

FORMULATION OF GUAVA LEAF COATING AS ANTICORROSION OF Cu METAL CULTURAL HERITAGE MATERIAL

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Abstract

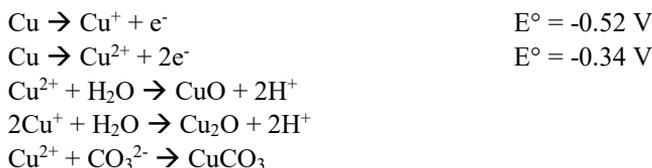
This paper explores the successful use of guava leaf extract for inhibiting corrosion of metal coin CHM. The metal content is determined using X-ray Diffractometer instruments for qualitative and Atomic Absorption Spectroscopy for quantitative analysis. Two different bases are used for the coating formulation – Vaseline album and lanolin – and the mix of both. By testing three different options, such as physical properties (spread and adhesion test), coating quality (pH and viscosity test) and anti-corrosion test (weight addition and copper content). Analytical results show that the constituent element is Cu, while corrosion solids contains Cu, Zn, and Fe with the concentration of 0.135; 0.0029; 0.0029 g/g corrosion respectively, and the minerals contained in corrosion solids are Cu₂O, CuO, Cu₂(OH)₂CO₃, dan FeCO₃. F1 had the widest spread ability test, measuring 10.75 cm², while F3 had the largest adhesion result, measuring 15.33 s. F3, with 950 dPas, was the thickest coating. F1 had the highest pH test result of 4.84. F1, F2, and F3 have weight additions of (-2.7) mg, 0 mg, and 3.1 mg, respectively. F3 has the lowest copper content (5.667 ppm). The F3 with a 100% lanolin base was deemed to be the best result based on the results.

Keywords: Metal coin conservation; Tannin; Oinment; Anti-corossion; AAS

Introduction

Copper and copper based alloys (e.g. bronzes) undergo degradations mainly caused by water (rain, moisture) but also related to air pollution in outdoor environment [1]-[4]. The atmospheric corrosion of these objects leads to their physical and aesthetic modifications by the interaction of the environment. The corrosion process in copper metal and its copper-based alloys occurs due to the presence of water vapor, which forms two oxide layers, Cu₂O and CuO, while the presence of CO₃²⁻ (carbonate) ions can cause precipitation in the form of CuCO₃, which is greenish blue in color [5]. The reaction to form copper oxide due to the presence of air in the environment, which forms CuO, Cu₂O and CuCO₃.

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Based on the reaction, oxidation of Cu metal produces Cu^+ ions, these Cu^+ ions are further oxidized to become Cu^{2+} . The E° value is the reduction potential which describes the ease with which the metal can be reduced or oxidized. The more negative the E° value, the easier it will undergo oxidation, Cu metal oxidized to Cu^{2+} will experience corrosion more easily than Cu^+ . Cu^{2+} ions react with carbonate (CO_3^{2-}) in CuCO_3 deposits, and also form CuO and Cu_2O oxides in the presence of water vapor.

The corrosion layer, also called patina for bronze artwork [6], is known to be formed through a two-stage process in the case of pure copper [7]-[11] and bronze [12]-[18]. Corrosion damages cultural heritage objects made of metal, and can eliminate important information contained in the cultural heritage objects, so cleaning corrosion on cultural heritage objects made of metal needs to be done.

XRD, SEM-EDX, and micro-FTIR analysis methods have been widely applied in the conservation studies of ancient metals, for example in Bronze Age artifacts and spearheads. These techniques enable the identification of primary, secondary, and contamination patinas [19], [20], [21], [22].

To protect the cultural heritage objects from the atmospheric corrosion, different types of treatments are used, varnishes, waxes, and corrosion inhibitors [23], [24]. The advantage of varnishes and waxes is that they do not significantly modify the surface appearance but are efficient. Indeed, such treatments create a physical barrier on the surface and thus prevent interactions between the atmosphere and the objects. However, varnishes have the disadvantage of being difficult to reprocess and waxes of having a bad hold in time. Concerning corrosion inhibitors, including benzotriazole (BTA) which is currently used for restoration, several studies have reported a possible toxicity of this type of inhibitor for the environment [25], [26], [27].

The search for new corrosion inhibiting treatments, non-toxic, is a major issue in the field of heritage. In addition to synthetic compounds, plants are considered to function as a substitute for inhibitors in the corrosion process. The use of plants can also reduce environmental pollution, cheaper prices and easily available, and the effectiveness of good corrosion process inhibition is the basis for choosing extracts from plants expected to replace toxic corrosion process synthetic inhibitors [28]. Therefore, there is a need for corrosion process inhibitors that are safe, cheap, biodegradable and environmentally friendly. Examples of natural corrosion inhibitors derived from plant secondary metabolite compounds are tannins [29], [30].

Tannins have the ability to form complex compounds because they have free pair elements that function to donate electrons to M^+ metals and form chelates with M^+ metals. This chelate functions to coat the CHM metal coin to avoid the formation of corrosion again (Fig. 1). Tannin compounds can be found in plants that have an astringent taste. The largest tannin-producing plants are moringa leaf [31], butterfly pea [32], papaya seed [33], guava leaf [34], red betel [35], gotu kola herb [36], white radish [37], lamtoro leaf [38], and tea. Tannin is an effective functional ligand that functions as a chelating agent for Cu (II) metal [39].

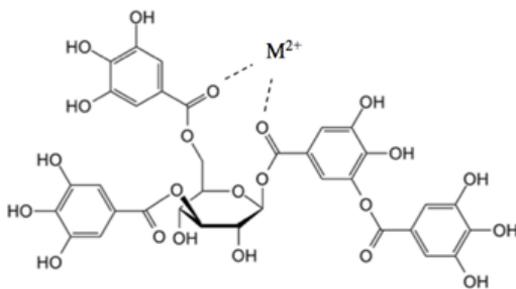


Fig. 1. Complex formation of tannin with metals

The use of guava leaves as a natural inhibitor of metal corrosion is done by immersing the CHM metal in guava leaf powder solution or guava leaf extract solution. Immersion of CHM metal in inhibitor solution is considered impractical, because large CHMs require space and a large volume of inhibitor solution and must be removed. In addition, this method is technically difficult to perform. Coating CHMs with natural inhibitor compounds can be made easier by dispersing the inhibitor compounds in coating preparations. This is because the use of coating is considered to be able to facilitate the application of inhibitor compounds on large-sized CHMs.

In general, coatings are used on porous objects such as human skin and animal skin. The use of anti-corrosion coating on CHM metal coins is based on the appearance of the surface morphology of CHM metal coins which proves the presence of pores on the surface of CHM metal coins which are believed to appear due to corrosion (Fig. 2) [40].

The difference in morphology between the surface of human skin or animal skin and the surface of CHM metal coins is not expected to affect the effectiveness of the coating applied to CHM metal coins, so that the active inhibitor compounds contained in the coating can maximally protect the surface of CHM metal coins.

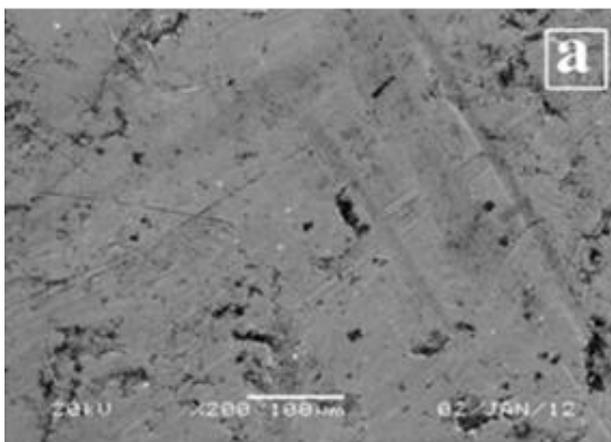


Fig. 2. SEM image of the surface of CHM metal coins from Najran Hoard, Saudi Arabia [40]

Furthermore, the coating will be tested for physical properties including pH, viscosity, adhesion and spread ability as well as testing the ability of the coating as an anticorrosive.

Experimental part

The primary objective of this research is to examine the metal content of metal coin corrosion and how guava leaves extract inhibit metal coin corrosion. This semi solid product are unique and never been made before.

Materials

Metal coins of CHM category were purchased at Beringharjo Market Yogyakarta 20 coins; Guava leaves taken from Bantul area; Nitric Acid (HNO₃); Ethanol 70%; 1 M Hydrochloric Acid (HCl); and 1000 ppm standard solutions for Cu, Fe, Zn, and Pb metals which are all Pro Analysis (PA) quality E-Merck brand; aqua bides; distilled water; Vaseline Album and Adeps Lanae/Lanolin with pharmaceutical grade.

Laboratory glassware, analytical balance, X-ray Diffractometer (XRD) (Shimadzu-6000), pH meter, Atomic Absorption Spectrophotometer (AAS), viscometer (Rion Viscotester VT-04), maceration vessel, rotary evaporator, water bath, equipment test for adhesion, and spread ability.

Methods

The analytical techniques selected to conduct this investigation

Analysis with XRD instrument

A layer of approximately 1 mg was taken from the surface of the CHM metal coin and pulverized. This sample was then loaded into a sample specimen and inserted into an XRD device operated with Cu target metal and Ni pellet at a diffraction angle of 3-80°, so that the diffraction pattern of the sample was obtained. The sample diffraction pattern obtained was then interpreted by comparing the sample diffraction pattern obtained with the corresponding standard diffraction pattern [41].

Analysis and identification with AAS device

A weight of 0.041 g of the coating on the surface of CHM metal coins was dissolved in 10 mL of concentrated nitric acid solution and heated to a volume of approximately half of the initial volume. This solution was then diluted to a volume of 100 mL and then analysed with an AAS device at a wavelength corresponding to the elements Cu, Fe, Pb, and Zn whose concentrations were to be determined. The concentration calculation is done by comparing the absorbance of the sample with the absorbance of the standard solution. From this step, the levels of Cu, Fe, Pb, and Zn metals in the corrosion solids of CHM metal coins can be obtained.

Preparation of guava leaf concentrated extract

Guava leaves were dried for 7 days to reduce their moisture content, then blended. Guava leaf powder of 250 grams was soaked in 750 mL of 70% ethanol, covered with aluminum foil and left for 24 hours while occasionally stirring. Next, the soaked sample was filtered using filter paper to produce filtrate 1 and pulp 1. The dregs were then added with 750 mL of 70% ethanol solution, covered with aluminum foil and left for 2 days, the sample was filtered using filter paper to produce filtrate 2 and pulp 2. Filtrates 1 and 2 were mixed together. The filtrate obtained was evaporated using a rotary evaporator with a temperature of 70°C and a speed of 40 rpm. Furthermore, the extract is evaporated with a water bath at a temperature of 60°C so that a thick extract of guava leaves is obtained [42].

Preparation of guava leaf extract

In this study, three formulas were made with different variations in the amount of base used. Formula one used a water-insoluble hydrocarbon base, vaseline album, formula two used 50% hydrocarbon base and 50% absorbent base, while formula three used an absorbent base, adeps lanae. The formulation of each coating is listed in Table 1.

Table 1. Guava leaf extract formulation with hydrocarbon base and absorbent base

| Materials | Formula 1 | Formula 2 | Formula 3 |
|------------------------|-----------|-----------|-----------|
| Guava leaf extract (g) | 0.9 | 0.9 | 0.9 |
| Vaseline album (g) | 9.1 | 4.55 | 0.0 |
| Adeps lanae (g) | 0.0 | 4.55 | 9.1 |
| Total (g) | 10.0 | 10.0 | 10.0 |

Preparation of guava leaf extract begins with heating the mortar and stamper to be used while weighing the necessary ingredients. Once hot, the coating base is added first, followed by

guava leaf extract. After the ingredients enter, stirring is done quickly so that the coating is homogeneous.

Examination of physical properties, quality and anticorrosive effect of guava leaf extract

Examination of the physical properties of the coating solution

Spreadability test

This test is conducted to determine the area of spread of the coating on the object. This test is carried out in the following way: 0.5 gram of coating is placed on the glass in the center. The other glass is weighed and placed on top of the coating mass and left for 1 minute, then measured the diameter of the coating that spreads (by taking the average length of the diameter from several sides). Next, add 50 grams of additional weight, then let it stand for 1 minute and record the diameter of the coating spread as before. Next, add another 50 grams load and record the diameter of the coating that spreads as before until a constant diameter is obtained. A coating is said to be good if its spreading power is large which can be seen from the large diameter.

Adhesion test

This test is conducted to determine the length of coating attached by weighing the coating on a glass object that has been determined in area, then placing another glass object on top of the coating and pressed with a load of 1 kg for 5 minutes. Next, install the glass object on the test device, then release a load weighing 80 grams and record the time until the two glass objects are released. Coating is said to be good if the adhesion is large on the smeared object, because the active substance will stick longer so that it can produce the desired effect.

Quality inspection

pH examination

The pH examination is carried out by preparing 0.5 grams of coating dissolved in 30 mL of distilled water, then measuring the pH value using a pH meter until it shows a constant pH value.

Viscosity test

The coating solution viscosity test was conducted with a viscosimeter (Rion Viscotester VT-04). The viscosimeter was mounted on its clamps in a horizontal direction or perpendicular to the direction of the clamps. The rotor was then attached to the viscosimeter by locking it counterclockwise. The bowl is filled with the sample to be tested, the rotor is placed right in the middle of the coating, then the device is turned on and when the rotor starts rotating the viscosity pointer needle will automatically move to the right then after stabilizing, the viscosity is read on the scale of the rotor used.

Anticorrosive activity test

Anticorrosive activity testing on guava leaf extract was carried out by applying 1 gram of coating from three formulas on the surface of CHM metal coins. Furthermore, inserting metal coins that have been smeared with coating into a container and added with 1M HCl solution [43] for 24 hours. After 24 hours the coin was removed and cleaned to dry and weighed to determine the weight gain on the coin due to corrosion during immersion using 1M HCl. The remaining coin soaking solution was then diluted and read using AAS to determine the Cu concentration of CHM metal coins dissolved during soaking using 1M HCl.

Results and discussion

Analysis of corrosion solids found on CHM metal coins

Analysis of corrosion solids on the surface of CHM metal coins was carried out to determine the composition of corrosion solids covering the surface of the metal coins. The analysis of the types of elements and compounds that make up the corrosion solids attached to the surface of the metal coins was carried out with an X-ray Diffractometer (XRD), while the analysis of the composition of the corrosion solids on the surface of the metal coins was carried

out with an Atomic Absorption Spectrophotometer (AAS). The elemental and compound diffractograms of the corrosion solids on the surface of CHM metal coins are shown in Fig. 3.

Fig. 3, shows that the corrosion solids on the surface of CHM metal coins are semi-crystalline, this is evidenced by the peak broadening at d values of 3.7045; 2.8588; 2.4662; 2.095; 1.8145; and 1.279 with the largest peak broadening. Identification of the constituents of the corrosion solids attached to the surface of CHM metal coins was carried out by comparing the diffraction angles (2θ) of the samples with the standard diffraction angle (2θ) of JCPDS (Joint Committee on Powder Diffraction Standard). The comparison of the 2θ value of the sample with the standard 2θ value is presented in Table 2.

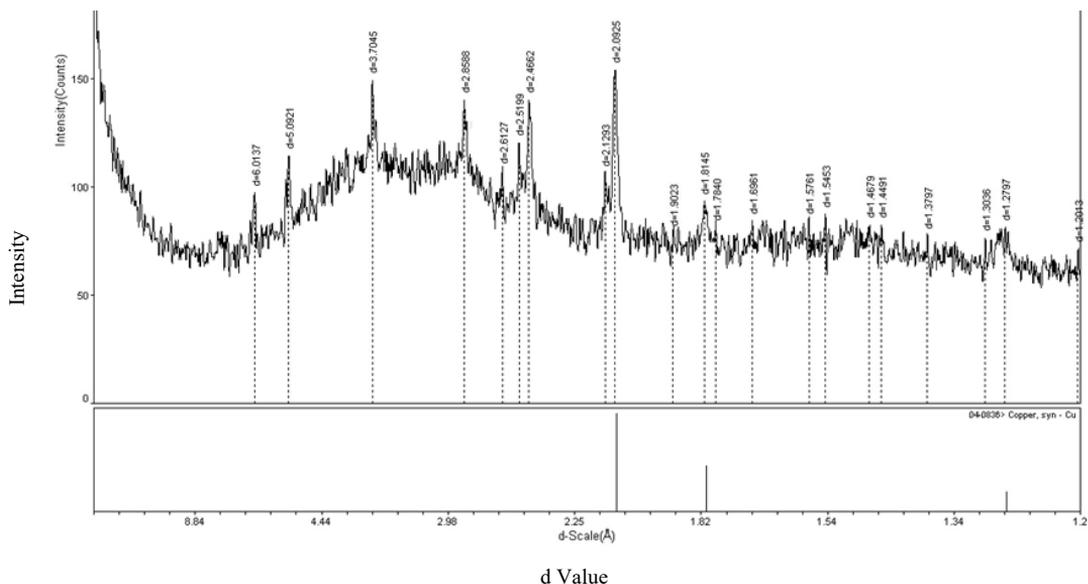


Fig. 3. Elemental diffractograms of corrosion solids on the surface of CHM metal coins

Table 2. Diffraction angles of Cu in corrosion solids and standard Cu

| Diffraction angle of the corrosion solid (2θ) | Standard diffraction angle (2θ) JCPDS of copper: 04-0836 |
|--|--|
| 43.200 | 43.297 |
| 50.241 | 50.433 |
| 74.018 | 74.130 |

Based on the data presented in Table 2, the diffraction angle values in the sample and Cu standard obtained from JCPDS, Copper: 04-0836 data have no significant difference. This it can be confirmed that the constituent element of corrosion solids on the surface of CHM metal coins is Cu metal.

Furthermore, the identification of the compounds that make up the corrosion solids on the surface of CHM metal coins is done by comparing the inter-plane distance (d) in the sample with the d value of several metal oxides from JCPDS. Comparison of sample peaks with metal oxide peaks is presented in Table 3.

Table 3. Data on d values in samples and standards

| d(A) sample | d(A) JCPDS standard mineral | | | |
|-------------|-----------------------------|-------|---|-------------------|
| | Cu ₂ O | CuO | Cu ₂ (OH) ₂ CO ₃ | FeCO ₃ |
| 1.545 | 1.510 | 1.510 | 1.560 | 1.506 |
| 2.129 | 2.135 | 2.120 | 2.128 | - |
| 1.784 | - | - | 1.760 | 1.731 |
| 1.696 | - | - | - | 1.738 |
| 2.466 | 2.465 | 2.450 | 2.477 | - |
| 1.280 | - | 1.280 | 2.320 | 2.346 |

Table 3, shows that the d values of the samples are consistent with the d values of Cu₂O, CuO, Cu₂(OH)₂CO₃, and FeCO₃. This indicates that the substances contained in the corrosion solids on the surface of CHM metal coins are Cu₂O, CuO, Cu₂(OH)₂CO₃, and FeCO₃. The elemental composition of the corrosion solids on the surface of CHM metal coins is presented in Table 4 and appendix 3.

Table 4. Elemental content of Cu, Fe and Zn metals in corrosion solids on the surface of CHM metal coins

| Elements | g/g corrosion |
|----------|---------------|
| Cu | 0.1350 |
| Fe | 0.0029 |
| Zn | 0.0029 |

Table 4, shows that the metal elements Cu, Fe and Zn are present in the corrosion solids on the surface of the metal coins. This indicates that the metal alloy used in the CHM metal coins is brass. Brass is a metal alloy between copper (Cu) and zinc (Zn) and other metals white tin (Sn), lead (Pb), iron (Fe) in small amounts as accompanying metals [44].



Fig. 4. CHM metal coin covered with corrosion solids on its surface

Fig. 4, shows the green color of the corrosion solids that dominate on the surface of the coin, this indicates that the most component in the corrosion solids is Cu carbonate which is green in color. This is reinforced by the XRD results (Table 3) where the component (Cu₂(CO₃)(OH)₂) dominates over other minerals.

Extraction

The results of guava leaf extraction by maceration using 70% ethanol solvent obtained a concentrated extract weighing 5.54 g, so that the yield obtained was 11.08%. Images and tables of guava leaf extract characteristics are presented in Fig. 5 and Table 5.

Table 5. Guava leaf extract characteristics

| Characteristics of the Extract | Results |
|--------------------------------|-------------------------|
| Yield | 11.08% |
| Color | Dark Green |
| Texture | Thick |
| Smell | typical of guava leaves |

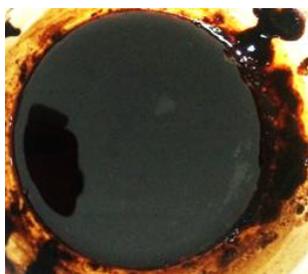


Fig. 5. Concentrated extract of guava leaves

Formulation of coating

Guava leaf extract (*Psidii guajava* L) that has been formulated is observed organoleptically, namely color, odor and consistency of the coating solution. The observation results of guava leaf extract (*Psidii guajava* L) coating preparation formulation using hydrocarbon base and absorbent base can be seen in Fig. 6 and Table 6.

Based on visual observations, the three coating formulas have colors and consistencies that are not much different, and have the same smell, namely the typical smell of guava leaves. The consistency of F3 coating produces a stickier and thicker coating mass when compared to F1 and F2 coating, this is because F3 coating uses adeps lanæ base which causes the coating to become stickier and denser.

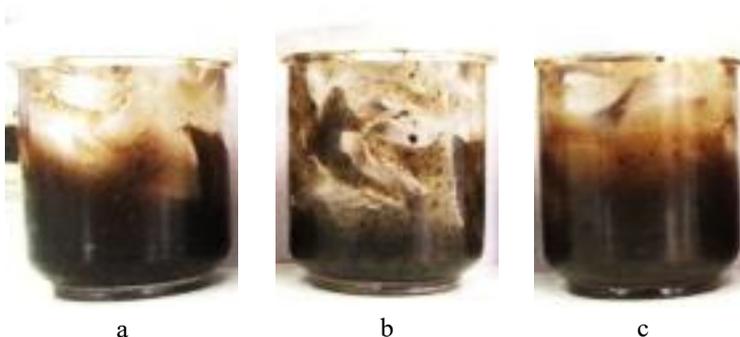


Fig. 6. Guava leaf extract coating based on its formulation:
 (a) F1 100% Vaseline, (b) F2 50% Vaseline: 50% Adeps Lanæ, (c) F3 100% Adeps Lanæ

Table 6. Organoleptical observation of the coating

| Formula | Color | Smell | Consistency |
|---------------|------------------------|----------------------|---------------------------|
| F1: 100%V | Blackish brown | Typical guava leaves | Thick |
| F2: 50%V:50%A | Fainter blackish brown | Typical guava leaves | Thick and slightly sticky |
| F3: 100%A | Deeper blackish brown | Typical guava leaves | Very thick and sticky |

Description: V = Vaseline Album; A = Adeps Lanæ

Test Results of Coating Physical Properties and Coating Quality

Examination of the physical properties of the coating was carried out to compare the two coating formulations between hydrocarbon base and absorbent base. The examination of the physical properties of the coating included the adhesion test and the spreadability test, while the examination of the quality of the coating with viscosity test, pH test and corrosion inhibition test on CHM metal coins with hydrochloric acid media.

Spreadability Test

This coating spreadability test is to determine how wide the surface area of the object can be reached by the coating. An coating preparation is expected to be able to spread easily on the

surface of the object, without using significant pressure. The easier it is to spread, the greater the surface area of contact with the active substance contained in the coating with the object of application, so that the absorption of the active substance in the place of application is optimized. The spreading power is inversely proportional to the viscosity, the lower the viscosity, the higher the spreading power [45]. The spreading surface that is produced with an increase in the added load is characteristic of the spreadability of the coating. The results of observation data on the spreadability of guava leaf extract coating can be seen in Table 7.

Table 7. Observation of spreadability of guava leaf extract coating

| Formula | Diameter average (cm) | Radius (cm) | Area of a circle (cm ²) |
|---------------|-----------------------|-------------|-------------------------------------|
| F1: 100%V | 3.70 | 1.85 | 10.75 |
| F2: 50%V:50%A | 3.50 | 1.75 | 9.62 |
| F3: 100%A | 3.30 | 1.65 | 8.55 |

Description: V = Vaseline Album; A = Adeps Lanae

Based on the results of the spreadability test, F1 with 100% vaseline album composition has the largest spreadability, while the smallest spreadability is owned by F3 with 100% adeps lanae base. Overall, the surface area that can be reached is better by the hydrocarbon base. This is because the consistency of hydrocarbon base coating (vaseline album) is softer than absorbent base coating (adeps lanae).

Adhesion Test

Adhesiveness shows the tendency of a material to stick to other materials. This test serves to determine how long the coating adhesion time on the object. The thicker or more concentrated the consistency, the longer the time needed to separate the two glass objects. So, if the consistency of the coating is thicker, the contact of the active substance on the surface of the object is also longer. The results of the guava leaf extract coating adhesion test can be seen in Table 8.

Table 8. Observation results of guava leaf extract coating adhesion test

| Formula | Adhesion time (seconds) |
|---------------|-------------------------|
| F1: 100%V | 4.67 |
| F2: 50%V:50%A | 8.67 |
| F3: 100%A | 15.33 |

Description: V = Vaseline Album; A = Adeps Lanae

Based on Table 8, the results of the adhesion test, F3 has a longer adhesion time than F1 and F2, this shows that the thicker the consistency of a preparation, the higher the adhesion time. The longer the adhesion time of an coating, the longer the contact of the coating with the object, so the longer the possibility of the active substance will protect the object. The adhesion value is inversely proportional to the resulting spreadability value. This shows that the adhesive ability of the coating has an inverse correlation with its spreading ability, namely the lower the spreading ability, the greater the adhesive ability [45]. Overall, absorbent bases have a longer adhesion time than hydrocarbon bases. This is because the absorbent base (adeps lanae) has a higher viscosity than the hydrocarbon base (vaseline), so its ability to adhere to the object is also longer.

Viscosity Test

Viscosity testing was conducted to determine the viscosity of the coating using a viscometer (VT-04E Rion Viscotester). Viscosity is a statement of the resistance of the liquid to flow, the higher the viscosity, the greater the resistance [46]. The observation results of the coating viscosity test can be seen in Table 9.

Table 9. Observation results of viscosity test of guava leaf extract coating

| Formula | Coating viscosity (dPas) |
|---------------|--------------------------|
| F1: 100%V | 503 |
| F2: 50%V:50%A | 600 |
| F3: 100%A | 950 |

Description: V = Vaseline Album; A = Adeps Lanae

Table 9 shows that the hydrocarbon base (F1) has a smaller viscosity than the coating using Table 9, shows that the hydrocarbon base (F1) has a smaller viscosity than the coating using absorbent base (F3). Absorbent base coating in its formulation uses adeps lanae, so the consistency is denser when compared to hydrocarbon coatings that use vaseline album.

The denser the coating mass, the greater the viscosity. Viscosity is closely related to the spreadability of the coating on the object. The greater the viscosity, the smaller the spreading power. Coatings that have a low viscosity will make it easier to use and take from the container easier because the consistency is soft [45]. The viscosity of the coating is also closely related to its adhesiveness, because the higher the viscosity, the longer the ability of the coating to adhere.

pH Test

pH examination is one part of the physico-chemical examination criteria in predicting the stability of coating preparations. The pH profile will determine the stability of active ingredients in an acidic or alkaline atmosphere [47]. The results of the pH test observations are presented in Table 10.

Table 10. Observation results of pH test of guava leaf extract coating

| Formula | pH |
|---------------|------|
| F1: 100%V | 4.84 |
| F2: 50%V:50%A | 4.12 |
| F3: 100%A | 3.88 |

Description: V = Vaseline Album; A = Adeps Lanae

Based on Table 10, the pH for hydrocarbon base coating (vaseline album) is higher than absorbent base coating. This is because hydrocarbon coatings contain a lot of oil. States that the pH of o/w coating bases is always lower than w/o because water is more acidic than oil.

Polyurethanes (PUs) can be tailored into various forms, including coatings, where they serve as protective barriers against aggressive environments. The durability of PU coatings, however, is often compromised by electrolyte diffusion beneath the film, leading to corrosion product formation, coating disbondment, and reduced charge transfer resistance (R_{ct}) [48], [49].

A comparable mechanism applies to guava leaf extract coating used as a bio-based coating for copper heritage materials. While tannins act as natural inhibitors by complexing with Cu(II) ions, the coating's effectiveness depends on physical properties such as viscosity, adhesion, and spreadability. Insufficient adhesion or porosity allows corrosive agents to penetrate, producing compounds like CuO, Cu₂O, and Cu₂(OH)₂CO₃ at the metal coating interface.

Corrosion Inhibition of CHM Metal Coins Using Guava Leaf Extract Coating

CHM metal coins that have been cleaned of corrosion solids will experience corrosion again during storage. The use of tannin obtained from guava leaf extract is then dispersed into coating formulated in three formulas. The coating was used to prevent the formation of corrosion solids on the surface of CHM metal coins. The effect of preventing further corrosion process was expressed as weight gain between before immersion in 1M HCl solution and after immersion, and dissolved Cu content in 1M HCl solution. The weight gain of CHM metal coins in 1M HCl solution for 24 hours is presented in Table 11.

Table 11. Weight addition of CHM metal coins in 1M HCl solution for 24 hours

| Formula | Weight gain (mg) |
|---------------|------------------|
| F1: 100%V | -2.7 |
| F2: 50%V:50%A | 0.0 |
| F3: 100%A | 3.1 |

Description: V = Vaseline Album; A = Adeps Lanac

Table 11, shows that the formation of 3.1 mg of corrosion solids on the surface of CHM metal coins cannot be prevented using F3. While in the application using F1 there is a dissolution of the main metal which results in a reduction in the weight of the CHM metal coin, this is because F1 cannot withstand dissolution by 1M HCl. Furthermore, F2 provides the best protection for CHM metal coins in 1M HCl solution media for 24 hours, it can be seen that in F2 there is no increase or decrease in weight on the coin. The appearance of CHM metal coins before and after being soaked using 1M HCl solution for 24 hours with F1, F2 and F3 can be seen in Fig. 7, Fig. 8 and Fig. 9.



Fig. 7. View of CHM metal coins smeared with F1 coating before (a) and after (b) immersion in 1M HCl solvent



Fig. 8. View of CHM metal coins smeared with F2 coating before (a) and after (b) immersion in 1M HCl solvent



Fig. 9. View of CHM metal coins smeared with F3 coating before (a) and after (b) immersion in 1M HCl solvent

In addition to using three coating formulas, tests were also conducted using positive and negative controls to determine the difference in results between coatings and controls. The weight difference of CHM metal coins using the control in 1M HCl solution is presented in the following table.

Table 12. Weight difference of CHM metal coins using control in 1M HCl solution

| Control | Weight gain (mg) |
|--------------------------------|------------------|
| Control (-) | -472.5 |
| Control (-) V100% | -81.7 |
| Control (-) V50%:A50% | -402.2 |
| Control (-) A100% | -12.8 |
| Control (+) guava leaf extract | -228.3 |

Description: V = Vaseline Album; A = Adeps Lanae

Table 12, shows that the best protection is given by the negative control with 100% adeps lanae, while the lowest protection is shown in the negative control which is not given any application. The appearance of CHM metal coins before and after being soaked using 1M HCl solution for 7 days with various controls can be seen in Fig. 10 to Fig. 14.



Fig. 10. View of CHM metal coin control (-) before (a) and after (b) immersion in 1M HCl solvent



Fig. 11. View of CHM metal coins smeared with 100% Vaseline Album before (a) and after (b) immersion in 1M HCl solvent



Fig. 12. View of CHM metal coins smeared with Vaseline Album 50%:Adeps lanae 50% before (a) and after (b) immersion in 1M HCl solvent



Fig. 13. Appearance of CHM metal coins smeared with 100% Adeps Lanae before (a) and after (b) immersion in 1M HCl solvent



Fig. 14. View of CHM metal coins treated with guava leaf extract before (a) and after (b) immersion in 1M HCl solvent

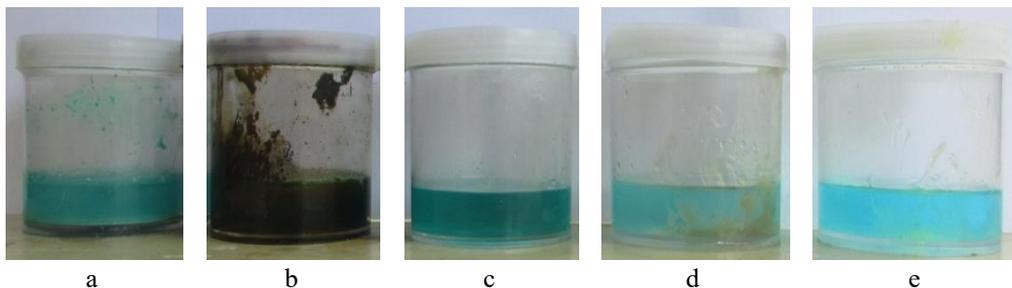


Fig. 15. Residual soaking solution of CHM metal coins in 1M HCl at negative control (a), positive control (b), 100% vaseline album negative control (c), 50% vaseline album:50% adeps lanae negative control (d), and 100% adeps lanae negative control (e).

Furthermore, the corrosion process inhibition test on CHM metal coins can be determined based on the dissolved Cu content. The dissolved Cu content in three coating formulas using 1M HCl media can be seen as follows.

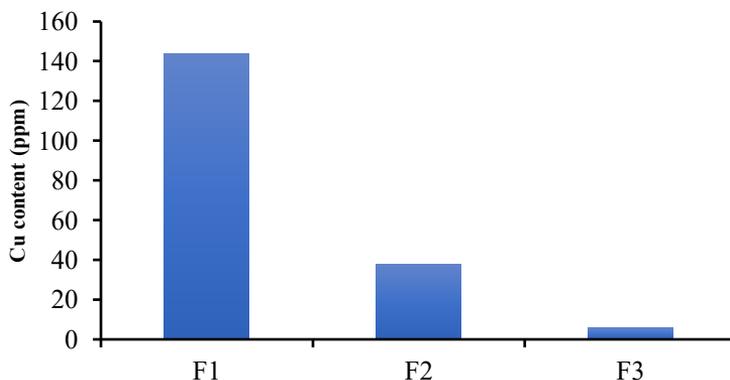


Fig. 16. Cu content dissolved in 1M HCl media for 7 days:
 F1 = Guava leaf extract coating with 100% Vaseline Album base;
 F2 = Guava leaf extract coating with a base of 50% Vaseline Album: 50% Adeps Lanae;
 F3 = Guava leaf extract coating with 100% Adeps Lanae base

Fig. 16, shows that the lowest Cu content is found in the application of F3, which uses 100% Adeps lanae base, this is because the higher the Cu content, the less the ability of the coating to prevent the corrosion process of CHM metal coins in 1M HCl solution media.

The test results on weight gain and dissolved Cu content show that F3 provides the best protection against CHM metal coins with 1M HCl solution media compared to F1 and F2. This is because the structure of adeps lanae has a CN⁻ ligand that functions to bind metal M. Based on the spectrochemical sequence, the difference in orbital energy (Δ) generated in the CN⁻ ligand is quite large, so the bond strength between N and metal M is also large, this causes the chelate

formed to be difficult to oxidize. In contrast, the interaction between M metal and O occurs electrostatically, where the bond between the two is weaker than the bond between N and M metal, so that the chelate formed between O and M metal is more easily oxidized.

Predicted reactions to prevent further corrosion process by vaseline album with tannin and adeps lanae with tannin are presented in Fig. 17 and Fig. 18. The reaction that occurs between vaseline album and tannin in Fig. 17 is an esterification reaction with an acid catalyst, while the reaction between adeps lanae and tannin in Fig. 18 is a pinner reaction.

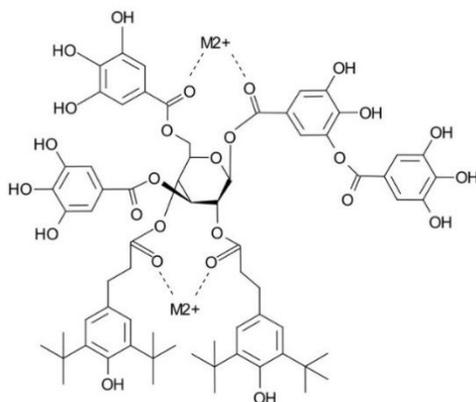


Fig. 17. Predicted reaction of tannins contained in guava leaf extract with Vaseline Album and metal

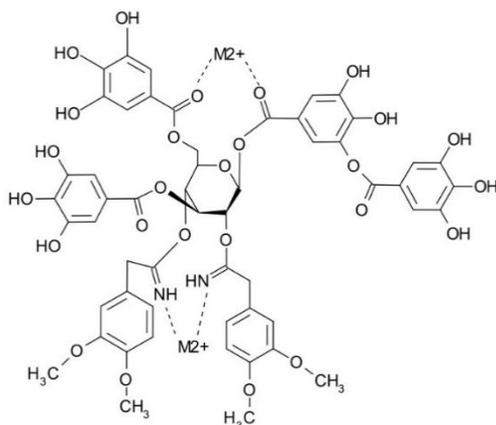


Fig. 18. Predicted reaction of tannins contained in guava leaf extract with adeps lanae and metals

Conclusions

The result obtained from the experiment that the constituent element of corrosion solids on the surface of CHM metal coins is Cu metal by using XRD instrument, while corrosion solids on the surface of CHM metal coins contains Cu, Zn, and Fe with the concentration of 0.135; 0.0029; 0.0029 g/g corrosion respectively, and the minerals contained in corrosion solids are Cu₂O, CuO, Cu₂(OH)₂CO₃, dan FeCO₃. The spread ability test for F1, F2, and F3 are 10.75 cm²; 9.62 cm²; and 8.55 cm². The adhesion for F1, F2, and F3 are 4.67 s; 8.67 s; and 15.33 s. The viscosity test for F1, F2, and F3 are 503; 600; and 950. The pH test for F1, F2, and F3 are 4.84; 4.12; and 3.88. The weight addition for F1, F2, and F3 are (-2.7) mg; 0 mg; and 3.1 mg. The Copper content was analyzed using AAS, and the result for F1, F2, and F3 are 143.909 ppm; 37.977 ppm; 5.667 ppm.

In conclusion, the optimum coating formula was found in the F3 with 100% lanolin base formulation. Because it has the best protection against CHM metal coins with 1M HCl solution media compared to F1 and F2. It's because the structure of adeps lanae has a CN⁻ ligand that functions to bind metal M. Based on the spectrochemical sequence, the difference in orbital energy (Δ) generated in the CN⁻ ligand is quite large, so the bond strength between N and metal M is also large, this causes the chelate formed to be difficult to oxidize.

Acknowledgments

We would like to thank to Alma Ata University and Gadjah Mada University for providing exceeding adequate research facilities.

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Received: June 02, 2025

Accepted: January 24, 2026