



Henna leaves extract as a corrosion inhibitor in acrylic resin coating



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ABSTRACT

This study utilizes optical measurements, thermo-impedance analysis, potentiodynamic polarization studies and morphology observations of henna leaves extract (HLE) incorporated in an acrylic resin coating. The acrylic resin coating with 0.2 wt/vol% HLE (AC2) had the best performance protecting metal from corrosion. XRD and DSC analysis demonstrate that an increase in the crystallite size limits the close packed structure, which increases the free volume and reduces the T_g of the coating. Open circuit potential (OCP) measurements demonstrate that the AC2 coating has a uniform potential due to the lower rate of coating barrier destruction. Electrochemical impedance spectroscopy (EIS) indicates that AC2 has the highest coating resistance, R_c ($4.79 \times 10^8 \Omega$), and lowest coating capacitance, C_c ($3.32 \times 10^{-9} \text{ F/cm}^2$). An elevation in temperature caused coating deterioration for all of the coatings. AC2 has the lowest dielectric constant, ϵ_r , indicating less water uptake and lower ionic conductivity. An additional study of potentiodynamic polarization demonstrates that AC2 has shifted to the noble potential and gives the lowest corrosion current density, i_{corr} , reading. The corrosion rate is the lowest for AC2 ($3.93 \times 10^{-7} \text{ mm/year}$), while the polarization resistance is the highest at $7.44 \times 10^7 \Omega$. An SEM morphology study indicates that AC2 has lesser delamination and greater coverage of HLE in the coating.

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1. Introduction

Corrosion is a natural phenomenon in which metals deteriorate because of electrochemical processes. Corroded metals cause major losses due to the wide application of metals in various industries. Hence, anticorrosive coatings were introduced to retard the corrosion process. For reliable long term performance, inorganic pigments that release corrosion inhibiting substances are usually added to the coatings. However, nearly all powerful corrosion inhibitors have detrimental effects on both the environment and health due to their toxic and carcinogenic nature.

In recent times, researchers have focused their attention on developing greener products due to several social and economic challenges such as increasing greenhouse gas emissions, global warming, depletion of petroleum sources and fluctuations in crude oil prices. Due to these concerns, green coatings have been a new trend in corrosion protection and extensive study is necessary to understand the mechanism of the coatings [1]. Organic coatings are an effective way to protect metallic substrates from corrosion. They are used widely due to low manufacturing costs and

ease of application, suitable bulk properties, versatility and aesthetic attributes. Organic coatings normally consist of a polymeric material, pigments, fillers and various additives [2]. Overall, acrylic resin coatings are the most commonly used anticorrosive coatings because of the excellent weathering and adhesion properties and chemical and abrasion resistance [3].

Henna leaves extract (HLE) has been chosen in this study because it effectively inhibits corrosion for a variety of metals exposed to a wide range of electrolytes. The aqueous extract of the henna leaves, also known as *Lawsonia inermis*, has been tested as a corrosion inhibitor for carbon steel, nickel and zinc in acidic, neutral and alkaline solutions using a polarization technique [4]. *Lawsonia inermis* was also found to have other attributes and may act as an anti-oxidant, anti-inflammatory, anti-cancer and anti-corrosion agent [5–7]. The advantages of *Lawsonia inermis* are attributed to the presence of lawsone (2-hydroxy-1,4-naphthoquinone).

The direct incorporation of an inhibitor into a coating must fulfil a number of requirements; the inhibitor must be effective for the specific metal being protected, the inhibitor shall not damage the barrier properties and cannot be prematurely leached from the coating [8]. In this work, an eco-friendly corrosion inhibitor extracted from henna leaves (HLE) was incorporated in an acrylic resin coating to protect the aluminium alloy 5083 from corrosion. The impact of using HLE in the acrylic resin coating was analysed

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Table 1
Composition of the aluminium alloy 5083 (AA5083).

Element	Mg	Mn	Fe	Si	Zn	Cr	Ti	Cu	Al
Weight percent, wt%	4.5	0.7	0.4	0.4	0.25	0.15	0.15	0.1	Remaining

Table 2
Abbreviations for coatings.

Coating mixture	Abbreviation
Acrylic resin	AC
Acrylic resin + 0.1 wt/vol% HLE	AC1
Acrylic resin + 0.2 wt/vol% HLE	AC2
Acrylic resin + 0.3 wt/vol% HLE	AC3
Acrylic resin + 0.4 wt/vol% HLE	AC4

by X-ray diffraction, differential scanning calorimetry, open circuit potentiometry and electrochemical impedance spectroscopy. The acrylic resin coating with HLE was capable of protecting the aluminium from corrosion and an intensive study should be made to understand its mechanism.

2. Materials and methods

2.1. Surface preparation of aluminium alloy 5083 (AA5083)

The AA5083 samples were prepared as squares with the dimensions 25 mm × 25 mm × 3 mm and polished with emery paper of different grades (600, 900 and 1200). The samples were cleaned with acetone and rinsed with distilled water, dried in the air and stored in desiccators prior to use. The composition of AA5083 is tabulated in Table 1 [9].

2.2. Henna leaves extraction (HLE) process

The fresh henna leaves were dried at room temperature and then crushed into powder. The henna powder was mixed with ethanol and left for one week. Then, the mixture was extracted with a rotary evaporator. The residue left in the flush was incorporated into the acrylic resin coating.

2.3. Incorporation of HLE into the acrylic resin coating

The acrylic resin coating was purchased locally. The concentration of HLE in the acrylic resin coating was from 0.1–0.4 wt/vol%. The mixtures were stirred for 2 h and were coated onto the aluminium alloy using brushes with thicknesses of 25 ± 5 μm. Corrosion tests were performed using seawater collected at Aquatrop Hatchery, Universiti Malaysia Terengganu. To easily identify the coating, the following abbreviations are used as shown in Table 2.

2.4. X-ray Diffraction (XRD)

A Rigaku miniflex X-ray diffractometer (XRD) was used to study the morphology of the acrylic resin coating in the presence and absence of HLE. The XRD measurements were performed at room temperature. Coated specimens were dried at room temperature and cut into small strands before being placed on the sample holder. The samples were directly scanned at 2θ angles between 3° and 80° with X-rays of 1.5406 Å generated by a Cu K_α source.

2.5. Surface tension

A force tensiometer from Biolin Scientific (Sigma 701) was used to study the surface tension of the acrylic resin coating in the absence and presence of HLE. Each test was repeated three times

and the average and the standard deviation were calculated. The measurements were performed at room temperature.

2.6. Differential scanning calorimeter (DSC)

The glass transition temperature (*T_g*) of the coating film was determined with 5–10 mg samples of the dried coating. The heating rate was set to 10 °C/min from 0 to 250 °C.

2.7. Open circuit potential (OCP)

OCP measurements were conducted to investigate the potential of the electrode at various temperatures. The OCP was run for 120 s while the limit *dV/dt* was set to 1 μV/s. The average potential was taken as the OCP of the electrode.

2.8. Electrochemical Impedance Spectroscopy (EIS)

EIS was used to study the impedance and the capacitance of the coated AA5083 in the presence and absence of HLE. The EIS measurements were conducted using an alternating current (AC) with an Autolab PGSTAT302N with respect to the open circuit potential (OCP). All the potentials recorded were relative to a saturated calomel electrode (SCE). The impedance measurements were conducted over a frequency range from 10000 Hz down to 0.01 Hz. The temperature range was 30°–70 °C. The results were analysed using the fitness programme NOVA 1.10. An immersible pump circulated water through the inlet and outlet of the sample holder. The heated water increased the temperature of the seawater in the cell to the desired temperature.

Dielectric analysis used the following equation [10]:

$$\varepsilon_r = \frac{Z_r}{\omega C_0(Z_r^2 + Z_i^2)}$$

where *Z_r* is the real impedance, *Z_i* is the imaginary impedance, *C₀* is the coating capacitance and $\omega=2\pi f$ is the angular frequency in Hz.

2.9. Potentiodynamic Polarization (PP)

Potentiodynamic polarization (PP) is the most common polarization method used for measuring corrosion resistance. The cell used was a conventional three electrode cell with a platinum wire counter electrode (CE) and a saturated calomel electrode (SCE) as the reference electrode (RE). The working electrode (WE) is in the form of a square cut so that the flat surface would be the only surface in the electrode. The potentiodynamic current-potential curves were recorded as the electrode potential was automatically changed from −1.60 V to 0.50 V with the scanning rate of 10 mVs^{−1}. The results were analysed and fitted using the NOVA 1.10 programme.

3. Result and discussion

3.1. X-ray Diffraction (XRD)

The XRD diffractograms of the acrylic coating in the presence and absence of HLE are shown in Fig. 1. At the low position of 2θ approximately 20°, all samples display a hump that indicates the

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