

## Study on the anti-corrosion performance of multi-functional additives used in coolant for solder material

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### ABSTRACT

*This work presented the corrosion protection performance of anticorrosion multi-metal hybrid (AC-2MH) complex additive for solder materials in corrosive media. The AC-2MH complex contained traditional additives and organic acids. The Electrochemical impedance spectroscopy results demonstrated the formation of active and passive protective layers on the surface of the alloys, which prevented the solder materials from corrosion. The electrochemical analysis also indicated that the multifunctional additive had the capability of inhibiting the corrosion rate up to 97-99% under normal conditions (30-40 °C). At high temperatures (60-70 °C), the corrosion inhibition efficiency was significantly reduced to 89-94%.*

**Keywords:** Coolant; Ethylene glycol; Corrosion inhibitors; Electrochemical analysis.

### 1. INTRODUCTION

For heat exchanger systems using ethylene glycol (EG) - water as a cooling solution, the materials used to fabricate the heat exchangers and the arrangement of heat channels determine the heat exchange efficiency [1]. Currently, in addition to materials with low specific heat, light, and facile to processing, such as aluminum, copper, and their alloys, solder materials and steel are also among the materials commonly employed to fabricate the cooling components. In which, some types of alloys are commonly used to make cooling systems such as aluminum alloy (3003), steel (SAE 1010), copper, brass (Cu-70, Zn-30), and solder consumables (Pb-70, Sn-30) [2]. Coolant is often served under high temperature, long time conditions, and in contact with many different metals. This facilitates the decomposition of EG to organic acids that are corrosive to the cooling system [3, 4]. Therefore, the addition of additives to reduce the rate of corrosion of metals, as well as hinder the electrochemical reactions occurring on their surfaces, thereby enhancing the ability to protect against corrosion, against deposit formation for cooling systems which are studied and applied in different ranges [5, 6].

With water-glycol coolant systems, there is no single anti-corrosion additive that will work in all cases, so there is always a combination of different inhibitors to compensate and/or enable a synergy effect for the best corrosion protection. In which there are complex additive systems containing traditional inorganic additives and new additives of carboxylic acids (OAT technology), which is known as a new trend for EG-water cooling systems [7, 8]. The corrosion protecting mechanism and performance of these additive systems for steel, aluminum, copper, and magic alloys have been reported in recent years [9-11]. However, the anti-corrosion mechanism of these additives for solder materials results are still very limited.

Herein, the anti-corrosion performance of multifunctional additive-contained EG-water coolant is investigated for multi-metallic substrates including G10200 steel, A23190

aluminum alloy, and C11000 red copper at different temperature conditions [12]. The corrosion protection efficiency of the complex additive system for solder materials at different temperature conditions is also investigated, and evaluated their surface corrosion protection mechanism through electrochemical impedance measurement.

## **2. EXPERIMENTAL SECTION**

### **2.1. Materials and equipment**

Chemicals: NaCl, NaHCO<sub>3</sub>, Na<sub>2</sub>SO<sub>4</sub>, Na<sub>2</sub>SiO<sub>3</sub>, Na<sub>2</sub>B<sub>4</sub>O<sub>7</sub>·10H<sub>2</sub>O tolyltriazole, octanoic acid, and ethylethylene glycol were purchased from Macklin (China), and Merck (Germany). Deionized water was used for preparation using deionized water with a conductivity of lower than 1 μS/cm.

Preparation of solder materials-contained electrode: SAE30 alloy was coated on a round copper electrode with a diameter of 0.5 cm, followed by the ASTM D1384. Before measuring, the electrode surface was polished using sandpaper with a fineness ranging from 1.200 to 2.000.

Autolab PGSTAT 302N multi-function electrochemical analyzer and Nova 2.1.4 software at Institute of Chemistry and Materials were employed for the electrochemistry analysis.

Auxiliary equipment in the laboratory: an electric stove with a magnetic stirrer and thermostatic tank was used during the experiment process.

### **2.2. Sample preparation**

The AC-2MH additive system was prepared following compositions: Sodiumbenzoate (60.0-65.0%), Sodium nitrite (10.5-12.5%), Tolyltriazole (10.0-12.0%), Sodium nitrate (3.5-4.0%), octaoic acid (3.5-4.5%), sodium metasilicate (1.0-1.5%), sodium borate (1.0-1.5%), in addition, anti-scale and anti-foam additives accounted for from (1.0-1.5%). The cooling solution was fabricated by adding 5% w/w of AC-2MH additive into solution containing 95% of EG and 5% of deionized water is added 5% by weight of the solid additive system AC-2MH to make the research solution system.

Preparation of corrosive solution: Corrosive water is prepared according to ASTM D 1384:148 mg of Na<sub>2</sub>SO<sub>4</sub>, 165 mg of NaCl, and 138 mg of NaHCO<sub>3</sub> were dissolved in deionized water with a final volume of 1.000 mL.

Corrosion measurement electrolyte solution: Corrosion measurement solution consists of additive-contained cooling solution and corrosive water with a ratio of 1:2 by volume.

### **2.3. Electrochemical analysis**

Electrochemical analysis was performed on Autolab PGSTAT 302N device with a 3-electrode system: Ag/AgCl comparison electrode (RE); Pt counter electrode (CE), and the solder material working electrode (WE).

Polarization curve measurement: Polarization curve was studied on Autolab device with Nova 2.1.4 software, the scan range of ± 0.15 V over open circuit potential (OCP), the scan speed of 0.001 V/s. Corrosion inhibitory ( $\eta_p$ , %) was calculated as follows (1):

$$\eta_p = \frac{i_{corr}^0 - i_{corr}}{i_{corr}^0} \times 100 \quad (1)$$

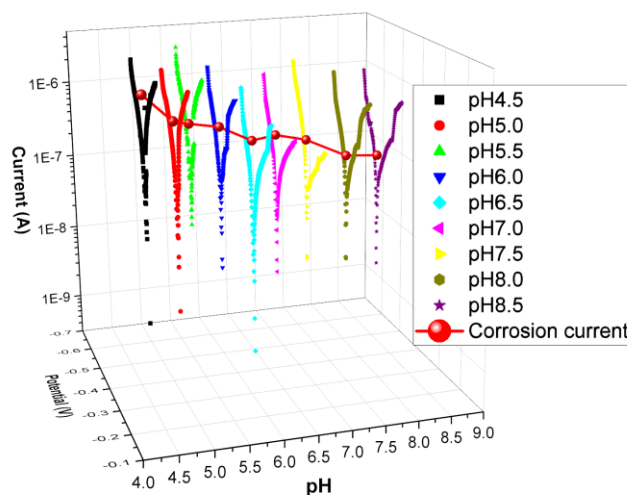
Where,  $i_{corr}^0$  and  $i_{corr}$  are the corrosion current density ( $A/cm^2$ ) in the absence and presence of inhibitors, respectively.

Total impedance measurement was determined on Nova 2.1.4 software with scanning range from  $10^5$ -0.1 Hz, soak time of 20 min before measurement at open circuit potential.

### 3. RESULTS AND DISCUSSIONS

#### 3.1. Effect of pH on anti-corrosion performance of the multifunctional additive

The pH of the corrosion measurement solution containing the 5% AC-2MH additive was adjusted from 4.5 to 8.5 by KOH agents and glycolic acid. The polarization curve measurement results on the solder consumable electrode are shown in figure 1. It can be clearly seen from figure 1 that the shift of the corrosion potential towards the positive side when the pH increases from 4.5 to 8.5, proving the corrosion rate for the solder material linearly decreases with the increase of the pH solution. The corrosion parameters of the system obtained at different pH values were determined by Tafel extrapolation, as presented in table 1. The calculated corrosion rates from the polarization curves indicate that the AC-2MH additive has a remarkable inhibiting effect on the corrosion of solder materials, especially at neutral pH to weak alkaline. The results also show that at pH 8.0 and 8.5, the corrosion rate is almost constant, thus, the solution pH of 8.0 is selected as the optimal condition.



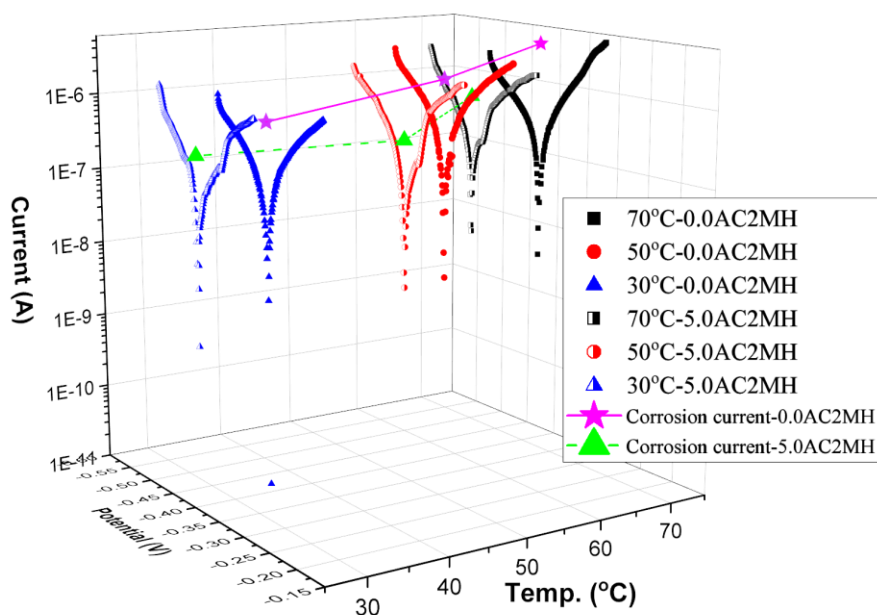
**Figure 1.** Polarization curve of solder consumables at different pH values.

**Table 1.** Corrosion parameters for solder materials at different pH conditions.

| No. | pH  | E <sub>cor</sub> (V) | I <sub>cor</sub> (A)   | Corrosion rate (mm/year) |
|-----|-----|----------------------|------------------------|--------------------------|
| 1   | 4.5 | -0.301               | $2.346 \times 10^{-6}$ | 0.0744                   |
| 2   | 5.0 | -0.339               | $9.197 \times 10^{-7}$ | 0.0292                   |
| 3   | 5.5 | -0.481               | $4.743 \times 10^{-7}$ | 0.0150                   |
| 4   | 6.0 | -0.495               | $3.807 \times 10^{-7}$ | 0.0121                   |
| 5   | 6.5 | -0.489               | $2.372 \times 10^{-7}$ | 0.0075                   |
| 6   | 7.0 | -0.539               | $2.163 \times 10^{-7}$ | 0.0069                   |
| 7   | 7.5 | -0.538               | $1.722 \times 10^{-7}$ | 0.0055                   |
| 8   | 8.0 | -0.480               | $1.218 \times 10^{-7}$ | 0.0039                   |
| 9   | 8.5 | -0.475               | $1.155 \times 10^{-7}$ | 0.0037                   |

### 3.2. Effect of temperature on anti-corrosion performance of the multifunctional additive

To evaluate the effect of temperature on the effectiveness of corrosion inhibition for solder materials, the corrosion experiments were carried out with and without addition of AC-2MH additive at the pH solution of 8 and the temperature range from 30 to 70 °C. This is also known as the operating temperature range of the ethylene glycol/water-based cooling system [8]. The results are shown in figure 2 and table 2.



**Figure 2.** Polarization curves of solder consumables with and without AC-2MH additive at different temperatures.

**Table 2.** Corrosion parameters for solder consumables at different temperatures.

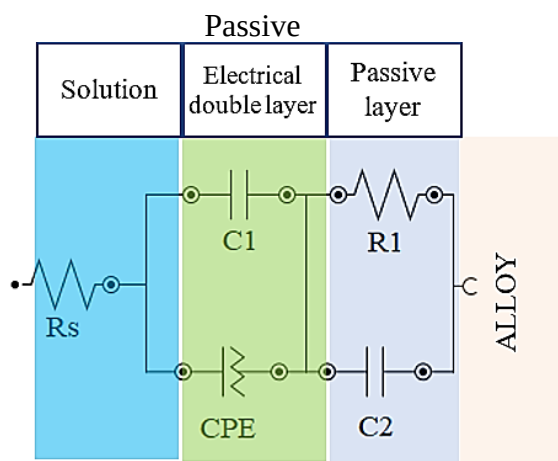
| No. | Temp (°C) | Additive content AC-2MH (%) | Ecor (V) | Icor (A)               | Corrosion rate (mm/year) | Protection effectiveness (%) |
|-----|-----------|-----------------------------|----------|------------------------|--------------------------|------------------------------|
| 1   | 30        | 0.0                         | -0,325   | $1.807 \times 10^{-6}$ | 0.0319                   | -                            |
| 2   |           | 5.0                         | -0,470   | $2.199 \times 10^{-7}$ | 0.0070                   | 93.73                        |
| 3   | 50        | 0.0                         | -0,293   | $2.823 \times 10^{-6}$ | 0.0800                   | -                            |
| 4   |           | 5.0                         | -0,363   | $3.337 \times 10^{-7}$ | 0.0106                   | 88.18                        |
| 5   | 70        | 0.0                         | -0,376   | $4.118 \times 10^{-6}$ | 0.1306                   | -                            |
| 6   |           | 5.0                         | -0,500   | $5.365 \times 10^{-7}$ | 0.0170                   | 86.97                        |

It can be seen from figure 2 and table 2 that the corrosion rate of the solder materials increases significantly with the increase in temperature. Without the additive, the corrosion rate for the solder consumables increases by 4.3 times, while using the additive, the corrosion rate only increases by 2.4 times with the increase of temperature from 30 °C to 70 °C. The results clearly indicate that the increase of corrosion rate along with temperature rising significantly minimized upon the addition of the AC-2MH

additive system with enhanced corrosion protection efficiency of more than 86%. The results of this study also show the similarity with the protective performance of AC-2MH additive system for aluminum, steel, and copper alloys at 70 °C, protection reaches 89.2% to 94.8%, as reported previously [12].

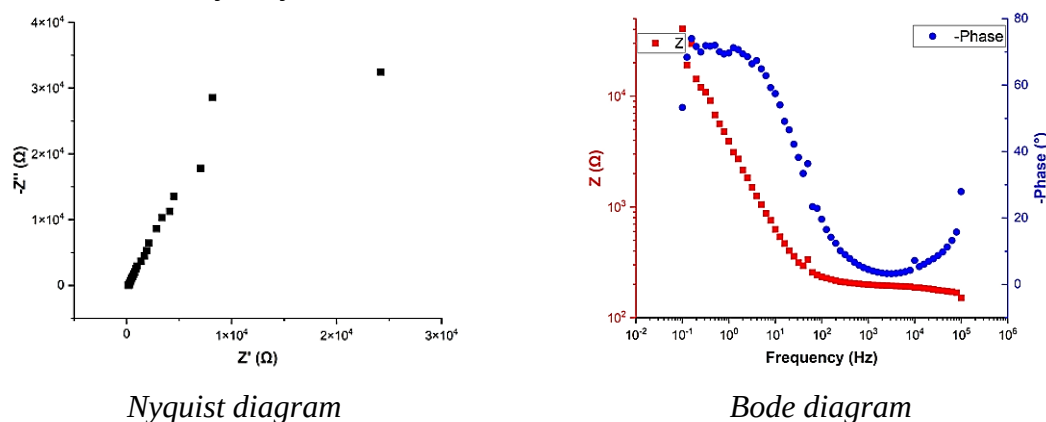
### 3.3. Mechanism for the corrosive protection of solder consumables corrosion by the AC-2MH additive system

The corrosion protecting mechanism of the additive on solder consumables was demonstrated through the Nyquist and Bode diagrams. From the resultant diagram, the evolution of the electrochemical process on the electrode surface of the solder consumables using equivalent circuit electricity was simulated. The equivalent circuit diagram simulating the electrochemical evolution on the electrode surface is shown in figure 3.



**Figure 3.** Equivalent circuit diagram simulating the electrochemical process on the electrode surface of solder consumables.

It can be obvious that the matching circuit is built quite consistent with the results given in the schematic diagram of figure 3. The equivalent circuit diagram is built from the total impedance spectrum in figure 4, showing that the electrode surface could be coated with a thin layer by the additives.



**Figure 4.** Total impedance spectroscopy of the solder consumable sample with AC-2MH additive.

From the impedance results, the corrosion protection mechanism of the AC-2MH additive for the solder consumables electrode could be explained as follows: Firstly, the active parts of the additives were chemically absorbed on the surface of solder material (passive layer), and as a result, formed a passive layer on the metal surface. This coating formation is usually created by inorganic additives such as nitrite, nitrate, silicate, etc. The parameters of resistor R1 and capacitor C2 are typical for this coating and are given in table 3. The second layer is a double charge layer, which is formed by the hydrocarbon tail of the organic additive present in the AC-2MH system. The electric dual class is characterized by the capacitor C1 and the unrealistic constant phase element CPE. The CPE element is built upon the non-ideal capacitor element and is characterized by the inductance value  $Y_0$  and the non-ideal exponent N. For the electric double layer with a higher impedance value, then the less charge transport capacity, the higher the protective effect, the higher the resistance to electrochemical corrosion [13]. With the results shown in table 3, the low C and  $Y_0$  indicate that the protection ability of AC-2MH additive for solder consumables is relatively good. The part of the solution in contact with the outer double charge film has electrochemical properties by the solution resistance  $R_s$ , the calculation results are given in table 3.

**Table 3.** Electrochemical characteristics in equivalent circuit.

| Parameter | $R_s$    | C1                   | Q                    |      | R2       | C2                   |
|-----------|----------|----------------------|----------------------|------|----------|----------------------|
| Unit      | $\Omega$ | F                    | $Y_0, Mho$           | N    | $\Omega$ | F                    |
| Result    | 79.915   | $6.3 \times 10^{-6}$ | $5.7 \times 10^{-5}$ | 0.83 | 112.68   | $1.3 \times 10^{-8}$ |

#### 4. CONCLUSIONS

In summary, the corrosion protection of solder consumables by AC-2MH additive was thoroughly investigated using the electrochemical method. The results show that the additive was of good protection at pH 8. The protection ability decreases along with the increase in temperature. The protection efficiency at 30 °C, 50 °C and 70 °C was determined to be approximately 93.73%, 88.18%, and 86.97%, respectively, corresponding to the corrosion rate of 0.0070 mm/year, 0.0108 mm/year and 0.0170 mm/year, respectively. The results of studying the protection mechanism of the electrode surface of the solder material show that there is a formation of protective layers on the electrode surface, including a passive film, a double charge layer that prevents attack on the electrode surface.

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### TÓM TẮT

#### **Nghiên cứu hiệu quả chống ăn mòn của phụ gia đa chức năng sử dụng trong nước làm mát cho vật liệu hàn**

*Nghiên cứu này trình bày hiệu quả bảo vệ ăn mòn của hệ phụ gia phức AC-2MH cho vật liệu hàn trong môi trường ăn mòn. Hệ phụ gia AC-2MH bao gồm các phụ gia truyền thống và phụ gia axit hữu cơ. Kết quả đo phổ tổng trở EIS chứng minh sự tạo thành các lớp bảo vệ chủ động và thụ động trên bề mặt hợp kim trong môi trường ăn mòn. Phân tích điện hóa cũng cho thấy hiệu quả ức chế ăn mòn của hệ phụ gia nghiên cứu lên đến 97-99% tại điều kiện thường (30-40 °C). Trong khi đó, tại vùng nhiệt độ cao (60-70 °C), hiệu quả ức chế ăn mòn giảm đáng kể xuống còn 89-94%.*

**Từ khóa:** Nước làm mát; Etylen glycol; Ức chế ăn mòn; Phương pháp điện hóa.