Influence of Extraction Methods and Solvents on Tannin Content and Performance of Bio Inhibitor In Inhibiting Corrosion Of Seawater Battery Electrodes

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Abstract. This study aims to evaluate the potential of tannin extract from ketapang leaves as a corrosion inhibitor for seawater battery electrodes. The tannin content obtained from Soxhlet extraction, maceration, and ultrasonic-assisted extraction (UAE) using ethanol and acetone solvents at various extraction times was evaluated. The results showed that maceration method using ethanol solvent for a 7-day extraction period produced a tannin content of 1236 ppm. The extract was then tested to determine its effectiveness as a corrosion inhibitor on seawater battery electrodes experiencing power changes due to saturated electrolyte solutions and decomposed electrode layers. In the test, an extract with a concentration of 3% was able to reduce the corrosion rate by 0.00212 mmp and generate a power output of 0.00218 W. These findings provide insights into the use of tannin extract from ketapang leaves as an effective corrosion inhibitor to enhance the performance of seawater batteries.

Keywords: Tannin extract, bio inhibitor, ketapang leaves, corrosion, battery.

1 Introduction

Corrosion in metals is a natural phenomenon that can be mitigated and controlled. It is a degradation process of materials, particularly metals, caused by natural or artificial interactions with the environment [1]. Several commonly used methods to prevent corrosion include metal surface coating, cathodic protection, addition of corrosion inhibitors, among others [2], [3]. Corrosion inhibitors are substances that reduce the reaction rate between the corrosive medium and the metal, thus preventing corrosion [4], [5]. Corrosion inhibitors can be categorized into two types: inorganic inhibitors and organic inhibitors. Inorganic inhibitors such as silicate, borate, tungstate, phosphate, chromate, dichromate, and arsenate have disadvantages including their hazardous nature, cost, and lack of environmental friendliness [6]. On the other hand, organic inhibitors are derived from abundant natural resources. In addition to inhibiting corrosion rates, organic inhibitors are non-toxic, cost-effective, readily available, and renewable [7].

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One interesting example of an organic inhibitor is tannin, a polyphenol compound obtained from plants [8]. Tannins have been used as environmentally friendly corrosion inhibitors, particularly for carbon steel alloys, as they can react with iron ions to form Ferri-tannate complexes that act as a barrier against direct contact between water and iron metal [9]. Tannins can be found in various natural sources, including the leaves of Terminalia catappa Linn, a widely available plant in Indonesia [10]. The leaves of Terminalia catappa contain various compounds such as flavonoids, tannins, saponins, and triterpenoids, while the bark and fruits contain different compounds [11].

In seawater batteries, an electrochemical reaction occurs between the anode and the cathode, where aluminum (Al) is oxidized to copper ions (Al3+) at the anode, and copper ions (Cu2+) are reduced to copper metal (Cu) at the cathode. This electrochemical process produces electrical current and voltage. However, during the electrochemical process, voltage drop occurs at the electrode due to resistance, polarization, and electrode corrosion [12]. Corrosion on the electrode can lead to a reduction in the surface area, which serves as the site for electrochemical reactions, thereby impeding electron flow and decreasing voltage. Therefore, it is crucial to understand and control the corrosion rate, as well as perform appropriate maintenance and care for seawater battery components to ensure efficient operation and optimal power output.

Previous studies have examined the use of guava leaf extracts as corrosion inhibitors for galvanized pipes [13]. However, in this study, we focus on the utilization of Terminalia catappa leaf extracts as organic corrosion inhibitors. We will vary the extraction methods, solvents, and extraction durations to determine the conditions that yield the highest tannin content in the extract. Subsequently, we will immerse the electrodes in Terminalia catappa leaf extract at specific concentrations and observe the power output of the seawater battery over a 14-day period.

Through this research, we aim to contribute to the development of environmentally friendly methods to prevent corrosion on seawater battery electrodes using organic corrosion inhibitors derived from Terminalia catappa leaves. The results of this study are expected to enhance our understanding of tannin utilization as a bio inhibitor for corrosion and provide practical guidelines for selecting the optimal extraction methods, solvents, and durations to yield extracts with the highest tannin content.

In the context of Indonesia's diverse flora, the Terminalia catappa tree represents an intriguing plant to be developed for the utilization of underexploited natural resources [14]. The Terminalia catappa tree grows abundantly throughout Indonesia, especially along coastlines and roadsides. Consequently, Terminalia catappa leaves can be easily obtained as a source of tannin compounds used in this research.

In the process of extracting tannin compounds, the variation in extraction methods becomes an important aspect to investigate. Commonly used extraction methods include Soxhlet extraction, maceration, and ultrasonic-assisted extraction (UAE). The choice of solvent is also crucial since tannin compounds are polar in nature, containing hydroxyl groups. Therefore, polar solvents such as methanol, ethanol, acetone, and water are commonly employed to extract tannins [15]. The polarity level of the solvent should be matched with the extracted compound to achieve optimal extraction results [16]. Additionally, the extraction duration needs to be varied to determine the required time for the extract to contain the highest tannin content.

The extracted results from Terminalia catappa leaves containing tannin compounds will be used as a bio inhibitor for corrosion. The electrodes will be immersed in Terminalia catappa leaf extract at specific concentrations, and the power output of the seawater battery will be observed over a period of 14 days. Through this research, we aim to understand the impact of using Terminalia catappa leaf extract as a corrosion inhibitor on the performance of the seawater battery.

By gaining a better understanding of utilizing tannins from Terminalia catappa leaf extracts as corrosion inhibitors, this research aims to contribute to the development of environmentally friendly methods to prevent corrosion on seawater battery components. Furthermore, the use of organic corrosion inhibitors from natural sources has the potential to reduce dependence on hazardous and expensive chemicals while promoting the optimal utilization of natural resources.

This research holds significant relevance in supporting the development of more efficient and sustainable seawater battery technology. With a better understanding of corrosion inhibition and the use of organic corrosion inhibitors, it is expected to enhance the durability and performance of seawater batteries while reducing losses caused by electrode corrosion.

2 Literature Review

Terminalia catappa L., commonly known as the Indian almond or ketapang tree, is a plant from the combretaceous family that has been widely used in traditional medicine in Southeast Asia. This plant can be found growing wild in almost all regions of Indonesia, particularly along coastlines and roadsides. Despite its wild growth, the ketapang tree (Terminalia catappa Linn) holds high economic value and is extensively utilized by local communities. Ketapang leaves are broadly elliptic in shape with pointed bases and blunt tips. The leaves have parallel veins and wavy leaf margins, and they typically undergo two annual shedding cycles.

The ketapang tree also produces small flowers that cluster into spikes near the branch tips. These flowers have a yellowish-green color and measure approximately 8-25 cm in length. Ketapang leaves (and fruits) contain various natural components, including flavonoids (20-25%), tannins (11-23%), saponins (20%), phytosterols (10-15%), metals (Ca, Mg, Cu, and Zn) (5%), as well as other elements such as sulfur, nitrogen, and phosphorus (20%) [17].

One of the frequent issues encountered with metals is corrosion, which refers to the deterioration of metal quality due to electrochemical reactions with the environment. Corrosion can be described as a galvanic cell, where different surface areas of the metal act as cathodes and anodes. The corrosion mechanism in iron (Fe) can be explained as follows:

$$Fe\,(s) + H_2O\,(l) + \tfrac{1}{2}\,O_2(g) \rightarrow Fe(OH)_2(s) \tag{1}$$

Ferrous hydroxide [Fe(OH)2] formed in this process can naturally oxidize in the presence of water and air, transforming into ferric hydroxide [Fe(OH)3]. The subsequent reaction mechanism is:

$$4\ Fe(OH)_2(s) + O_2\ (g) + 2H_2O(l) \rightarrow 4Fe(OH)_3(s) \tag{2}$$

The resulting ferric hydroxide eventually converts to reddish-brown Fe2O3, commonly known as rust [18]. Corrosion occurs as a consequence of electrochemical reactions with the surrounding environment.

2.1 Tannins

Inhibitors are substances that, when added in small concentrations to an electrolyte, can decrease the corrosion rate. Inhibitors can be considered as catalysts that retard the corrosion process. Generally, corrosion inhibitors are derived from organic and inorganic compounds containing functional groups with free electron pairs, such as nitrite, chromate, phosphate, urea, phenylalanine, imidazoline, and amine compounds. However, these synthetic chemicals are typically hazardous, expensive, and environmentally unfriendly. Consequently, small and medium-sized industries rarely employ inhibitors in their cooling systems, piping systems, and water treatment systems to protect iron/steel from corrosion.

In this regard, the use of safe, readily available, biodegradable, inexpensive, and environmentally friendly inhibitors is highly desirable. Extracted inhibitors from natural sources offer a solution as they are safe, easily obtainable, biodegradable, low-cost, and environmentally friendly [19]. Tannins are polyphenolic aromatic compounds that possess an astringent taste and have the ability to tan hides. Tannins are amorphous, lightweight, have a distinct and highly astringent taste, and can precipitate alkaloids and glycosides from solutions. Tannins are insoluble in benzene, chloroform, petroleum ether esters, carbon tetrachloride, and carbon disulfide [20]. Tannins are present within plant cells in vacuoles, thus not interfering with cellular metabolism. Tannin-containing cells appear brown when observed under a microscope [21]. Tannins are found in almost all parts of plants, including seeds, stems, leaves, and fruit peels. Ethanol extraction is a commonly used method to obtain tannins since tannins are soluble in ethanol. However, the lower the purity of ethanol, the lower the yield of tannin extract obtained. This occurs because the polarity of ethanol increases with increasing water content. Tannins that can undergo hydrolysis will experience it if there is more water in the solvent. Ethanol is the best solvent for extracting tannins compared to methanol, n-hexane, and acetone. It is recommended to use 96% pure ethanol for easier separation of the extraction product [22]. The optimal extraction time for tannins from soursop leaves is 2 hours. At 1 hour of extraction time, the tannin concentration tends to be low. Subsequently, the tannin concentration increases at 2 hours of extraction time but decreases again at 3 hours of extraction [23].

One of the methods to obtain tannins is through distillation processes. Tannic acid resulting from tannin hydrolysis can accelerate the corrosion process by lowering the pH and forming complexes with iron adhered to the surface. The dissolution of anodic carbon iron involves the initial oxidation of Fe to Fe2+. Subsequently, oxygen oxidizes Fe^{2+} to Fe^{3+}. Fe^{3+} is then reduced to Fe^{2+} ions through contact with iron in the pores, leading to a color change [24]. Tannic acid acts on available iron ions through three ways. First, tannin can form complex compounds with Fe^{2+} ions, producing ferro-tannate, which is easily oxidized to ferric tannate in the presence of oxygen. Second, tannin can directly react with Fe^{3+} ions, forming ferric tannate. Third, due to tannin's reducing properties, Fe_2O_3 can be reduced to Fe^{2+} ions [25]. Ferro-tannate can directly oxidize to ferric tannate upon contact with oxygen and water.

3 Materials and Methods

The experiment was conducted using the Soxhlet extraction method and maceration in the process of tannin extraction from Indian almond (Terminalia catappa Linn) leaves. Ethanol and acetone were used as solvents with various extraction times. Quantitative analysis was performed using a UV-V is spectrophotometer to determine the extracted tannin content in each sample. The sample with the highest tannin content underwent a concentration process using a rotary evaporator to remove the polar solvent content from the extract. The extracted Indian almond tannin, which served as a bio-inhibitor, was then used to immerse the electrodes with inhibitor concentrations of 0% (w/v), 3% (w/v), 6% (w/v), and 9% (w/v). The power generated by the seawater battery was observed over a period of 14 days. The corrosion rate on each electrode was determined by conducting weight loss tests. Surface analysis of the samples was performed using a digital microscope to examine the visual appearance of the corroded areas on the electrodes.

4 Result and Discussion

4.1 Soxhlet Extraction Tannin Content

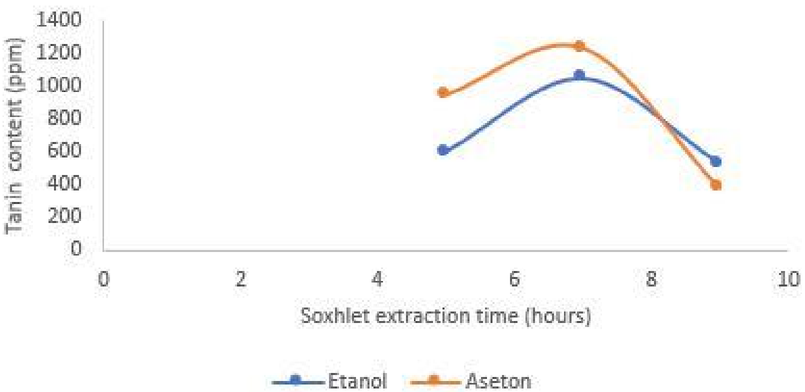

Fig. 1. The relationship tannin content of the extract and the extrusion prayer time (hours)

The test results revealed that the tannin content obtained from Soxhlet extraction using ethanol and acetone exhibited an increase at the extraction time of 2 hours, followed by a decrease at the extraction time of 3 hours [26], which align with Fick's second law of diffusion.

Fick's second law describes the diffusion process of solutes from a solid matrix into a solvent, predicting that a final equilibrium between the solute concentrations in the solid matrix and the solvent will be achieved after a certain time. Consequently, this leads to a deceleration in the extraction results.

In the present study, the increase in tannin content observed at the 2-hour extraction time can be attributed to the diffusion process occurring during extraction. During the extraction, ethanol or acetone acts as a solvent, extracting tannins from the solid matrix. Initially, the concentration of tannins in the solid matrix surpasses that in the solvent, resulting in the movement of solutes from the solid matrix into the solvent and subsequently increasing the tannin content in the solvent.

However, at the 3-hour extraction time, a decline in tannin content was observed. This phenomenon can be explained by the attainment of a final equilibrium between the tannin concentrations in the solid matrix and the solvent after a specific time. At this point, the diffusion process of tannins from the solid matrix into the solvent becomes slower, impeding a rapid increase in tannin content.

These findings validate the utilization of a 2-hour extraction time as the optimal duration in this study. Prolonging the extraction time does not yield a significant enhancement in tannin content and may even lead to a decrease. Thus, to achieve optimal extraction results, a recommended extraction time of 2 hours is advised

4.2 Levels of Maceration Extraction Tannins

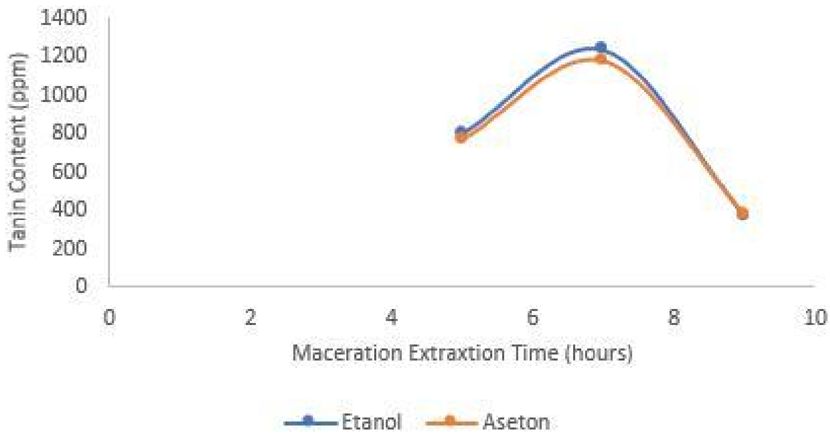

Fig. 2. Maceration extract tannin content

Fig. 2 shows that the tannin content in maceration extraction using ethanol and acetone increased on the 7th day but decreased on the 9th day. This finding suggests that the extraction time affects the tannin content in maceration extraction.

In the process of maceration, the extraction time is an important factor that needs to be considered. The longer the maceration time, the longer the contact between the solvent (ethanol or acetone) and the material being extracted. A longer time allows for more cell breakage and enhances the extraction rate of the active components dissolved in the solvent [27]). The increase in tannin content on the 7th day might be attributed to the greater amount of tannins dissolved in the solvent as a result of the extraction process that occurred during that time period. During the maceration period, tannin compounds dissolve in the solvent and contribute to the overall tannin content.

However, on the 9th day, a decrease in tannin content was observed. This could be due to several factors, such as the saturation of the solvent with tannins or the degradation of tannins over time. These factors may have led to a reduction in the overall tannin content in the maceration extraction.

These findings emphasize the significance of selecting an appropriate extraction time in maceration processes to achieve the desired tannin content. The results suggest that a 7-day extraction time provides an optimal balance between tannin extraction and potential degradation or saturation effects. Further investigations are warranted to explore the specific mechanisms underlying the changes in tannin content during maceration extraction.

4.3 Seawater Battery Power

The following are the results of measuring the electric power of seawater batteries that have been soaked in bio inhibitors with concentrations of 0% (w/v), 3% (w/v), 6% (w/v), and 9% (w/v):

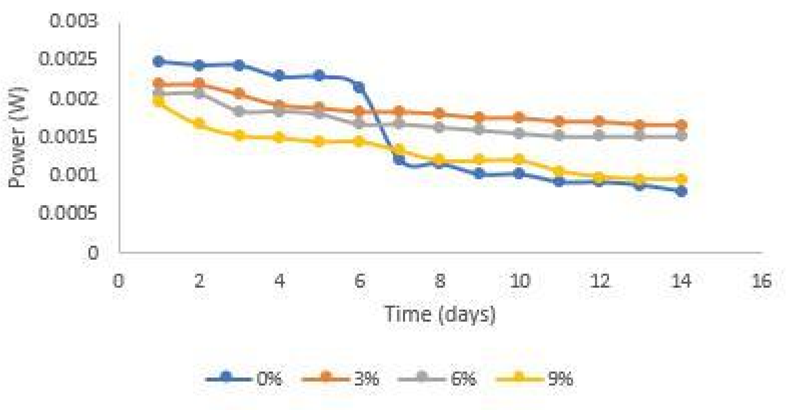

Fig. 3. The results of measuring electric power using various concentrations of inhibitors

Based on Fig. 3, it can be seen that the seawater battery electrolysis process by immersing the electrodes using bio inhibitors at various concentrations (0%, 3%, 6%, and 9% w/v) produces different electrical power.

By immersing the electrodes with a concentration of 0% (w/v), the electric power generated on the first day was 0.00248 W. However, there was a significant decrease in electric power on the 7th day to 0.00119 W. This decrease could be caused by the formation of corrosion on the electrodes on the 7th day, which resulted in a decrease in the generated electric power.

By immersing the electrodes with a concentration of 3% (w/v), the electric power generated on the first day was 0.00218 W. Even though it decreased to 0.00205 W on the third day, the electric power generated tended to be more stable compared to to the electrodes. without bio-inhibitors. This shows that the bio-inhibitor with a concentration of 3% (w/v) forms a barrier layer on the copper and aluminum electrodes, which inhibits the corrosion process and affects the electrical power generated.

By immersing the electrodes with a concentration of 6% (w/v), the electric power generated on the first day was 0.00205 W. Although it decreased to 0.00183 W on the 3rd day, the electric power generated also tended to be more stable compared to to electrodes without bio inhibitors. The concentration of electrode immersion with bio inhibitors affected the formation of a barrier layer on the electrodes, and at a concentration of 6% (w/v) the barrier layer formed tended to be more effective than at a concentration of 3% (w/ v).

Based on the results presented in Fig.3 it is evident that the immersion of electrodes using bio inhibitors at various concentrations (0%, 3%, 6%, and 9% w/v) in the seawater battery electrolysis process leads to different levels of electrical power generation.

When the electrodes were immersed with a concentration of 0% (w/v), the electric power generated on the first day was 0.00248 W. However, there was a significant decrease in electric power on the 7th day, resulting in a value of 0.00119 W. This decline can be attributed to the formation of corrosion on the electrodes, which ultimately led to a reduction in the electric power generated.

In contrast, when the electrodes were immersed with a concentration of 3% (w/v), the electric power generated on the first day was 0.00218 W. Although there was a slight decrease to 0.00205 W on the third day, the electric power generated remained relatively stable compared to the electrodes without bio inhibitors. This observation suggests that the bio inhibitor at a concentration of 3% (w/v) forms a protective barrier layer on the copper and aluminum electrodes, effectively inhibiting the corrosion process and positively influencing the electrical power generated.

Similarly, when the electrodes were immersed with a concentration of 6% (w/v), the electric power generated on the first day was 0.00205 W. Despite a decrease to 0.00183 W on the 3rd day, the electric power generated continued to exhibit stability compared to the electrode without a bio inhibitor. The concentration of the bio inhibitor used for electrode immersion affected the formation of a barrier layer on the electrodes, and at a concentration of 6% (w/v), the barrier layer formed showed a tendency to be more effective than at a concentration of 3% (w/v).

These findings highlight the significance of using bio inhibitors to protect the electrodes from corrosion and maximize the electrical power generated in the seawater battery electrolysis process. The concentration of the bio inhibitor plays a crucial role in the formation of an effective barrier layer, which directly influences the stability and efficiency of the power generation. Further investigations are warranted to optimize the concentration of the bio inhibitor and explore its long-term effects on the performance of the seawater battery system.

5 Conclusion

- The tannin content in the Soxhlet extraction using ethanol and acetone increased during a 2-hour period but decreased after 3 hours. The optimal extraction time was determined to be 2 hours, as longer durations did not yield significant increases. Extraction time was found to impact the tannin content in maseration extraction. An increase was observed on the 7th day, followed by a decrease on the 9th day. Therefore, the optimal extraction time is approximately 7 days.
- The electrolysis process of seawater batteries, involving electrode immersion with bio inhibitors at various concentrations (0%, 3%, 6%, and 9% w/v), resulted in varying levels of electrical power generation. The concentration of the bio inhibitor influenced the formation of a barrier layer on the electrodes. At concentrations of 3% and 6%, the barrier layer tended to be more stable. However, at a concentration of 9%, the formed barrier layer became thicker but less effective in preventing corrosion. These findings highlight the importance of determining the optimal concentration of the bio inhibitor to protect the electrodes and maximize electrical power generation.

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