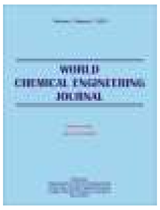
World Chemical Engineering Journal Vol.7, No.1, (2023), pp. 11 – 17

WORLD CHEMICAL ENGINEERING JOURNAL

Journal homepage: http://jurnal.untirta.ac.id/index.php/WCEJ



Synergistic Ability of Tannin – Silica as a Corrosion Inhibitor with the Addition of KI to Mild Steel in Demineralized Water

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ARTICLE HISTORY

Received 21 June 2023
Received in the revised form 28 June 2023
Accepted 29 June 2023
Available online 30 June 2023

ABSTRACT

The rapid industrial growth in Indonesia today requires effective and efficient operating and maintenance conditions. The application of corrosion protection in cooling towers and boilers shows that inhibitor materials such as nitrates, nitrites and chromates must be used in large concentrations; these inhibitors also cause environmental pollution. For this reason, bio-inhibitors derived from natural ingredients have begun to be developed. Tannins and silica are widely known as effective inhibitors in controlling carbon steel corrosion. The synergistic mechanism of inhibitors is known to increase the effectiveness of inhibitory substances. This study aims to study the effectiveness and understand the mechanism of tannin and silica inhibitors and the effect of adding potassium iodide. as a synergistic inhibitor. This study used samples of mild steel carbon steel with demineralized water media at a temperature variation of 30-60°C and immersion time of 1, 2 and 3 hours. Variations in the concentration of tannins and silica used are 0-1250 ppm, and the corrosion measurement method uses the weight loss method. The results showed that adding potassium iodide. They have increased the ability of mild steel corrosion inhibitors produced by tannin-silica synergy. The corrosion rate of mild steel can be reduced by 82.95% at a concentration of tannin-silica-KI 1250 ppm. Therefore, tannin-silica synergy with the addition of potassium iodide. It has the potential to be a good alternative in the use of corrosion inhibitors for mild steel in demineralized water.

Keywords: *tannin, silica, synergistic, inhibitor, potassium iodide*

1. INTRODUCTION

The development of the Indonesian industry in the last decade has experienced significant effects (Deny, 2019) besides being ranked 5th in the manufacturing industry of the G-20 countries. Indonesia was also removed from developing country status by the US Trade Representative Office (USTR) (Kembaren, 2020). This development is inseparable from the rapid growth of the industry in Indonesia.

This rapid growth requires that every operation and maintenance of the manufacturing industry is carried out in the best and most efficient way possible. One of the main problems plaguing the process of advanced industries is metal corrosion. Even though corrosion is a natural event for every metal, it still causes losses in every sector, both economically and safety-wise. The Saudi Aramco Technology Journal 2006 published research on the cost of corrosion prevention in the oil and refining industries. It is known that the cost of overcoming corrosion in the onshore oil industry is 17-28% of maintenance costs. In the offshore industry, these costs can swell even more, namely 60-70% of maintenance costs (Aramco, 2019).

To maintain the continuity of the industrial process, maintaining operating equipment is of particular concern to avoid the danger of corrosion. Many methods can be used to inhibit corrosion, including adding inhibitors. The National Association of Corrosion Engineers defines an inhibitor as a substance that slows down the corrosion rate when added to an environment in small concentrations (Nathan, 1973).

In cooling towers and boilers applications, an anodic (passivation) inhibitor is used to inhibit the anodic reaction. The mechanism used is to change the properties of the metal surface to be passive. Anodic inhibitors can be divided into oxidizing and non-oxidizing ions based on the need for oxygen ions. Oxidizing ions form a protective barrier on the metal devoid of oxygen ions. Examples are nitrate, chromate, and nitrite-based inhibitors. In comparison, non-oxidizing ions form a passive layer at the anode by involving hydrogen ions. Examples are phosphate, tungsten, and molybdenite inhibitors.

This inhibitor type is also used in recirculation-cooling systems, rectifiers, cooling towers and boilers. However, this anodic inhibitor has weaknesses in its application, apart from the content of nitrate, chromate, and other heavy metals, which are not environmentally friendly. This inhibitor material must be continuously maintained in the fluid concentration because decreasing the inhibitor content from the required limit will make corrosion occur faster. The most common type of corrosion that occurs is pitting corrosion (Putra, 2011). On this basis, it is necessary to carry out further research on replacement inhibitor materials in cooling tower systems.

Inhibitors work by forming a layer on the metal surface to protect the metal from corrosion attacks. The corrosion rate will decrease with environmental inhibitors (Prayogi, 2017). Corrosion inhibitors generally come from organic and inorganic compounds containing groups with lone pairs of electrons, such as nitrites, chromates, phosphates, urea phenylalanine, imidazoline, and amine compounds. However, synthetic chemicals are toxic and not environmentally friendly. Therefore, further research is needed to explore environmentally friendly corrosion inhibitors, for example, natural product inhibitors (Pramudita et al., 2018a).

Most of the currently developed inhibitors are bio-inhibitors or organic inhibitors, which are taken from plant extracts. Flavonoids, terpenes, and tannins are widely used bio-inhibitors (Rochmat et al., 2019b).

Tannins are organic compounds that are widely used as antioxidants or natural inhibitors. Tannin compounds can be obtained from extracts of Terminalia catappa leaves, rice grain, and other plants (Alaneme et al., 2015; Pramudita et al., 2018a). Tannins have inhibitory properties because of their ability to form complex compounds with metals. Tannins with hydroxy groups (-OH) are natural polyphenolic compounds widely contained in various types of plants in Indonesia. The ability of polyphenolic compounds to form enormous structures can potentially absorb heavy metals, transition metals, and even uranium (Rochmat et al., 2019b).

Another inhibitor material that is widely used is silica material. Pramudita has researched the inhibition of silica materials by taking silica extract from rice husks. In comparison, the addition of inorganic silica as an inhibitor synergized with phosphonates was carried out in 2007 (Salasi et al., 2007). The adhesion properties of silica provide an excellent barrier to the metal from ions in solution and oxygen, thereby protecting the metal from corrosion. Silica has good thermal stability and chemical resistance, making it an excellent anodic inhibitor (Pramudita et al., 2018b).

The synergy between tannins and silica in corrosion inhibitors ensures the maximum use of inhibitor materials in all operating conditions. Tannins have been proven to work effectively at low temperatures, while silica has optimal performance at high temperatures. Therefore, the synergy between tannins and silica is expected to be optimized by combining the two ingredients in every condition. In addition, adding KI (halide ions) can improve the performance of corrosion inhibitors because potassium iodide (KI) can accelerate the inhibitor reaction and increase corrosion resistance. Thus, the synergy between tannins and silica with the addition of KI is expected to produce effective and efficient corrosion inhibitors in various operating conditions.

2. METHODS

2.1 Sample Preparation

In this study, the samples used were mild steel samples with demineralized water corrosion media. Mild steel samples were prepared with dimensions of 20 x 30x10 mm for each sample and then sanded using a grinder and sandpaper of various sizes. From 60#, 80#, to 120# and 200# to show the shine on the metal surface. The mass of the sample before and after testing is weighed and recorded for the weight loss method.

2.2 Preparation of Inhibitors

Corrosive media (demineralized water) and inhibitors were prepared by dissolving the inhibitor in demineralized water. Concentration variations were made at 0, 250, 500, 750, 1000, and 1250 ppm for each inhibitor, namely tannin, silica, and tannin-silica-KI. A particular concentration of tannin-silica-KI solution is prepared by mixing the three inhibitor ingredients with the same concentration.

2.3 Immersion Process in Demineralized Water

Samples were immersed in demineralized water corrosive media. Other samples were given additional inhibitors in the following order: (1) Tannins, (2) Tannins + KI, (3) Silica, (4) Silica + Tannins. Various variations of temperature and immersion time were made in each inhibitor variable. Temperature variations were observed in each inhibitor medium and samples without inhibitors at 30, 40, 50, and 60 °C. Mild steel samples were also prepared for variations in immersion time at each immersion time. Namely: 1, 2, 3, and 4 hours.

2.4. Corrosion Measurement

Measurement of corrosion in this study is using the weight loss method. The weight loss method measures the corrosion rate of metal materials by measuring the corroded material's weight after exposure to a corrosive environment for a particular time.

First, prepare a sample of the metal to be tested in this corrosion measurement. Clean the sample's surface until it is clean from dirt or other substances that may affect the size. Weigh the sample using an analytical balance with an accuracy of 0.001 gram and record the initial weight of the sample.

Next, dip the sample into the corrosive medium that has been prepared and leave it for the specified time. After a particular time, remove the sample from the corrosive medium, clean it of any corrosion film that may have formed on its surface, and dry it. Weigh the sample again using the same balance, and record the final sample weight. The corrosion rate is calculated using Equation 1, and the inhibition efficiency is calculated using Equation 2.

$$Cr = (87500 * \Delta W)/A\rho t \qquad (1)$$

Where Cr is corrosion rate (mmpy), ΔW is the difference between the initial weight and the final weight (g), A is the surface area (cm^2), ρ is the density of mild steel (g/cm^3), and t is a difference in immersion time (h)

$$\boldsymbol{I.E\% = \left(1 - \frac{Cr_{inh}}{Cr_{blank}}\right) x\ 100} \qquad (2)$$

Where Cr_{inh} and Cr_{blank} are the corrosion rate with and without the inhibitor, I.E% is the percentage of inhibition efficiency.

3. RESULT AND DISCUSSION

3.1. Effect of tannin addition on efficiency inhibition

Tannins are organic polyphenols compounds and can act as corrosion inhibitors on carbon steel. Characteristics of tannins include water solubility, easy oxidation by oxygen, and easy colour change when exposed to UV light. This study used tannins with concentrations of 500, 750, 1000 and 1250 ppm in research.

The effect of variations in immersion temperature and immersion time on the efficiency of corrosion inhibition of carbon steel was measured under the addition of tannin. Carbon steel samples measuring 20x30 mm and 3 mm thick were immersed in a tannin solution and demineralized with variations in temperature of 30, 40, 50 and 60 ^{0}C and variations in immersion time of 1, 2 and 3 hours.

The highest corrosion inhibition efficiency occurred at a tannin concentration of 1000 ppm, immersion temperature of 40 °C and immersion time of 3 hours, with an inhibition efficiency of 83.75%. Meanwhile, the lowest corrosion inhibition efficiency was at a tannin concentration of 250 ppm, immersion temperature of 30 Celsius and immersion time of 1 hour with an inhibition efficiency of 4.00%.

Based on the data that has been presented, the addition of tannins to demineralized water can increase the corrosion inhibition efficiency of mild steel. The protective mechanism occurs due to a reaction between Fe2+ and tannins, producing complex compounds. The mechanism can be shown as follows:

$$Fe \longrightarrow Fe^{2+} + 2e$$
$$Fe^{2+} + 2e \longrightarrow Fe$$
$$Fe^{2+} + \text{tannin} \longrightarrow [Fe(\text{tannin compound})]^{2+}$$

The results showed that the highest inhibition efficiency was achieved at a tannin concentration of 1000 ppm, immersion at 40°C, and immersion time for 3 hours with an inhibition efficiency of 83.75%.

However, the inhibition efficiency at a tannin concentration of 250 ppm, immersion at 30°C, and immersion time for 1 hour were very low, namely only 4.00%. It shows that adding tannin as a corrosion inhibitor has optimal limits at a specific temperature and immersion time.

In previous studies, it has been known that temperature affects the effectiveness of corrosion inhibitors, where the effectiveness of inhibitors decreases at higher temperatures. It can be explained because the corrosion reaction rate will increase at higher temperatures, and the inhibitor molecule will be more easily separated from the metal surface so that its effectiveness decreases. Therefore, higher concentrations of inhibitors are required at high temperatures to achieve the same inhibition efficiency at lower temperatures. (Behpour et al., 2012; Pramudita et al., 2020).

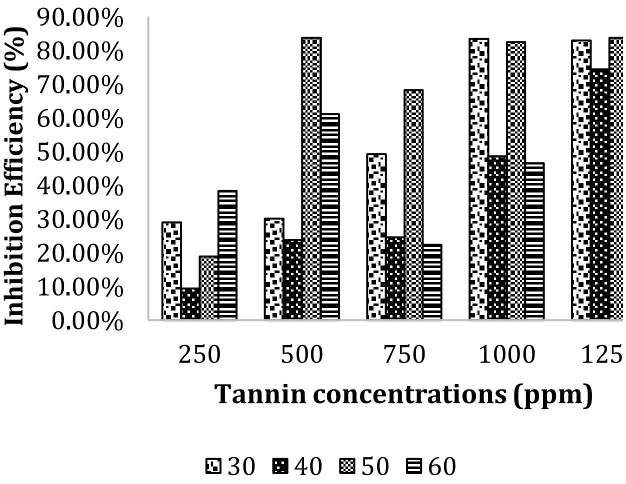

Fig. 1. Effect of tannin concentration (ppm), temperature (^{0}C), and inhibition efficiency (%)

Figure 1 shows the effect of varying tannin concentrations on the efficiency of tannin inhibition. The decrease in inhibition efficiency at lower tannin concentrations is probably due to insufficient inhibitors to protect the mild steel surface optimally. In comparison, the increase in inhibition efficiency at higher tannin concentrations may be due to enough inhibitors to form a better protective layer on the mild steel surface.

However, remember that using very high concentrations of tannins can also cause damage to metal surfaces. Therefore, selecting the right concentration of tannins is very important to achieve optimal inhibition efficiency and prevent damage to the metal surface. In this case, it can be concluded that the tannin concentration significantly affects the inhibition efficiency of mild steel in demineralized water, and the proper concentration needs to be selected to achieve optimal protection.

3.2. Effect of silica addition on efficiency inhibition

Figure 2 shows adding silica to demineralized water effectively reduces the corrosion rate of mild steel. Based on the data obtained, the highest inhibitor efficiency occurred in immersion with the addition of 1000 ppm silica, the temperature of 60 ^{0}C, and an immersion time of 2 hours, with an efficiency of 94.65%. In contrast, the lowest inhibitor efficiency occurred in immersion with the addition of 500 ppm silica, a temperature of 30 ^{o}C, and an immersion time of 3 hours, with an efficiency of only 0.85%.

Several factors can cause the difference in inhibitor efficiency in each sample. First, the concentration of silica used in the test can affect the efficiency of the inhibitor. This study's most excellent inhibitor efficiency occurred at 1000 ppm silica concentration. It may be because higher silica concentrations can provide more silica particles suspended in water and adhere to metal surfaces, forming a better protective layer.

Second, immersion temperature and time can also affect the efficiency of the inhibitor. In this test, the highest inhibitor efficiency occurred at 60 ^{0}C and 2 hours of immersion time. It may be because at higher temperatures, the chemical reaction between the silica and the metal surface occurs more rapidly, and the longer immersion time provides a longer time for the silica to form a more robust protective layer.

In conclusion, the test results show that adding silica to demineralized water can effectively reduce the corrosion rate of mild steel. Using a silica concentration of 1000 ppm at 60 ^{0}C and an immersion time of 2 hours produces the highest inhibitor efficiency. It can be a good choice for use in industry to protect metals from corrosion.

Several studies show the effect of temperature on the effectiveness of silica inhibitors in preventing corrosion. Several studies have shown that the effectiveness of silica inhibitors at high temperatures is lower than at low temperatures. However, research also indicates silica inhibitors can be effective at high temperatures. (Pramudita et al., 2018b; Rochmat et al., 2019a).

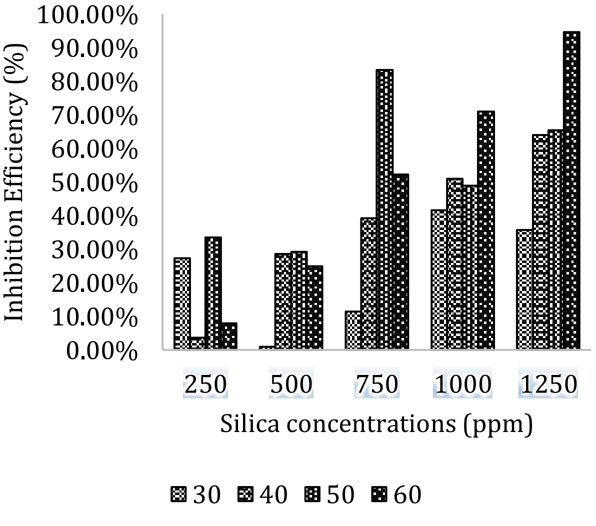

Fig. 2. Effect of silica concentration (ppm), temperature (^{0}C), and inhibition efficiency (%)

One of the studies discussing the effect of temperature on silica inhibitors was conducted by Rahman et al. (2014). This study tested the effectiveness of silica inhibitors in preventing corrosion of mild steel at 30°C and 60°C. The results showed that the effectiveness of silica inhibitors at 30°C was higher than at 60°C. Another study conducted by Togun et al. (2019) also showed similar results, where the effectiveness of silica inhibitors decreased at higher temperatures. (Awizar, Othman, Jalar, Daud, et al., 2013; Salasi et al., 2007)

However, studies also show silica inhibitors can be effective at high temperatures. Research conducted by Ávila-Gonzalez et al. (2011) tested the effectiveness of silica inhibitors at temperatures between 25°C and 80°C. The results showed that silica inhibitors were still effective at high temperatures and could provide corrosion protection for mild steel. (Ávila-Gonzalez et al., 2011)

In general, the effect of temperature on the effectiveness of silica inhibitors in preventing corrosion is still controversial and needs further research. This research shows that the efficiency of silica still increases with an increase in temperature up to 60 ^{0}C.

3.3. The Effect of the Combination of Tannin, Silica, and KI as a Synergistic Inhibitor

In this study, immersion was carried out at various concentrations of tannin-silica-KI 250, 500, 750, 1000, and 1250 ppm with temperature variations of 30, 40, 50, and 60 ^{0}C and immersion times of 1, 2, and 3 hours. Figure 3 shows the inhibitor efficiency of each sample, with the highest efficiency reaching 95.75% at an immersion temperature of 50 ^{0}C, immersion time of 2 hours, and the addition of tannin-silica-KI in 1250 ppm demineralized water. The lowest efficiency reached 0.51% at 30 ^{0}C immersion temperature, 1-hour immersion time, and adding tannin-silica-KI in 500 ppm demineralized water.

In this study, the synergistic effect between tannins, silica, and KI provided a higher inhibition efficiency than a single inhibitor. The results of this study indicate that the addition of KI can increase the efficiency of synergistic inhibitors of tannins and silica. In addition, the efficiency of the inhibitor is also affected by temperature and immersion time. Synergistic inhibitor efficiency increases at high temperatures, where at 60 ^{0}C, the inhibitor efficiency reaches 92.33% when added tannin-silica-KI in 1000 ppm demineralized water and 2 hours of immersion time.

The increased efficiency of synergistic inhibitors at high temperatures can be explained by the mechanism of chemical reaction acceleration at high temperatures, which increases corrosion activity, so the inhibitors' effect is also required at a higher level. In addition, using tannins and silica together provides a better protective layer on the steel surface and increases the efficiency of the inhibitor. Other studies have also shown that using a combination of silica and organic inhibitors can increase the efficiency of inhibitors at high temperatures. (Hao et al., 2017; Pramudita et al., 2019)

This study's results indicate that using a combination of tannins, silica, and KI as synergistic inhibitors can provide higher inhibition efficiency for mild steel in demineralized water, especially at high temperatures. Therefore, this synergistic inhibitor can be a more effective alternative in inhibiting steel corrosion.

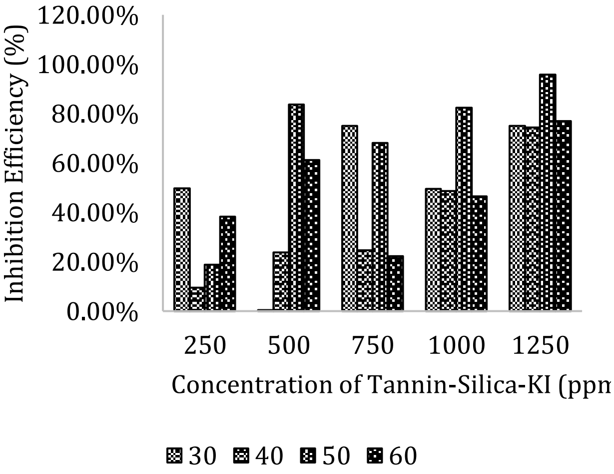

Fig. 3. Synergistic of Tannin-Silica-KI on Variation of Concentration (ppm), temperature (^{0}C), and inhibition efficiency (%)

The mechanism of inhibitor synergism in this study is thought to involve several effects, including increased protective power from interactions between different inhibitor molecules, increased solubility and spread of inhibitors in the broader medium, and reduced electrochemical activity of metals associated with combined inhibitors.

Research conducted by Hao et al. (2017) explained that the synergistic effect of inhibitors could occur due to several inhibitor molecules with different mechanisms that complement each other in protecting metals. Tannins, for example, form an adsorption layer on the metal surface that protects the metal from corrosion. Meanwhile, silica and KI work with physical and chemical inhibition mechanisms, in which silica forms a passive film that protects the metal from corrosion. At the same time, KI can absorb metal surfaces and form a thin layer with iodide ions, forming bonds with the metal. These three molecules complement each other in protecting the metal, thereby providing a higher synergistic effect than using each inhibitor separately.

4. CONCLUSION

Based on the results of this study, it can be concluded that using corrosion inhibitors separately or in combination can increase the efficiency of inhibition in mild steel. From the test results, it was found that the addition of tannin as a corrosion inhibitor was able to inhibit corrosion by forming a thin layer on the mild steel surface. In contrast, silica, a corrosion inhibitor in demineralized water, had high inhibition efficiency, especially at higher temperatures. While adding tannins, silica and KI as synergistic inhibitors provides a synergistic effect on inhibition efficiency and higher inhibition efficiency than using tannins or silica separately. It shows that the combination of inhibitors can increase the effectiveness of inhibiting the corrosion process on mild steel.

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