

GREEN SYNTHESIS OF MILD STEEL CORROSION INHIBITOR IN A SALT SOLUTION MADE FROM PEPPER FRUIT (*PIPER NIGRUM* LINN), ARECA FRUIT (*ARECA CATHECU* LINN) AND TEA LEAVES (*CAMMELLIA SINENSIS* L. KUNTZE)

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ABSTRACT

This study aims to synthesize a corrosion inhibitor based on natural materials. The synthesis material of this research consists of extracts of three types of plants which are widely found in the Indonesian environment, namely pepper, areca nut and tea leaves. The extraction of these three basic ingredients is carried out using the maceration method. The extract is used as an inhibitor in mild steel. The functional groups extracted were observed using FTIR (*Fourier Transform Infra Red*), while the microscopic structure of the steel surface given the inhibitor was observed using a metallurgical microscope. The measurement of the corrosion rate was carried out using the mass loss method in 1M NaCl medium. The experimental results showed that the betel nut extract was more efficient as an inhibitor than the tea leaf and pepper extract. The betel nut extract can maximally reduce corrosion with an efficiency of 85.5% and a corrosion rate of 1.65 mpy. However, in general, the corrosion rate of steel decreased as the inhibitor increased, using either betel nut extract, tea leaves or pepper fruit.

KEY WORDS : Corrosion inhibitor, Fruit on, Areca nut, Tea leaves

INTRODUCTION

Generally, metals are easily oxidized so that metal equipment will quickly experience corrosion (damage). In Indonesia, the problem of corrosion needs serious attention, considering that two-thirds of the archipelago consists of oceans and is located in tropical areas with high rainfall, high chloride content, where an environment like this is known to be very corrosive. The environment that causes corrosion is greatly influenced by the presence of waste gases (sulfur dioxide, sulfate, hydrogen sulfide, chloride), O₂ content, solution pH, temperature, humidity, flow rate, and microbial activity (Callister, 1991).

Corrosion is the degradation of metal qualities due to chemical reactions with its environment. Corrosion is a major problem for metal equipment (Smallman, 2007). The impact caused by corrosion

will have a major impact on human life, for example, the economy and the environment. From an economic perspective, corrosion can result in high maintenance costs and production losses in an industry. In addition, corrosion of metals can cause environmental pollution. One way to inhibit the metal corrosion process can be done using an inhibitor. The inhibitor is coated on the metal surface to prevent corrosion reactions with the environment (Maria and Hinton, 2015).

In general, corrosion inhibitors come from organic and inorganic compounds. Inorganic inhibitors are characterized by the presence of a lone pair in their chemical structure and contain nitrite, chromate, and phosphate groups (Reardon, 2011). While imidazoline and amine compounds are examples of organic inhibitors (Francis *et al.*, 2007).

From the research that has been done (Hale, 2012) shows that imidazoline is a very effective inhibitor

of mild steel corrosion in salt water medium. In addition, imadazolin also has high activity as an inhibitor of steel corrosion in acidic medium. In general, organic inhibitors have a nitrogen atom that has a lone pair. Many plants contain compounds that contain nitrogen atoms, including peppers, areca nuts and tea leaves (Schweitzer, 2007).

Peppers, areca nuts, and tea leaves are types of spices and have many health benefits. The content of nitrogen atoms and alkaloid compounds in these spices can be used as reagents against Fe^{2+} ions which form complex compounds. Currently, pepper, areca nut and tea leaves are widely planted in Indonesia, it is proven that Indonesia is one of the largest spice exporting countries in the world. In connection with the above, it is very important to study the use of extracts of pepper, areca nuts and tea leaves as corrosion inhibitors of mild steel in a salt solution. This research will study the corrosion behavior of steel which is widely used as equipment in machinery and industry. Steel material is immersed in a corrosive environment and inhibitor solution in various concentrations

MATERIALS AND METHODS

In general, the materials in this study consisted of experimental metals and inhibitor solutions. The corrosive metal used in this research is mild steel St-41. The inhibitor solution consists of three different natural sources, namely pepper, areca nut and tea leaves. Metal samples are formed into cylinders which are then subjected to grinding (from 240 grit to 2000 grit) and polishing to remove dirt that can hinder the corrosion process.

Preparation of an inhibitor solution from pepper fruit extract (*Piper nigrum* Linn) was carried out through maceration of 700 g of ground pepper using distilled methanol for two weeks. Filtering the filtrate from the sediment was carried out using filter paper, then evaporated using a rotary evaporator at 45 °C and a speed of 60 rpm. This treatment is to separate the extract from the solvent which is then carried out by the drying process. The mother liquor with a concentration of 500 ppm was prepared by dissolving 500 mg of pepper fruit extract in methanol and water with a ratio of 50 ml and 950 ml. Then the inhibitor solution was diluted successively with various concentrations of 50, 100, 150, 200, and 250 ppm.

Preparation of an inhibitor solution from areca nut extract (*Areca Catechu* Linn) is carried out

through maceration of 1 kg of crushed betel nut using a distilled methanol solvent for two weeks. The next step is the same as that for providing an inhibitor made from pepper fruit. Meanwhile, the tea leaf inhibitor was provided by mixing 25 g of dry tea leaves and 20 g of sodium carbonate and 225 ml of boiling water into an erlemeyer flask. Let stand for 7 minutes, then be decanted into another Erlenmeyer flask. To the tea leaves, 50 ml of boiling water is added, then the tea extract is immediately decanted and combined with the previous extract. To extract any remaining caffeine that may be present, boil water containing the tea leaves for 20 minutes, then decant the extract. A solution of 1 M corrosive sodium chloride media is prepared by dissolving 58.5 g of sodium chloride in a 1000 ml erlenmeyer flask. The desired solution of the salt corrosive medium is prepared by diluting the mother liquor 1 M

RESULTS AND DISCUSSION

The results of observations of functional groups in the extracts of pepper, areca nuts and tea leaves using Bruker Tensor 27 are shown in Figure 1. Identification of absorption bands from the FTIR spectrum of Figure 1 is stated in Table 1. These spectra are formed due to the presence of vibration bands and rotational bands with numbers waves between 4000 cm^{-1} - 400 cm^{-1} . The results of the corrosion rate test using the immersion corrosion method and the percent protection for extracts of pepper, areca nut and tea leaves, can be shown in Table 2.

Based on Table 2, it appears that the betel nut extract provides the greatest inhibitor efficiency compared to the inhibitor derived from the extract of pepper fruit and tea leaves. Likewise occurs in the corrosion rate it causes. However, in general it appears that the increase in inhibitor concentration in the three extracts (pepper, areca nut and tea leaves) can reduce the rate of corrosion and increase its efficiency. The smallest corrosion rate of the three types of inhibitors is 1.6547 mpy with an efficiency as an inhibitor of 85.4802 % which occurs at a concentration of 150 ppm of betel nut extract. Meanwhile, for other concentrations and inhibitors from the extracts of pepper and tea leaves, the corrosion rate is still quite large and the inhibition efficiency is still relatively low. This occurs because in this concentration range, the complex compounds formed between the alkaloid compounds contained

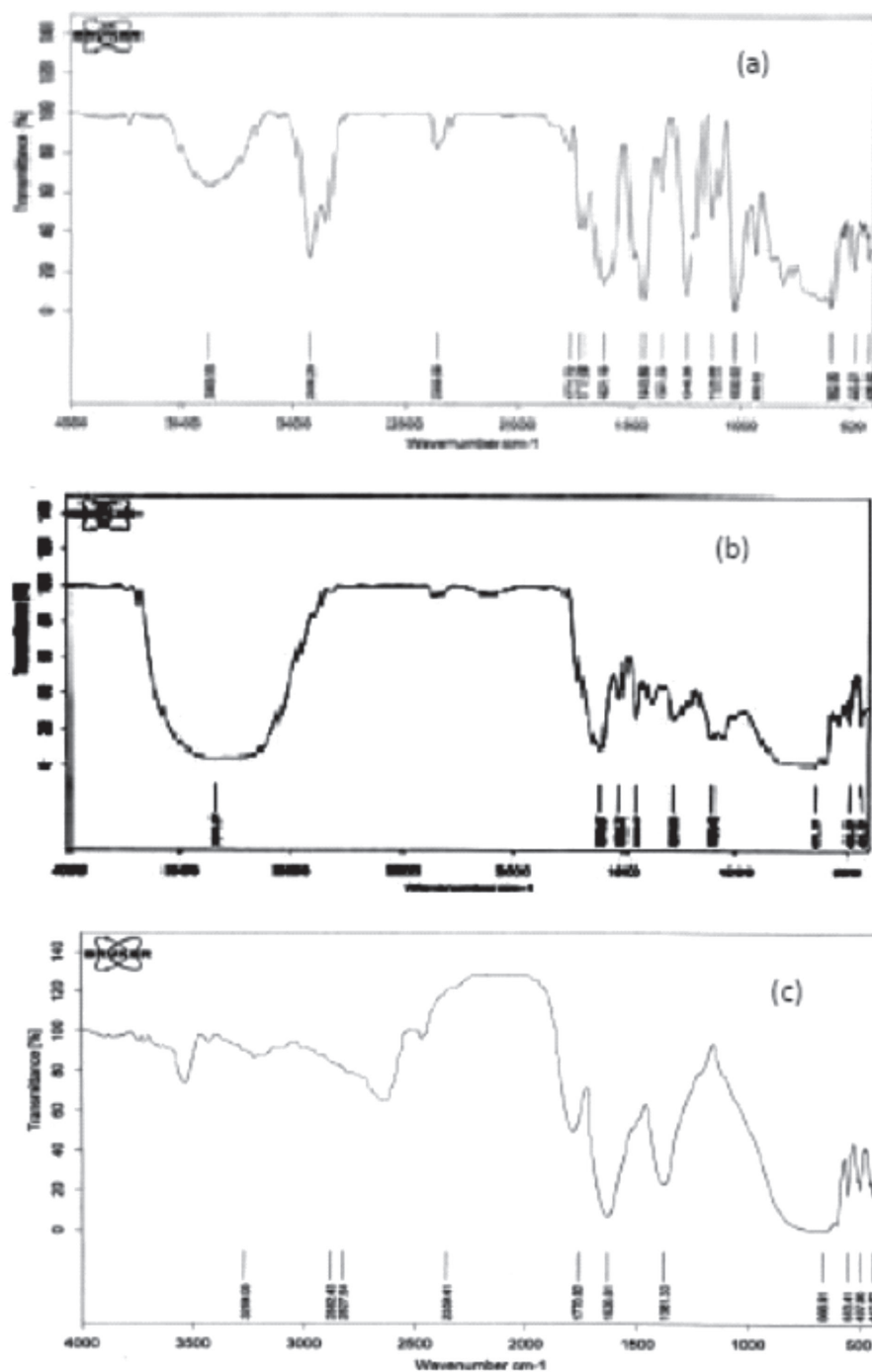


Fig. 1. FTIR spectrum FTIR of (a) pepper fruit, (b) betel nut extract, (c) tea leaves

in the pepper fruit and the Fe^{2+} ion are still not perfect (a little), so that the protective layer formed is still small. Thus, for the prevention of corrosion on mild steel St-41 it is recommended to use betel nut extract with a concentration of 150 ppm.

Alkaloid compounds also have two oxygen atoms which can react with Fe^{2+} ions to form

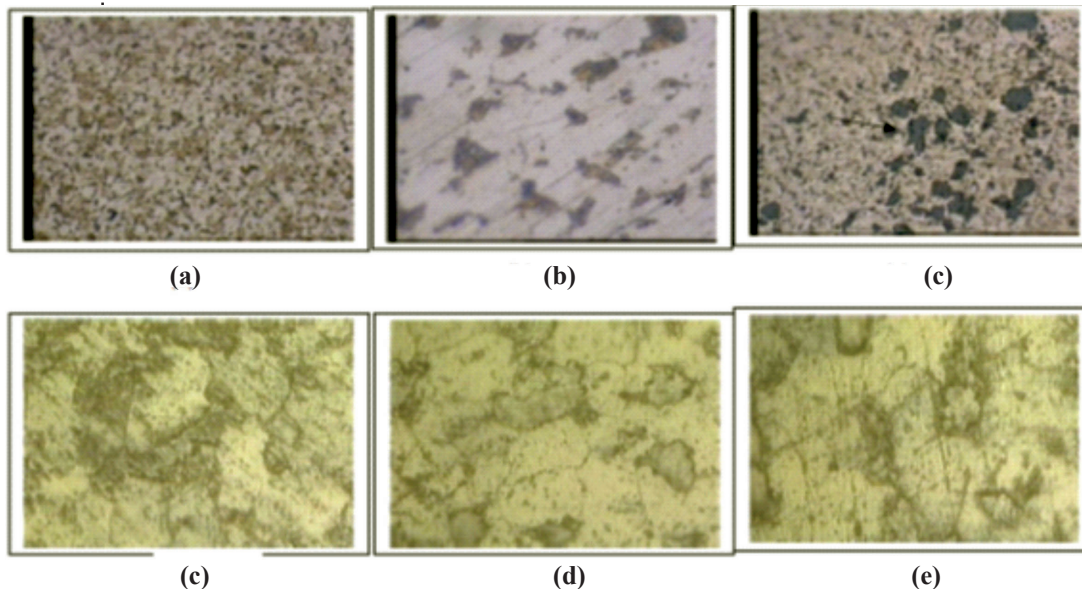
complex compounds. This oxygen atom is in a freer position compared to the oxygen atom contained in piperine and caffeine, so it is easier to react with Fe^{2+} ions to form complex compounds. This is also supported by the results of the microstructure test as presented in Figure 2

Table 1. The result of FTIR analysis

Wave number spectrum (cm ⁻¹)	Variaty of vibration	Wave number (cm ⁻¹)		
		Pepper fruit	Betel nut extract	Tea leaves
1160 – 1030	C - OH	1125.651030.02	1106.40	—
1300 – 1100	C – O	1246.361125.68	1280.961106.40	1106.40
1445 – 1465	C – CH ₂ Scissor	-	1461.921454.01	—
1666 – 1610	C – C	1621.19	1613.07	1636.81
1820 – 1760	C = O	1598.08	1645.071644.06	1770.82
2250 – 2000	C = C	-	2097.392148.25	1645
2850 – 2750	C – H	abel-	-	2827.54
3500 - 3000	N - H	3383.56	3346.57	3269.05

Table 2. Data of corrosion test results for mild steel with and without inhibitor of pepper fruit, betel nut extract and tea leaves.

No	Concentration of inhibitor (ppm)	Pepper fruit		Betel nut extract		Tea leaves	
		Corrosion Rate (mpy)	Efficiency of Inhibitor (%)	Corrosion Rate (mpy)	Efficiency of Inhibitor (%)	Corrosion Rate (mpy)	Efficiency of Inhibitor (%)
1	0	11.3962	0	11.3962	0	11.3962	0
2	50	6.7957	40.3687	3.2705	71.3018	5.3292	53.2370
3	100	6.7119	41.1041	2.6790	76.4922	5.3747	52.8378
4	150	5.8497	48.6697	1.6547	85.4802	5.3328	53.2055
5	200	5.9992	47.3579	2.2811	79.9837	4.9198	56.8295
6	250	5.7478	49.5639	1.9511	82.8794	4.1767	63.3501

**Fig. 2.** (a) and (b) Microstructure of St-41 steel before immersion in media with a magnification of 100x and 500x., (c) without the addition of inhibitor with magnification of 100x, (d) with the addition of a pepper inhibitor with 1000x magnification, (e) with the addition of 1000x magnification of betel nut inhibitor, and (f) with the addition of 1000x enlargement of the tea leaf inhibitor.

CONCLUSION

From the results of testing, observations, and the results and discussions that have been carried out in

this study, several conclusions can be obtained. First, extracts of pepper, areca nuts and tea leaves can be used as a corrosion inhibitor in NaCl corrosion medium. Second, the value of the corrosion rate

tends to decrease as the inhibitor concentration increases. Finally, betel nut extract is the most effective organic inhibitor used as a mild steel corrosion inhibitor in a salt solution rather than an inhibitor of pepper and tea leaf extracts.

REFERENCES

- Callister, Jr. D. William, 1991. *Materials Science and Engineering an Introduction*, second Edition, John Willey and Sons, New York, Hal 281-290.
- Francis A. Carey, Richard J. Sundberg, 2007. *Advanced Organic Chemistry Part A. Structure and Mechanisms* [5th ed.], Springer
- Hale, Arthur James, 2012. *The Synthetic Use of Metals in Organic Chemistry*, Nobu Press
- Maria Forsyth; Bruce Hinton, 2015. *Woodhead Publishing series in metals and surface engineering, no.61 Rare earth-based corrosion inhibitors [1 ed.]*, Publisher. Woodhead Publishing, Elsevier Ltd.
- Reardon, Arthur, C. 2011. *Metallurgy for the Non-metallurgist*, second edition, Publisher Materials Park, Ohio : ASM International
- Schweitzer, P.A. 2007. *Corrosion of Linings and Coatings: Cathodic and Inhibitor Protection and Corrosion Monitoring [Second Edition]*.
- Smallman, R.E. and Ngan, A.H.W. 2007. *Physical Metallurgy and Advanced Materials*, Seventh Edition [7 ed.], Publisher Butterworth-Heinemann.