ปัจจัยของอุณหภูมิต่อประสิทธิภาพของยูเจนอล สำหรับการยับยั้งการกัดกร่อนของโลหะทองแดง

EFFECT OF TEMPERATURE ON INHIBITION EFFICIENCY OF EUGENOL FOR COPPER CORROSION

อาทิตยา สามณฑา* และณัฐวุฒิ จิตรสมัคร
Atitaya Samontha*, and Natthawut Jitsamak

Faculty of Science and Technology, Bansomdejchaopraya Rajabhat University,
*corresponding author e-mail: rabbitsamontha@gmail.com
(Received: 9 October 2018; Revised: 19 February 2018; Accepted: 4 March 2019)

บทคัดย่อ

ปัจจัยของอุณหภูมิต่อประสิทธิภาพของยูเจนอล สำหรับการยับยั้งการกัดกร่อนของโลหะทองแดง ศึกษาโดยการตรวจวัดน้ำหนักที่หายไปของโลหะทองแดง และเทคนิคกล้องจุลทรรศน์อิเล็กตรอนแบบส่องกราด การศึกษาการกัดกร่อนของโลหะทองแดงได้รับความสนใจเป็นอย่างมาก เพราะโลหะทองแดงถูกนำมาใช้เป็นวัตถุดิบหลักในอุตสาหกรรมต่าง ๆ ซึ่งปัจจุบันวิธีที่นิยมสำหรับการป้องกันหรือการลดการกัดกร่อนของโลหะทองแดง คือการเติมสารยับยั้งการกัดกร่อนที่เป็นสารจากธรรมชาติที่ไม่ส่งผลอันเป็นอันตรายหรือเป็นพิษต่อสิ่งแวดล้อม สำหรับงานวิจัยนี้ยูเจนอลซึ่งสามารถสกัดได้จากพืช ถูกนำมาใช้เป็นสารยับยั้งการกัดกร่อนของโลหะทองแดง จากการศึกษาพบว่ายูเจนอลสามารถลดการกัดกร่อนของโลหะทองแดงที่อยู่ในสภาวะการเป็นกรดด้วย 1 โมลาร์ ของกรดไฮโดรคลอริก อย่างไรก็ตามประสิทธิภาพการยับยั้งการกัดกร่อนของโลหะทองแดงที่อยู่ในสภาวะการเป็นกรดด้วย 1 โมลาร์ ของกรดไฮโดรคลอริกลดลงเมื่ออุณหภูมิของการศึกษาเพิ่มขึ้นจาก 30 ถึง 70 องศาเซลเซียส ทั้งนี้ผลการศึกษาจากการตรวจวัดน้ำหนักที่หายไปของโลหะทองแดง และภาพจากเทคนิคกล้องจุลทรรศน์อิเล็กตรอนแบบส่องกราดมีความสอดคล้องกัน ผลงานวิจัยนี้ถือเป็นการแสดงถึงความสามารถของยูเจนอลในการยับยั้งการกัดกร่อนของโลหะทองแดงภายใต้อุณหภูมิที่แตกต่างกัน อีกทั้งเป็นการแสดงถึงคุณสมบัติอีกอย่างหนึ่งของสมุนไพรไทยซึ่งมียูเจนอลเป็นองค์ประกอบ

คำสำคัญ: อุณหภูมิ ประสิทธิภาพการยับยั้ง ยูเจนอล การกัดกร่อนของทองแดง

Abstract

The effect of temperature on inhibition efficiency of eugenol for copper (Cu) corrosion was studied using the weight loss measurement, and scanning electron microscope (SEM). The corrosion of Cu has attracted much attention because Cu has been applied as raw material in various industries. Nowadays the popular method to prevent/reduce Cu corrosion is the addition of green corrosion inhibitor. In this work, eugenol, a natural fragrance and flavor extracted from plants, was applied as an eco-

friendly corrosion inhibitor. Eugenol could be used to reduce the corrosion of Cu in the presence of 1 M HCl. However the inhibition efficiency of eugenol for Cu plates at 1 M HCl was found to decrease with increasing temperature from 30°C to 70°C. The data from the weight loss measurement corresponded with images from SEM. Eugenol was found to perform well as a corrosion inhibitor of Cu and also possibly provided another capability of Thai tradition herbs in this research.

Keywords: temperature, inhibition efficiency, eugenol, copper corrosion

Introduction

Corrosion has received a considerable attention because it can damage/ reduce the properties of metals. The effect of corrosion on economic and industry is related to the safety and efficient operation of equipment and structures. Copper (Cu) has been various applied in many industries such as electronic circuit and pipelines (Finsgar & Milosev, 2010) because of its properties e.g. high electrical and thermal conductivities. The corrosion of Cu can occur by many factors e.g. acidic medium, time, pressure, and temperature. Therefore many works have been focused to protect the corrosion of copper from various purposes. One of the several methods of the prevention of corrosion is the use of corrosion inhibitor. Corrosion inhibitors are substances, which contain hetero-atoms such as nitrogen (N), sulphur (S), and oxygen (O) (Özcan et al., 2008; Ramesh & Rajeswari, 2004; Wang, 2006) Mechanism of corrosion inhibitor is the donation of lone pair electron to metal atoms (Ostovari et al., 2009). The addition of corrosion inhibitors to system can control/reduce the corrosion of metals. The adsorption of corrosion inhibitors to metals surface is affected by nature and charge of metals, electronic structure of inhibitors, temperature, and time (Abiola et al., 2009; Ma et al., 2002; Souza & Snelli, 2009) However there are many factors such as cost, toxicity, and availability to consider for the selection of the corrosion inhibitors. Benzotriazole (BTA) is one of the most important inhibitor for copper corrosion because it can be widely applied at broad range of pH and temperature (Stupišek-Lisac et al., 2002). Nevertheless it provides the toxicity. Nowadays green corrosion inhibitors/ eco-friendly corrosion inhibitors have been popularly utilized because of their environmentally friendly (Abd El-Maksoud, 2004; Ismail, 2007; Rocca et al., 2001). Several literatures have been proposed natural corrosion inhibitor for the prevention of metals' corrosion. Plant extracts, animal proteins from meat and milk, and natural honey were used as green corrosion inhibitors for many metals (El-Etre, 1998). The successful ability of corrosion inhibitor for green corrosion inhibitors especially plant extracts is its constitution including the heterocyclic compounds like alkaloids, flavonoids. The purpose of this work was to study the effect of temperature on

inhibition efficiency of eugenol for copper corrosion using the mass loss measurement, and surface characterization. Eugenol is a natural compound, which can be extracted from plant e.g. Pimenta dioica (Rao et al., 2012) and Thai traditional herbs e.g. Alpinia galangal, Curcuma longa, Piper betle, Ocimum africanum, and Zingiber officinale (Teanpaisan et al., 2017). The various qualities of eugenol have been published such as antioxidant, antibacterial activity, food flavoring agent, and additives for cosmetic (Atsumi et al., 2005; Cheng et al., 2008; Zhang et al., 2000). Therefore it would be useful if eugenol can be possibly performed to control copper corrosion and improve the capability of Thai traditional herbs at the same time.

Material and Methods

Material

Copper plates of size 0.5 x 1.5 cm^2 were polished by sandpaper, then were rinsed with deionzed water, ultrasonically degreased in acetone for 1 hour, dried in oven at 80°C for 30 minutes and stored in dessiccator before use. All chemicals and reagents e.g. acetone, hydrochloric acid (HCl), and ethanol were analytical grade and purchased from MERCK, Germany. Eugenol representing inhibitor was analytical grade and purchased by Sigma Aldrich.

Weight loss measurement

The cleaned Cu plates were weighed and immersed in 10% (w/v) eugenol for 5 minutes. Eugenol was performed as inhibitor in this study. Then HCl was added into the test solution at 1 M final concentration. The temperature and homogeneity were controlled using a water bath shaker. The immersion time for the weight loss test was 1 hour at 30 / 40 / 50 / 60 / 70°C. To determine the loss of weight, after the period of immersion was finished, Cu plates were removed from the test solution, washed with ethanol, dried in oven at 80°C for 30 minutes and reweighed. The inhibition efficiency (%IE) was calculated using Equation (1). To compare the inhibition efficiency of eugenol, test solution with and without inhibitor were studied.

$$\%IE = \frac{\left(\Delta m_i - \Delta m_u\right)}{\Delta m_u} \times 100 \quad \text{..... (1)}$$

where Δm_i and Δm_u are the mass loss of Cu with and without eugenol, respectively.

SEM characterization

The morphology of the Cu plates' surface was examined by scanning electron microscope (SEM) (JSM-IT300, Japan).

Statistical analysis

Data were expressed as mean and standard deviation (S.D.) by computational analysis from the triplicate independent experiments.

Results and Discussion

Effect of temperature on inhibition efficiency

The plot of the inhibition efficiency at various temperatures is presented in Figure 1. It is found that at a fix concentration of inhibitor and HCl concentration, the highest inhibition efficiency (50%) occurs at 30°C. Trend of the inhibition efficiency continuously decreases from 30 to 70°C. The decrease may be due to the reduction of physical adsorption of eugenol on the Cu surface and the stability of eugenol at higher temperature (Shimodal et al., 2006). Higher temperature can destroy the physical adsorption between eugenol and Cu surface and also decrease the stability of eugenol.

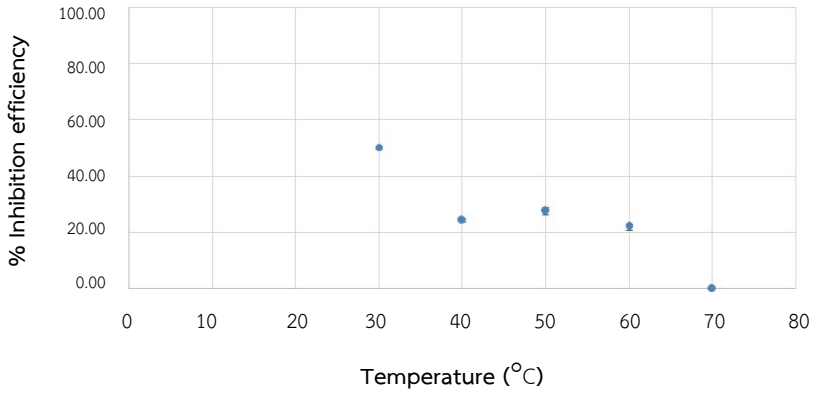

Figure 1 The inhibition efficiency of eugenol in acidic medium at various temperatures

In this work the inhibition efficiency of eugenol for Cu corrosion in the presence of 1 M HCl at 30°C was approximately 50% therefore eugenol acted as effective inhibitor. The reaction between eugenol and HCl could be explained using the transformation of functional group in eugenol from oxy- to hydroxyl in acidic solution. However other inhibitors such as cysteine (Ismail, 2007) and alginate biopolymer (Jmiai et al., 2018) presented as successful inhibitor, which were better than eugenol. The inhibition efficiency of cysteine and alginate biopolymer for Cu corrosion at 30°C was approximately 81 and 78%, respectively.

Surface characterization

Surface examination using SEM was carried out to perform the effect of temperature on Cu surface in acidic medium (1 M HCl) with and without eugenol as corrosion inhibitor. Figures 2a-2b show the surface of Cu at 30°C without/with eugenol,

respectively. Figures 2c-2d show the surface of Cu at 70°C without/with eugenol, respectively. It could be seen that surface of Cu plate at 30°C with eugenol (Figure 2b) is smooth. The adsorption of eugenol on Cu plate can be labeled as physical adsorption and applied for the inhibition of Cu corrosion. Therefore it can be concluded that the inhibition efficiency improves in the presence of eugenol. These results were corresponded with the mass loss measurement. However the stability of eugenol decreases with increasing temperature. Cu corrosion was observed in Figures 2c-2d.

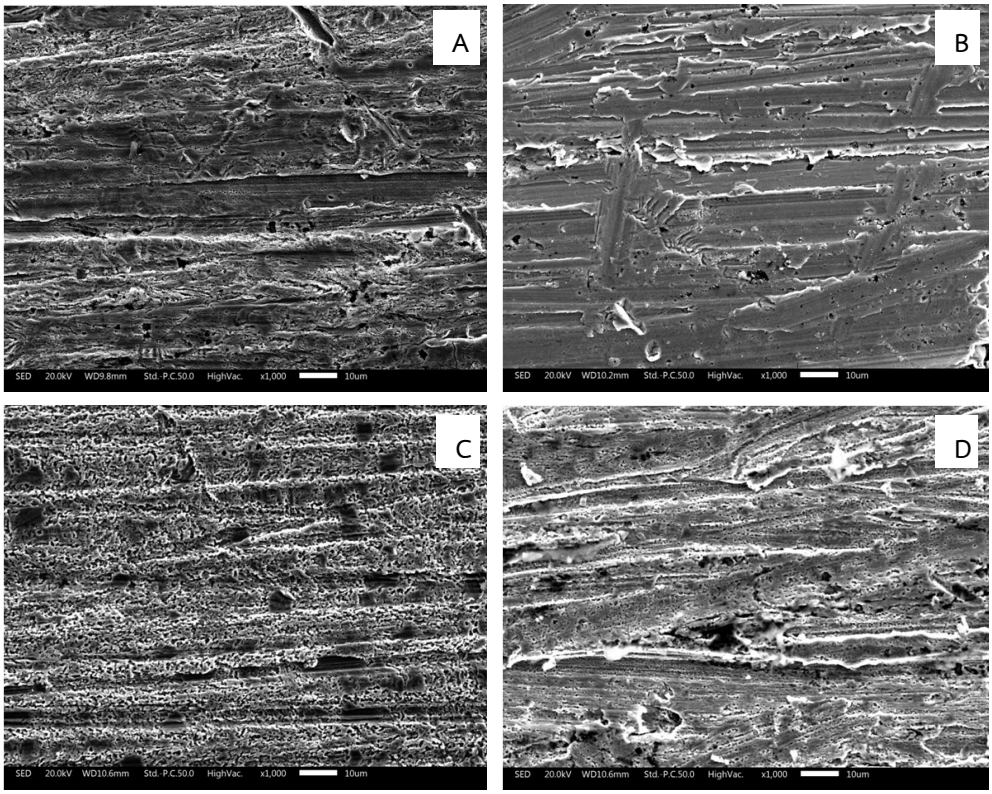

Figure 2 SEM images for Cu plates A) 30°C without eugenol B) 30°C with eugenol C) 70°C without eugenol D) 70°C with eugenol

Conclusion

In this paper, the effect of temperature on inhibition efficiency for Cu corrosion was investigated using the mass loss measurement, and surface characterization. Eugenol acted as a good green inhibitor for copper corrosion in acidic medium (1 M HCl). However the inhibition efficiency decreased with increasing temperature due to the instability of eugenol. The capability of eugenol for the inhibition of Cu corrosion was possibly presented another advantage of Thai traditional herbs.

Acknowledgement

The authors acknowledge Bansomdejchaopraya Rajabhat University for the financial support. This work was carried out at Program of Chemistry, Department of Science, Faculty of Science and Technology, Bansomdejchaopraya Rajabhat University.

References

Abd El-Maksoud SA. Some phthalazin derivatives as non toxic corrosion inhibitors for copper in sulphuric acid, *Electrochimica acta*. 2004; 49: 4205-4212.

Abiola OK, Otaigbe JOE, Kio OJ. *Gossipium hirsutum* L. extracts as green corrosion inhibitor for aluminium in NaOH solution, *Corrosion Science*. 2009; 51: 1879-1881.

Atsumi T, Fujisawa S, Tonosaki K. A comparative study of the antioxidant/peroxidant activities of eugenol and isoeugenol with various concentrations and oxidation conditions, *Toxicology in Vitro*. 2005; 19: 1025-1033.

Cheng SS, Liu JY, Chang EH, Chang ST. Antifungal activity of cinnamaldehyde and eugenol congeners against wood-rot fungi, *Bioresource Technology*. 2008; 99: 5145-5149.

El-Etre AY. Natural honey as corrosion inhibitor for metals and alloys. I. copper in neutral aqueous solution, *Corrosion Science*. 1998; 40: 1845-1850.

Finsgar M, Milosev I. Inhibition of copper corrosion by 1,2,3-benzotriazole: a review, *Corrosion Science*. 2010; 52: 2737-2749.

Ismail KM. Evaluation of cysteine as environmentally friendly corrosion inhibitor for copper in neutral and acidic chloride solutions, *Electrochimica acta*. 2007; 47: 4189-4194.

Jmiai A, Ibrahimi B, Tara A, Issami S, Jbara O, Bazzi L. Alginate biopolymer as green corrosion inhibitor for copper in 1 M hydrochloric acid: Experiment and theoretical approaches, *Journal of Molecular Structure*. 2018; 1157: 408-417.

Ma H, Chen S, Niu L, Zhao S, Li S, Li D. Inhibition of copper corrosion by several Schiff bases in aerated halide solutions, *Journal of Applied Electrochemistry*. 2002; 32: 65-72.

Ostovari A, Hoseinieh SM, Peikari M, Shadizadeh SR, Hashemi SJ. Corrosion inhibition of mild steel in 1 M HCl solution by henna and its constituents (lawsone, gallic acid, a-D-glucose and tannic acid), *Corrosion Science*. 2009; 51: 1935-1949.

Özcan M, Solmaz R, Kardas G, Dehri I. Adsorption properties of barbiturates as green corrosion inhibitors on mild steel in phosphoric acid, *Colloids and Surfaces*. 2008; 325: 57-63.

Ramesh S, Rajeswari S. Corrosion inhibition of mild steel in neutral aqueous solution by new triazole derivatives, *Electrochimica acta*. 2004; 49: 811-820.

Rao PS, Navinchandra S, Jayaveera KN. HPTLC analysis of the essential oil from Pimenta *dioica* leaf, *Journal of Applied Pharmaceutical Science*. 2012; 2(12): 141-148.

Rocca E, Bertrand G, Rapin C, Labrune JC. Inhibition of copper aqueous corrosion by non-toxic linear sodium heptanoate: mechanism and ECAFM study, *Journal of Electroanalytical Chemistry*. 2001; 503: 133-140.

Shimodal K, Kondo Y, Nishida T, Hamada H, Nakajima N. Biotransformation of thymol, carvacrol, and eugenol by cultured cells of Eucalyptus perriniana, *Phytochemistry*. 2006; 67: 2256-2261.

Souza FS, Snelli A. Caffeic acid as a green corrosion inhibitor for mild steel, *Corrosion Science*. 2009; 51: 642-649.

Stupišek-Lisac E, Gazivoda A, Modžarac M. Evaluation of non-toxic corrosion inhibitors for copper in sulphuric acid, *Electrochimica Acta*. 2002; 47: 4189-4194.

Teanpaisan R, Kawsud P, Pahumunto N. Screening for antibacterial and antibiofilm activity in Thai medicinal plant extracts against oral microorganisms, *Journal of Traditional and Complementay Medicine*. 2017; 7: 172-177.

Wang L. Inhibition of mild steel corrosion in phosphoric acid solution by triazole derivatives, *Corrosion Science*. 2006; 48: 608-616.

Zhang R, Kulkarni KA, Kulkarni AP. Oxidation of eugenol by purified human term placental peroxidase, *Placenta*. 2000; 21(2-3): 234-240.