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## Copper Atmospheric Corrosion Mechanism: A Review

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### ABSTRACT

This study examines the impact of multiple factors on the atmospheric corrosion of copper, including the effects of individual gases namely (SO<sub>2</sub>, NO<sub>x</sub>, and O<sub>3</sub>) and the synergistic interactions of mixed gases on copper's atmospheric corrosion. The synergistic effect of SO<sub>2</sub> and NO<sub>2</sub> on copper corrosion appears exclusively at an elevated humidity level of 90%. The combined action of SO<sub>2</sub> and O<sub>3</sub> is more potent than that of SO<sub>2</sub> and NO<sub>2</sub>, potentially elucidating the elevated corrosion rate of copper in O<sub>3</sub>-abundant rural settings. The impact of ammonium sulfate particles is notably highlighted, as the corrosion rate and mechanism of copper undergo major alterations at the critical relative humidity (CRH) of ammonium sulfate. Various copper corrosion products and their creation methods are also presented. A concise review of prevalent experimental and characterization approaches for investigating copper atmospheric corrosion is presented, and research trends in copper atmospheric corrosion are outlined.

### 1. Introduction

Copper is a nonferrous metal closely associated with humankind. It is widely used in electrical engineering, light industry, machinery manufacturing, construction, and defense, with the electrical and electronics industries being the most extremely used. Furthermore, copper and its alloys are also widely used in numerous works of art. When copper is exposed to the atmosphere, a thin green-brown or bluish-green corrosion layer typically forms on its surface, known as verdigris. Maintaining the original surface characteristics of copper and its alloys is crucial in many applications. For example, in electrical connections and conduction applications, the presence of verdigris is undesirable, as even the slightest corrosion can lead to failure of electronic components. This demonstrates that research on the corrosion behavior of

copper in atmospheric environments is crucial for various industrial sectors, especially in the electronics industry [1-4].

The electronics industry has significant practical significance. Copper is extensively used in architecture and sculpture because the patina it forms on its surface is considered a beautiful decorative feature. Once formed, the patina tends to stabilize and become a permanent part of the building or object it is attached to. Under certain conditions, any significant change in the patina is considered as a detrimental agent. Therefore, studying atmospheric corrosion of copper is of great significance for the preservation and restoration of copper artworks [2, 3].

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## 2. Effect of various factors on atmospheric corrosion of copper

Among the many factors affecting copper corrosion, atmospheric pollutants are a very important factor. Atmospheric pollutants are mainly transferred to the metal surface through two ways: dry and wet deposition. Sulfates, nitrates, nitrites, chlorides, carbonates, hydrogen ions, ammonium, metal ions, atmospheric particles, and organic compounds are often found in the corrosion layer or electrolyte components, and they have a certain influence on the corrosion process. These substances act directly through the deposition process, or through the reaction of atmospheric components and deposits in the aqueous phase [4,5].

### Effects of Single Polluting Gases

The main polluting gas in the atmosphere is  $\text{SO}_2$ , and its accelerated corrosion effect on metals is well known. The formation of verdigris occurs as  $\text{SO}_2$  in the water film adsorbed on the metal surface decomposes. Some of the chemical substances obtained from the decomposition combine with copper ions (copper ions are produced by the chemical decomposition of copper oxides and the action of corrosion cells). A large number of reactions are involved during this process [5, 6]. When copper is exposed to an atmosphere containing  $\text{SO}_2$  for a short period of time, the corrosion product film formed on its surface is mainly composed of sulfate, which is the basic copper sulfate  $\text{Cu}_4(\text{SO}_4)(\text{OH})_6$ , just like when it is exposed to an industrial atmosphere for a long time [10]. The corrosion reaction process act as following: the acidic medium environment formed causes the  $\text{Cu}_2\text{O}$  in the local area with poor stability to be destroyed, and the surface oxide film destroyed becomes the anode (area) of the corrosion micro battery, copper corrodes and dissolves, and further forms  $\text{Cu}_4\text{SO}_4(\text{OH})_6$  with lower solubility.  $\text{NO}_x$  is another major pollutant in industrial air. Nitrogen pollutants are emitted into the atmosphere in the form of  $\text{NO}$ . During atmospheric transport,  $\text{NO}$  is easily oxidized to  $\text{NO}_2$ .  $\text{NO}_2$  can then be further oxidized to  $\text{HNO}_3$ . However, under conditions of low  $\text{NO}_x$  concentrations ( $<0.1 \times 10^{-6}$ ), the oxidation reaction rate is very slow. Therefore, under normal circumstances, the content of  $\text{HNO}_3$  and  $\text{NO}^{-3}$  is very low [7, 8]. Some people estimate that the minimum  $\text{NO}_2$  content that can cause metal corrosion is  $30\mu\text{g}/\text{m}^3$  [13]. Compared with  $\text{NO}_2$ ,  $\text{SO}_2$  can promote the formation of a more continuous and more protective surface layer on copper. Laboratory results of copper exposed to  $\text{NO}_2$  show that the  $\text{Cu}_2\text{O}$  in the surface of the verdigris exposed to  $\text{NO}_2$  tends to be replaced by basic copper nitrate, and the color gradually becomes darker [14].  $\text{O}_3$  is a strong oxidant, and its redox potential is second only to fluorine. Ozone has high energy, so it is very unstable. At room temperature and pressure, its molecular structure is easy

to change, and it quickly decomposes into oxygen ( $\text{O}_2$ ) and single oxygen atoms ( $\text{O}$ ), the latter of which has a very strong oxidizing effect. Atmospheric pollutants  $\text{SO}_2$ ,  $\text{NO}_2$ , and  $\text{O}_3$  all have a significant impact on atmospheric corrosion. The effect of  $\text{O}_3$  on copper corrosion is stronger than that of  $\text{SO}_2$  and  $\text{NO}_2$  [13,14].

### Synergistic effect of mixed gases

In copper corrosion, the synergistic effect of  $\text{SO}_2$ ,  $\text{NO}_2$  occurs only at a relatively high humidity (90%) [8]. When copper is exposed to  $\text{SO}_2 + \text{NO}_2$  atmosphere, sulfate or sulfite first appears on the surface and then transforms into nitrate after a few days [17]. Similar experiments also obtained the same results [11]: under RH 80%  $\text{SO}_2 + \text{NO}_2$  mixed atmosphere conditions, the growth of copper sulfate is quickly inhibited and copper nitrate becomes the main product. In the high  $\text{NO}_2 +$  high  $\text{SO}_2$  atmosphere, the change of relative humidity and  $\text{NO}_2$  content has a significant effect on the weight gain of copper. Some analysis results show that the corrosion products obtained are copper sulfate, sulfite, nitrate and nitrite, mainly sulfate and nitrate [18, 19]. For copper corrosion, the synergistic effect of  $\text{SO}_2$ ,  $\text{O}_3$  is stronger than that of  $\text{SO}_2$ ,  $\text{NO}_2$ , which can explain why copper has a high corrosion rate in rural environments rich in  $\text{O}_3$  [13, 14].

In moist air containing  $\text{SO}_2$ ,  $\text{Cu}_2\text{O}$  is formed on the surface. A thin layer of sulfite similar to  $\text{CuSO}_3 \cdot x\text{H}_2\text{O}$  is covered on the surface. The introduction of  $\text{NO}_2$  causes  $\text{CuSO}_3 \cdot x\text{H}_2\text{O}$  to oxidize to  $\text{CuSO}_4 \cdot x\text{H}_2\text{O}$ , but no new  $\text{Cu}_2\text{O}$  is formed. The sulfite is gradually replaced by nitrate. After the introduction of  $\text{O}_3$ , the formation rate of  $\text{Cu}_2\text{O}$  and  $\text{CuSO}_4 \cdot x\text{H}_2\text{O}$  increases, resulting in a rapid increase in corrosion products [5].

### Effects of Corrosion Particles

Particles in the atmosphere can accelerate metal corrosion in many ways, such as increasing surface conductivity after dissociating soluble ions from the particles [21]. Ammonium and sulfate ions are the most abundant of the main dust particles in urban environments and may play a dominant role in the corrosion process [4, 16]. The corrosion of copper in air containing ammonium sulfate particles is largely affected by the relative humidity (RH). The corrosion rate and mechanism of copper change significantly at the critical relative humidity (CRH) of ammonium sulfate. Studies have found [23] that copper reacts with ammonium sulfate as long as the CRH of ammonium sulfate (the CRH of ammonium sulfate corresponds to the equilibrium partial pressure of water vapor above a saturated ammonium sulfate solution, which is 75% at  $100^\circ\text{C}$  and 81% at  $27^\circ\text{C}$ ) is exceeded (the CRH is the humidity at which the salt begins to absorb a large amount of water). Above the CRH of ammonium sulfate, the salt continues to absorb water even at concentrations

exceeding saturation, forming a sufficient solution to spread across the copper surface, causing corrosion over a larger area. The copper is severely corroded, forming a thick  $\text{Cu}_2\text{O}$  layer, which is then covered by basic copper sulfate. At CRH or above,  $\text{Cu}_2\text{O}$  is the first solid corrosion product formed (it is the primary product in the initial stages of atmospheric corrosion of copper in most natural environments). Within a given time, the amount of  $\text{Cu}_2\text{O}$  increases with increasing RH, temperature, and the amount of ammonium sulfate. At temperatures between  $27\text{ }^\circ\text{C}$  and  $100\text{ }^\circ\text{C}$  and humidities above CRH,  $\text{Cu}_3\text{SO}_4(\text{OH})_4$  and  $\text{Cu}_4\text{SO}_4(\text{OH})_6$  are the primary products of  $\text{Cu}_2\text{O}$  formation in natural environments. At temperatures above  $27\text{ }^\circ\text{C}$  and humidities above CRH, the primary corrosion product of copper, in addition to the aforementioned two, is  $\text{Cu}_4\text{SO}_4(\text{OH})_6 \cdot \text{H}_2\text{O}$ . study [12] studied the effect of the number of dust particles deposited on the copper surface on the atmospheric corrosion mechanism. When the concentration of ammonium sulfate particles on the copper surface ranges from  $1\text{ }\mu\text{g}/\text{cm}^2$  to  $10\text{ mg}/\text{cm}^2$ , the corrosion products are the same and the formation sequence is also the same: first, water adsorption and ammonium sulfate particles dissolve, then Cu dissolves in the form of  $\text{Cu}(\text{NH}_3)_2^+$ , then  $\text{NH}_3$  evaporates, basic copper sulfate precipitates, and then the equilibrium between copper and basic copper sulfate leads to the formation of the  $\text{Cu}_2\text{O}$  inner layer. Using the surface sensitive method, it can be seen that the first layer formed in the ammonium sulfate deposition area at  $25\text{ }^\circ\text{C}$  is the most important layer.

The solid corrosion product is the basic copper sulfate rather than  $\text{Cu}_2\text{O}$ . The main effect of increasing the number of particles deposited is the increase in the number of corrosion products, and the mutual equilibrium effect causes the basic copper sulfate on the surface of the few particles to be completely decomposed. On samples with more particles deposited, the reaction stops when the space between the metal and the basic copper sulfate is filled, and a stable basic copper sulfate is left on the  $\text{Cu}_2\text{O}$  layer. The experiments conducted [17, 21] found that the chloride deposition rate is an important factor in the copper corrosion process, even in areas far from the coastline. Chloride mainly exists in the form of particles (such as sea salt), which then dissolves into the electrolyte [25]. The interaction between the deposition rate of chloride in the outdoor atmosphere and the rainfall time causes changes in copper corrosion, while the interaction between the deposition rate of chloride in the indoor atmosphere and the wetting time at different temperatures, as well as the interaction between the wetting time at different temperatures and sulfur-containing compounds, can also cause changes in copper corrosion [26]. In addition to the above factors, seasonal changes also affect copper corrosion, and the patina will change. In the study [27], using AES technology, it was found that in the samples

exposed in summer, sulfides were present in the surface layer of the corroded material, while chlorides were distributed deeper. It was also observed that the distribution of chlorides in the samples exposed in summer and winter was different. The low diffusion rate at low temperatures in winter or the protective nature of the oxide film formed on the copper surface can explain the above results; the oxide film formed on the samples exposed in summer was thicker than that formed in winter. These results agree well with the surface observations and X-ray diffraction (XRD) results. This result can be explained by the differences in temperature, relative humidity, and the amount of sulfide and chloride on the copper surface.

### 3. Corrosion products of copper

Verdigris has a complex chemical composition, some of which are not protective, and some are only occasionally observed in verdigris.

#### Oxides

The most common corrosion product in verdigris is  $\text{Cu}_2\text{O}$ , and  $\text{Cu}_2\text{O}$  is also the first product formed when copper is exposed to the atmosphere.  $\text{Cu}_2\text{O}$  has a highly symmetrical cubic structure, with two oxygen atoms surrounding each metal atom, and each oxygen atom is surrounded by a tetrahedron of copper atoms. Moreover,  $\text{Cu}_2\text{O}$  is insoluble in water and slightly soluble in acid [28]. The  $\text{Cu}_2\text{O}$  layer has protective properties, and the protective properties of  $\text{Cu}_2\text{O}$  will gradually weaken due to the deposition of corrosive gases and atmospheric particles. In particular, the acidification caused by the alternating wet and dry deposition of acidic substances such as  $\text{SO}_2$  in the electrolyte on the copper surface will cause the  $\text{Cu}_2\text{O}$  to the protective property is weakened, thus accelerating the corrosion of copper [29].  $\text{Cu}_2\text{O}$  will be converted into other substances under certain conditions. In the study [6], pretreated  $\text{Cu}_2\text{O}$  reacts with NaCl in humid air to form basic copper chloride. At the same time, the pH value increases due to the oxidation of  $\text{Cu}_2\text{O}$ . When  $\text{O}_3$  participates in the reaction, CuO is also generated. When  $\text{SO}_2$  participates in the reaction, the surface alkalinity increases, causing the sulfidation rate to increase.  $\text{Cu}_2\text{O}$  fully absorbs  $\text{SO}_2$  to form basic copper chloride and basic copper sulfate. CuO is a corrosion product that can occasionally be observed in verdigris [30]. CuO can quickly react with  $\text{SO}_2$  in humid air to form  $\text{Cu}_4(\text{SO}_4)(\text{OH})_6$  and  $\text{Cu}_2(\text{SO}_4)(\text{OH})_3 \cdot 2\text{H}_2\text{O}$  [31]. This may explain why CuO rarely appears in outdoor environmental exposure.  $\text{Cu}_3\text{O}_2$  is a rarely reported phase among copper corrosion products. In an experiment conducted [32], in which copper was exposed to a mixture of  $\text{H}_2\text{S}$ ,  $\text{Cl}_2$ , and  $\text{NO}_2$ , information about  $\text{Cu}_3\text{O}_2$  was obtained through three different methods. Its presence has a great effect on the corrosion

protection of copper. However, more extensive research is needed to obtain more definitive conclusions about its origin and role.

#### **Chlorides**

In marine, urban, or industrial areas with high concentrations of water-soluble chlorides, copper ions are generated by the dissolution of  $\text{Cu}_2\text{O}$ , which reacts with chloride ions to form  $\text{CuCl}$ . Once formed,  $\text{CuCl}$  can serve as seed crystals to form  $\text{Cu}_2\text{Cl}(\text{OH})_3$  through many subsequent dissolution, ion pairing, and redeposition steps[33].

#### **Others**

Several other known copper minerals,  $\text{Cu}_2\text{NO}_3(\text{OH})_3$  and  $\text{Cu}_2\text{CO}_3(\text{OH})_2$ , are only occasionally observed in the verdigris component.  $\text{Cu}_2\text{NO}_3(\text{OH})_3$  is occasionally detected on electroluminescent copper surfaces, and  $\text{Cu}_2\text{CO}_3(\text{OH})_2$  is sometimes found as a verdigris component of ancient communion plates. As a more general result of corrosion, these products reflect chemical reactions between the metal and the environment to which it is exposed [26].

### **4. Research methods of atmospheric corrosion of copper**

#### **Atmospheric exposure corrosion test under natural conditions**

The so-called atmospheric exposure test is to place the sample on a test rack in an outdoor exposure field and conduct a corrosion test in a natural atmospheric environment. During the exposure process, the sample is observed regularly and its corrosion rate is measured by weight loss method. At the same time, various material characterization techniques are used to study its corrosion products and corrosion morphology. Atmospheric exposure test is a relatively reliable corrosion test method close to the use environment, and the results obtained are also close to the actual use situation. Its results are usually used as an important indicator to evaluate the quality of metal corrosion resistance. Twelve European countries, the United States and Canada cooperated in research to quantify the effects of acidic pollutants on different materials. Copper, as a structural metal and electrical contact metal, was included in the research scope[34].

#### **Laboratory simulation accelerated test**

Although atmospheric exposure test can obtain valuable data such as metal corrosion rate under reliable natural conditions, the long experimental period, especially for corrosion-resistant materials and protective layers, which may take several years or longer, and the strong regionality, is not conducive to the promotion and

application of test results. For decades, the engineering design and corrosion protection fields have been seeking to use the results of short-term accelerated corrosion tests in the laboratory to infer the results of long-term outdoor exposure tests, and then predict the atmospheric corrosion life of materials, products and protective layers, in order to partially replace or supplement atmospheric corrosion exposure tests. As a result, many accelerated test methods have emerged. Common atmospheric corrosion simulation accelerated tests include salt spray test, wet heat test, dry-wet cycle test and factor composite test. Study [17] used intermittent salt-water spray composite corrosion test method to explore the effect of  $\text{HSO}_3^-$  and/or  $\text{Cl}^-$  on copper corrosion, and to seek accelerated corrosion test methods suitable for the polluted atmospheric environment in this studies have shown that the corrosion effect of  $\text{NaHSO}_3$  on copper is greater than that of  $\text{NaCl}$ , and the synergistic corrosion effect of mixed media on copper is not significant.

#### **Application of Characterization Methods in Atmospheric Corrosion**

In corrosion research, characterizing corrosion products using various research methods is extremely important. The following describes the application of several of these techniques in research. Scanning electron microscopy (SEM) and atomic force microscopy (AFM) can both obtain microscopic corrosion morphology images. SEM is a relatively common characterization method. Although the operation of AFM is more complicated, it is not affected by the conductive properties of the material. It also has atomic-level spatial resolution and can obtain more information such as the average roughness of the sample surface and intermolecular forces [35]. X-ray photoelectron spectroscopy is usually used to obtain spectra from solid surfaces, but it has also been used on liquid surfaces after specific treatments. Similar treatments can be performed during in-situ testing during sustained atmospheric corrosion. Studies [12, 29] used XPS to study the effects of  $\text{SO}_2$  and  $\text{NO}_2$  on copper corrosion at relative humidity of 50% and 70%. Their study showed that a decrease in relative humidity (from 70% to 50%) had no significant effect on the formation rate and surface composition of the patina; in  $\text{SO}_2$ , the initially generated product  $\text{Cu}_2\text{O}$  was gradually replaced by  $\text{CuO}$  and basic sulfate, while in  $\text{NO}_2$ , it was replaced by basic nitrate[36].

### **5. Conclusion**

The behaviour of copper corrosion in the atmosphere under the impact of many environmental conditions, such as humidity, particulate matter, and single and mixed polluting gases, was thoroughly examined in this paper. It is clear that the corrosion of copper is a multi-phase, intricate process that is influenced by a wide range

of interrelated factors, including temperature, relative humidity, and gas composition. Because it may produce acidic surface coatings and basic copper sulphate, SO<sub>2</sub> continues to be the most significant individual gas in starting corrosion. But synergistic effects, especially between SO<sub>2</sub> and O<sub>3</sub>, intensify corrosion rates even more, underscoring copper's particular susceptibility in ozone-rich rural or urban transition zones. Additionally, the study demonstrates how copper corrosion entails intricate transitions between Cu<sup>+</sup> and Cu<sup>2+</sup> species, resulting in the development of many oxide, sulphate, and chloride phases, the stability of which is highly environment-dependent. Verdigris formation, although occasionally protective, frequently develops into porous or unstable layers when exposed to pollutants or fluctuating humidity, according to existing research, which limits its long-term durability in architectural and electrical applications.

There are still large research gaps in spite of tremendous advancements. The behavior of corrosion under the alternating dry-wet cycles that characterize actual air conditions is still poorly understood. To clarify reaction kinetics at the nanoscale, future research should concentrate on in-situ, real-time investigations that combine surface characterization with electrochemical monitoring (e.g., in-situ XPS, Raman spectroscopy, and environmental SEM). Furthermore, to more accurately forecast copper corrosion life, integrated modelling techniques that connect lab data with field exposure are crucial.

In summary, future research on copper atmospheric corrosion should prioritize mechanistic understanding under realistic fluctuating environments, multidisciplinary characterization, and development of sustainable protection strategies to ensure the long-term stability and functionality of copper materials

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