DOI: https://doi.org/10.33479/sakti.v5i1.151

Jurnal Sains dan Aplikasi Keilmuan Teknik Industri (SAKTI)
Journal of Industrial Engineering: Application and Research

Volume 05 No. 01 – June 2025
Journal homepage: www.sakti.machung.ac.id
ISSN: 2829-8519 (print) – ISSN: 2829-8748 (online)

Eco-friendly Corrosion Inhibitor Coatings Based on Plant Waste Extracts: A Comparative Study of Tea Waste and Orange Peel

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Received 10 April 2025 / Revised 28 May 2025 / Accepted 17 June 2025 / Published 30 June 2025

Abstract. This study seeks to assess the efficacy of plant-based waste extracts, namely orange peel extract and tea waste extract, as natural corrosion inhibitors for mild steel submerged in a 3.5% NaCl solution. The methodology adhered to ASTM G31 standards, employing mild steel specimens coated with each extract using three application techniques: brushing, immersion, and spraying. The weight loss method was used to measure corrosion rates (Mmpy), enabling the calculation of inhibition efficiency (IE%) for each specimen. The uncoated control specimen consistently exhibited the highest corrosion rate (3.4523 Mmpy), categorized as "Poor." Specimens treated with orange peel extract showed significantly lower corrosion rates, with the brushing method yielding an inhibition efficiency of 98.7%, categorized as "Excellent." Likewise, specimens coated with tea waste extract demonstrated remarkable inhibition, with the brushing and immersion techniques achieving efficiencies of 98.9% and 97.7%, respectively. These results indicate that both orange peel and tea waste extracts serve as promising, eco-friendly alternatives for corrosion protection of mild steel, with their effectiveness significantly influenced by the method of application.

Keywords: Corrosion rate; Inhibitor; Orange peel; Tea waste.

1. Introduction

Corrosion represents a widespread degradation process that impacts metallic materials via electrochemical interactions with their surrounding environment, including moisture, saline water, and aggressive chemical agents (Omran & Salam, 2020). In addition to physical degradation, corrosion presents considerable economic and safety issues, leading to notable financial setbacks in sectors dependent on metal structures, such as construction, transportation, and manufacturing. Global estimates indicate that the costs associated with corrosion reach billions of dollars each year, largely stemming from maintenance, component replacement, and infrastructure failures (Mazumder, 2020). Consequently, prioritizing the development of effective strategies for corrosion mitigation is essential to improve material durability and guarantee structural safety.

Surface coating technologies that include corrosion inhibitors are some of the most commonly utilized strategies for safeguarding metals from corrosive environments. These inhibitors operate by slowing down electrochemical reactions, usually by creating passive, protective films on metal surfaces (Bouoidina et al., 2021). While they are effective, many traditional corrosion inhibitors originate from synthetic compounds, including chromates, phosphates, and nitrites. The compounds in question present considerable risks to both the environment and human health, encompassing issues such as toxicity, bioaccumulation, and the contamination of soil and water resources (Al-Amiery et al., 2024). As a result, there is a pressing need for sustainable alternatives that provide similar protective capabilities while minimizing negative environmental effects.

In recent years, the use of plant-derived extracts as environmentally friendly corrosion inhibitors has surfaced as a promising direction. Plants inherently possess a variety of bioactive compounds: like flavonoids, tannins, saponins, and alkaloids, that can adhere to metal surfaces and create protective barriers, thus reducing corrosion processes (Salleh et al., 2021). The advantages of these natural materials are noteworthy, encompassing high biodegradability, low toxicity, renewability, and broad availability, which are in harmony with the principles of green chemistry and sustainable development. Furthermore, utilizing agricultural by-products as corrosion inhibitors plays a significant role in waste valorization within the context of a circular economy (Mandal et al., 2024).

Plant-based corrosion inhibitors typically function through the adsorption of organic compounds onto the metal surface. This process may proceed via physisorption, driven by weak Van der Waals forces, or chemisorption, involving the formation of coordinate or covalent bonds between functional groups, such as hydroxyls or carbonyls and the metal substrate. The adsorption behavior can often be modeled using isotherm theories such as the Langmuir and Temkin models, which help describe the nature and strength of inhibitor with metal interactions (Popoola, 2019). Such theoretical frameworks enhance the understanding of inhibition mechanisms and support the interpretation of experimental results, particularly for natural compounds like tannins and flavonoids.

This study explores the viability of orange peel extract as an eco-friendly corrosion inhibitor for metal coating applications. Orange peel, often overlooked as agro-industrial waste, possess a wealth of phenolic and flavonoid compounds that are recognized for their antioxidant and metal-chelating capabilities (Ferlazzo et al., 2016). This study investigates the effectiveness of various application methods, including spraying, brushing, and immersion, in creating a protective layer on metallic substrates. It also assesses the optimal concentration needed to achieve maximum inhibition efficiency.

The advancement of anti-corrosion coatings derived from plant extracts presents a twofold advantage: improving the corrosion resistance of metal materials and reducing the environmental hazards linked to synthetic chemicals. The development of corrosion-resistant coating technology based on plant extracts can provide an effective and environmentally friendly alternative to conventional corrosion inhibitors. The results of this study aim to enhance the development of eco-friendly corrosion inhibition technologies, fostering safer and more sustainable approaches in materials protection and industrial applications.

Table 1 Corrosion rate and inhibition efficiency of specimens coated with tea extract

Specimen	ΔW (g)	Corrosion Rate (Mmpy)	Inhibition Efficiency (%)
Specimen 1 (Control)	0.1188	3.4523^{a}	-
Specimen 2 (Brush)	0.0013	0.0377^{c}	98.9%
Specimen 3 (Immersion)	0.0027	0.0784^{c}	97.7%
Specimen 4 (Spray)	0.0113	0.3283^{b}	90.4%

*Values with different superscripts within the same column are statistically significantly different ($p<0.05$).

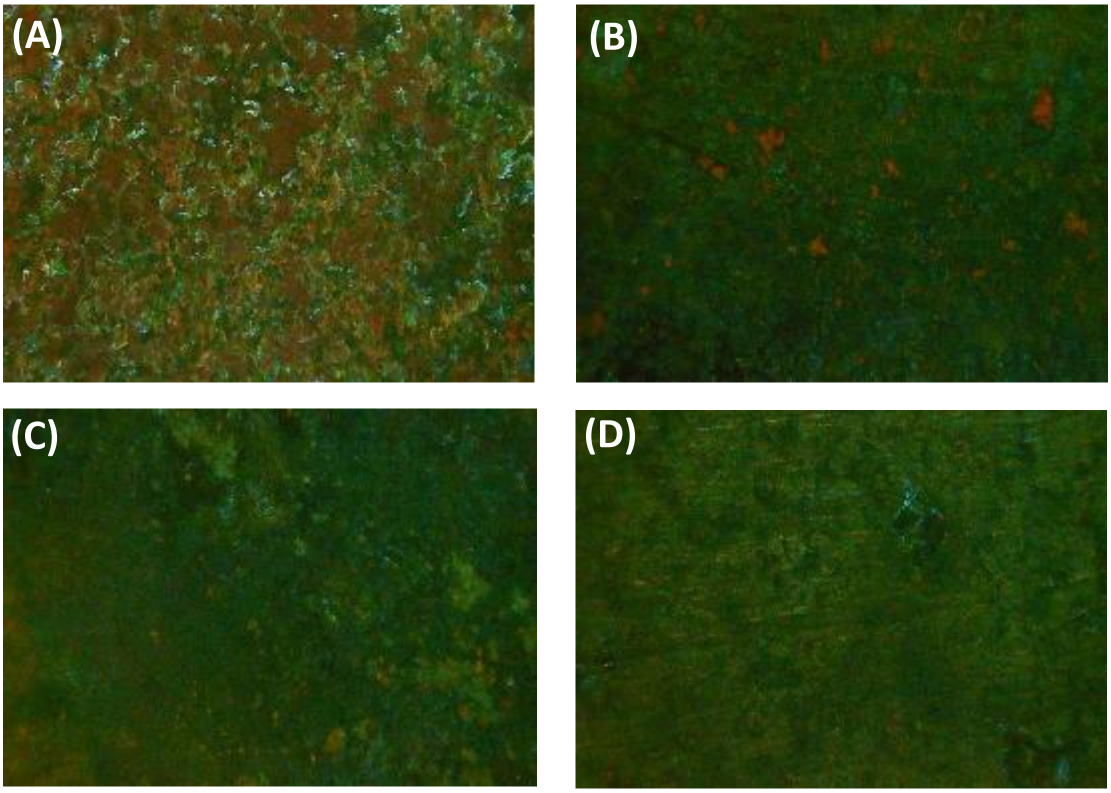

Fig. 1 Surface morphology of specimens: (a) uncoated, (b) tea-brushed, (c) tea-immersed, (d) tea-sprayed.

2. Methods

The experimental procedures in this study adhered to the ASTM G31 standard for laboratory immersion corrosion testing. Test specimens were prepared using mild steel plates measuring 40 mm × 20 mm, with four specimens designated for assessing the effectiveness of coatings derived from tea waste. The extract from tea waste, utilized as a natural corrosion inhibitor, was created by combining 20 g of dried tea waste with 200 mL of distilled water to achieve a 10% (w/v) concentration. The mixture underwent reflux in a sealed system at 100°C for a duration of 2 hours to facilitate the extraction of the active compounds. The steel specimens were precisely cut to the required dimensions utilizing the facilities of a metal workshop located in Banyumanik, Semarang. Following this, the specimens underwent polishing with 120-grit emery paper to eliminate surface oxides and contaminants, and the surface morphology was examined using a digital microscope at 400× magnification.

Before applying the coating, the steel specimens underwent a drying process in an oven set to 150°C for 1 hour, followed by conditioning in a desiccator for 30 minutes to remove any residual moisture. The initial weight of each specimen was meticulously documented for subsequent analysis. Three coating methods were utilized: (i) applying the extract onto the steel surface using a brush, (ii) immersing the specimen in the extract for a duration of 30 minutes, and (iii) uniformly spraying the extract onto the surface. A single specimen was retained uncoated to function as a control. Following the application of the coating, all specimens were allowed to dry at room temperature for a duration of 24 hours to ensure proper film formation and adhesion.

In the corrosion testing procedure, each specimen was suspended in 150 mL of a 3.5 wt.% NaCl solution. This was achieved using a straightforward apparatus comprising a sealed plastic bottle with a hanger system, following ASTM G31 guidelines to guarantee consistent exposure. The specimens underwent immersion for a duration of 48 hours. Following immersion, the specimens underwent re-drying in an oven set to 150°C for 1 hour. Subsequently, they were conditioned in a desiccator for 30 minutes, after which their final mass was recorded.

The corrosion rates were assessed through the weight loss method as outlined by ASTM G31, which involves calculating the difference in specimen weight before and after exposure to the corrosive environment. The corrosion rate (CR) in mils per year (Mmpy) was determined using the following formula (1), as specified by the ASTM G31 standard:

$$\text{Corrosion Rate (Mmpy)} = \frac{8.76 \text{ x } 10^4 \text{ x } \Delta W}{A \text{ x } t \text{ x } \rho} \quad (1)$$

where ΔW is the weight loss (g), A is the exposed surface area (cm^2), t is the exposure time (hours), and ρ is the density of mild steel (7.85 g/cm^3). The weight loss represents the mass of the specimen lost due to corrosion during the exposure period, and the corrosion rate is a measure of the degradation of the material over time. The inhibition efficiency (IE%) was then evaluated by contrasting the corrosion rates of both coated and uncoated specimens, as shown in the equation (2):

$$\text{IE (\%)} = \frac{CR_{control} - CR_{coated}}{CR_{control}} \, x \, 100 \quad (2)$$

These calculations enabled a quantitative assessment of the protective performance of the natural extracts applied using various coating methods, providing a clear understanding of how effectively each coating inhibited corrosion.

3. Results and Discussion

3.1. Tea Waste Extract Coating

The corrosion behavior of mild steel specimens treated with tea waste extract coatings was thoroughly assessed using the ASTM G31 weight loss method. The experimental data presented in Table 1 indicate a significant decrease in corrosion rates for the coated specimens when compared to the uncoated control group. The uncoated specimen (Specimen 1) demonstrated the highest corrosion rate at 3.4523 Mmpy, categorizing it as Poor based on the corrosion rate classification established by Fontana (1986). This aligns with anticipated outcomes, as the lack of any protective layer results in the steel surface being directly subjected to the aggressive chloride ions present in the 3.5% NaCl solution, facilitating swift anodic dissolution and pitting corrosion.

The exceptional efficacy of the tea waste extract coating, especially when applied through brush and immersion techniques, can be linked to the unique chemical composition of tea. Tea leaves contain a significant amount of polyphenolic compounds, mainly tannins, which make up about 7–15% of the total weight of tea (Prasetya, 2015). Tannins are complex polyphenolic compounds characterized by their high molecular weight and the presence of multiple hydroxyl (-OH) and carbonyl (C=O) groups, demonstrating a significant attraction to metal surfaces (Zhang et al., 2023). The presence of these polar functional groups enhances the adsorption of tannins onto the steel surface by enabling coordinate bonding with metal cations (Fe^{2+}/Fe^{3+}), thereby creating a dense and adherent protective layer (Vorobyova et al., 2023).

The organic film that has been adsorbed serves as a barrier, limiting the entry of harmful chloride ions and oxygen, which are essential factors in corrosion processes within saline environments (Tehrani et al., 2021). The effectiveness of corrosion inhibition is significantly affected by the consistency and extent of this protective film's coverage. The brushing and immersion techniques enabled a more comprehensive and consistent interaction between the extract and the steel surface than spraying, which could account for the enhanced inhibition efficiency noted for Specimens 2 and 3.

Furthermore, tannins demonstrate chelating abilities, allowing them to form complexes with iron ions that have leached from the steel surface, thereby further inhibiting anodic dissolution (Proença et al., 2022). The creation of these chelates effectively reduces the rate of metal dissolution while simultaneously enhancing the stability of the passive layer. Moreover, the flavonoids and various catechins found in tea extract could work together to further enhance this inhibition via comparable adsorption and chelation mechanisms (Bernatonienė, & Kopustinskiene, 2018). In contrast, the specimens treated with tea waste extract exhibited markedly reduced corrosion rates, thereby validating the efficacy of the natural inhibitor. Specimen 2 (brush application) demonstrated the lowest corrosion rate of 0.0377 Mmpy, categorizing it as Excellent. In a similar vein, Specimen 3 (immersion coating) demonstrated a corrosion rate of 0.0784 Mmpy, which is classified as Excellent. In contrast, Specimen 4 (spray application) showed a corrosion rate of 0.3283 Mmpy, categorized as Good. The observed reductions indicate inhibition efficiencies of 98.9%, 97.7%, and 90.4%, respectively, when compared to the uncoated specimen.

The observations made through digital microscopy supported these findings. The uncoated specimen demonstrated considerable surface degradation and pitting following immersion in NaCl solution, whereas the coated specimens displayed minimal surface damage, suggesting the integrity of the protective film. The combined weight loss measurements and surface morphology analysis confirm that tea waste extract serves as an effective and environmentally friendly corrosion inhibitor, with its performance significantly influenced by the application method (Fig. 1).

Table 2 Corrosion rate and inhibition efficiency of specimens coated with orange peel extract

Specimen	ΔW (g)	Corrosion Rate (Mmpy)*	Inhibition Efficiency (%)
Specimen 1 (Control)	0.1188	3.4523[a]	-
Specimen 2 (Brush)	0.0016	0.0455[c]	98.7%
Specimen 3 (Immersion)	0.0035	0.1007[b]	97.1%
Specimen 4 (Spray)	0.0030	0.0862[b]	97.5%

*Values with different superscripts within the same column are statistically significantly different ($p<0.05$).

3.2. Orange Peel Extract Coating

The effectiveness of corrosion inhibition provided by coatings made from orange peel extract was assessed using the same methodologies as those utilized for the tea waste extract. The findings, shown in Table 2, indicate that all specimens treated with orange peel extract displayed markedly reduced corrosion rates in comparison to the uncoated control specimen. The uncoated specimen (Specimen 1) exhibited a corrosion rate of 3.4523 Mmpy. According to the corrosion rate classification established by Fontana. (1986), this rate is categorized as "Poor," indicating a corrosion rate that ranges between 1 and 5 Mmpy. This outcome aligns with the anticipated behavior of mild steel when submerged in a corrosive 3.5% NaCl solution in the absence of any protective measures.

Specimen 2, coated via the brushing method, exhibited the lowest corrosion rate of 0.0455 Mmpy, categorizing it as "Excellent" (<0.1 Mmpy), and showing an inhibition efficiency of 98.7%. In a similar vein, Specimen 4, utilizing the spraying method, demonstrated a corrosion rate of 0.0862 Mmpy, placing it in the "Excellent" category, alongside an inhibition efficiency of 97.5%. Conversely, Specimen 3, which employed the immersion method, recorded a corrosion rate of 0.1007 Mmpy, classified as "Good" (0.1–0.5 Mmpy), with an inhibition efficiency of 97.1%. The results demonstrate that the use of orange peel extract coatings effectively decreased the corrosion rate of mild steel in a simulated chloride-rich environment.

The exceptional inhibition performance of orange peel extract coatings is due to the presence of bioactive compounds like flavonoids, phenolics, and tannins, which are plentiful in citrus peels

(Wu et al., 2020). These compounds possess functional groups that exhibit polar characteristics, including hydroxyl (-OH) and carbonyl (C=O) groups. These features promote adsorption onto the metal surface, leading to the formation of a protective film that prevents direct interaction between the metal and the corrosive medium (Pham et al., 2024). The findings indicate that the brushing and spraying application techniques produced more consistent and efficient protective layers in comparison to the immersion method (Fig. 2). The results obtained are consistent with the principles of green corrosion inhibition, showcasing the potential of orange peel extract as a sustainable and environmentally friendly substitute for synthetic inhibitors.

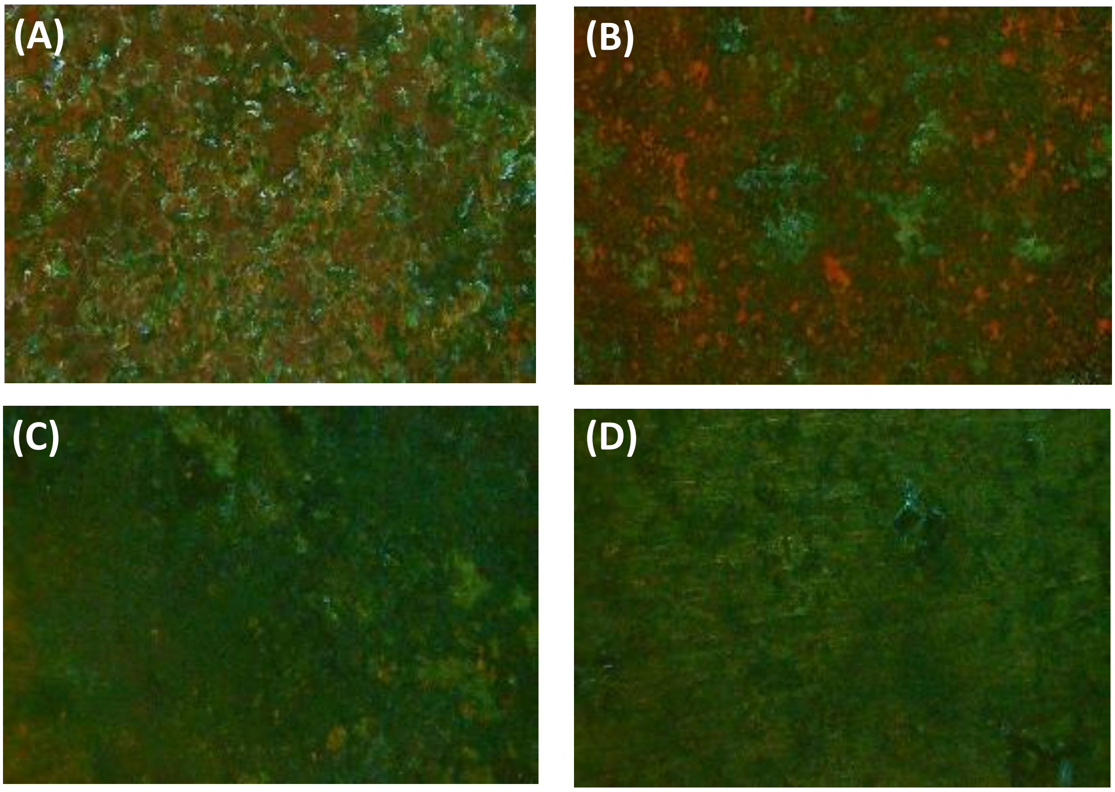

Fig. 2 Surface morphology of specimens: (a) uncoated, (b) orange peel-brushed, (c) orange peel-immersed, (d) orange peel-sprayed.

4. Conclusions

This study effectively demonstrated the potential of tea waste and orange peel extracts as natural corrosion inhibitors for mild steel in a 3.5% NaCl solution. The application of the weight loss method, as specified by ASTM G31, showed that the protective coatings developed from these extracts markedly decreased corrosion rates when compared to the uncoated control specimens. The inhibition efficiencies obtained with tea waste extract were recorded at 98.9%, 97.7% and 90.4%, whereas orange peel extract yielded inhibition efficiencies of 98.5%, 97.3%, and 96.8%, contingent upon the application method employed.

This study presents a distinctive approach by utilizing eco-friendly, natural materials as substitutes for the synthetic corrosion inhibitors that are typically employed in industrial applications. The bioactive compounds found in tea waste and orange peel, including tannins, flavonoids, and phenolics, have demonstrated efficacy in creating protective layers on metal surfaces, thereby inhibiting direct contact between the metal and corrosive environments. This study provides important insights into the use of organic waste materials as affordable and sustainable corrosion inhibitors, leading to more environmentally friendly approaches in corrosion management.

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