedures and are difficult to be analyzed due to the poor uniformity. In view of this, the corrosion models cannot be accurately calibrated and the predictive ability may be questionable.

In respect of the method, the conventional regression analysis method has its' own limitation in dealing with the corrosion data. For instance, component n is commonly obtained by regression analysis from the field exposure corrosion test data. However, in most cases the test time is generally within 8 years, and the uncertainty of corrosion data is huge. As a result, the accuracy of the estimated n is unpredictable. Meanwhile, the value of n is within a very narrow range of [0,1], a small deviation of n can result in a large relative error. In addition, the irrational selection of environmental variables and the simplification of environmental data can only support to build an empirical rather than a mechanism model between corrosion parameters and environmental variables.

6.2 Future research direction

(1) The environmental models should include more information. As the field environment changes continuously with periodic and random components, new environmental models should take full consideration of the variability, dependency, and randomness of the field environment. Furtherly, the spatial differences of the field environment should be considered, especially when the materials are suffered to different environmental conditions at different locations. The geographic information system (GIS) is a promising tool to represent the environment spatial difference.

(2) The corrosion effect models should consider the combined effect of multiple environmental factors on the corrosion process with interactions. There are probably two ways for the construction of the quantitative corrosion effect models. First, the models can be built with statistical learning method (ANN and SVR) technique and the response surface (RS) methodology, taking advantage of their ability in dealing with nonlinearity and interactive effects. On the other hand, the models can be built from laboratory accelerated corrosion tests data with the help of design-of-experiment (DOE) methodology and cross validated with outdoor observations.

(3) The corrosion kinetic models should be correlated with environmental factors to extrapolate the kinetic models to environment different from where the models are calibrated. As the conventional field corrosion tests are time and expense consuming, new experimental techniques are needed to produce enough data for the adjustment of the correlation functions. In view of this, online environment and corrosion data measured by corrosion monitoring sensors are promising alternative to gain an insight into the real-time relationship between the corrosion process and the environmental conditions.

(4) The corrosion prediction methods can be applied to various practical engineering problems, such as anticorrosion design, corrosion prevention and management. In extreme and indoor environments, the main influential factors may change and the models should be modified. The influence of climate change is also worthy of more research interest as it is able to influence the corrosion mechanism as well as the corrosion rate in various ways.

7 Conclusions

This paper gives a review of atmospheric corrosion prediction methods from a mathematical viewpoint. First, field and laboratory corrosion tests which provide the data for fitting the corrosion models are discussed. Then, corrosion influential factors and their effects are elaborated. Environmental models give a description of the corrosive environment and the corrosion kinetic models describe the corrosion development over time, while the corrosion effect models link the corrosion parameters with the environment variables.

Field exposure corrosion tests have produced large amount of environmental and corrosion data. They are useful for the construction of the corrosion map of different atmospheric environments around the world. Laboratory corrosion tests simulate or accelerate the actual corrosion process and the results can help to distinguish the influential factors and build the corrosion effect models. However, the limitations also call for continuous improvement of the test procedures and new test techniques. Among all the environmental factors, relative humidity, temperature, and pollutants are the main impact factors in most cases. The effects of each factor on the corrosion process are nonlinear in different degrees and interactive effects exist between different factors.

In most previous researches, the environment is assumed to be steady-state and use the annual average value to represent the actual environmental conditions. As the environment changes over time and varies at different locations, temporal and spatial models should be introduced to give an accurate and thorough description of the environment. The power function model, the general linear model, and the power-linear model are suitable to describe the corrosion kinetics. Further exploration is needed to extrapolate the models to environments different from where the models are calibrated. The dose-response functions are commonly used to correlate the environment variables with the corrosion parameters. More complex models should take the nonlinear and interactive corrosion effect and the dynamic environment into consideration. This means new models and methods should be developed. The response surface method and the statistical learning method are possibly promising tools to deal with this situation.

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