



# A Review on The Inhibition Efficiency of Green Materials as a Potential Corrosion Protection on Ferrous and Non-Ferrous Metals in Various Environments

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**Abstract:** Green material is a beneficial material in combating corrosion on any metallic structure. Since it is a green material, hence it can be classified as an environmental friendly material. Environmental friendly material (eco-friendly) is the best material choice to be opted for corrosion mitigation. Eco-friendly materials produce no pollution to the environment, safe to be used because of organic materials, made from a waste product or a natural resource, cheap, wide range of availability, renewable resource, free of heavy metals and free of toxic compounds. Therefore, numerous research studies have been done worldwide by researchers as to study the effectiveness in term of inhibition efficiency of the eco-friendly material as corrosion mitigation method. This review highlighted a few eco-friendly materials such as rice husk, egg shell, banana peel and henna that have been tested successfully in the previous research studies as corrosion prevention method. Many of related and relevant research articles to this review topic have been included as to review the inhibition efficiency of the eco-friendly materials in corrosion mitigation. The result showed that the inhibition efficiency for all green materials reviewed was mainly above 90%. This showed that the green materials acted as a remarkable material either as a direct corrosion inhibitor or incorporated into a coating. The corrosion rate on metal was decreased as the molecules of green material being absorbed on the metal surface, creating a protective barrier and protected the metal surface from corrosion attack. This review provided the significant and valuable information regarding the application of eco-friendly material as corrosion mitigation because the facts in this review have indicated that the eco-friendly materials have an excellent inhibition efficiency for corrosion protection purpose on different metals in various environments.

**Keywords:** Rice husk, egg shell, banana peel, henna, inhibition efficiency

## 1. Introduction

Corrosion is the degradation of materials as a result of reaction with its environment and corrosion will lead to many failures of metallic equipment [1]. Failure of any metallic equipment can lead to major failure of the metallic equipment. Major failure can be catastrophic and cost consuming. Basically, corrosion is inevitable, but it can be controlled or stopped by applying the appropriate corrosion protection methods. The use of corrosion protection methods such as corrosion inhibitors and coating are basically for protecting the metal surface from corrosion attack. Corrosion inhibitor is a substance that when added in a small amount, it can help in retarding the corrosion process and it was used in an internal part of any metallic structures. Major classes of inhibitor are organic inhibitor and inorganic inhibitor. Meanwhile, coating is a protective layer that is applied onto a metal surface with intention of protecting the metal surface from corrosive environment. Coating can be classified into metallic coating and non-metallic coating where non-metallic coating can be furthered extend to organic and inorganic coating.

Even though the corrosion inhibitor and coating have been used for many years, some of the inhibitor and coating materials used for corrosion protection purpose are not safe to be used as some are very toxic and can cause pollution to the environment as well as harmful to human health. For instance, benzotriazoles that were used to minimize the corrosion in the heat exchanger have been proven to poison marine creatures, even though the concentration used was low as 3 ppm [2]. Furthermore, for inorganic inhibitor, it might release toxic arsine gas as the production from corrosion process occurred. Moreover, the disposal of the corrosion inhibitor especially the inorganic inhibitor or synthetic inhibitor could be problematic since it can cause damage to the ecosystem. The use of heterocyclic synthetic compounds as corrosion inhibitor may cause permanent or non-permanent damage to the organ system, for instance, the liver or kidney [3]. In addition, the use of Bisphenol A as anti-corrosion coating could be the source of toxicity to the aquatic environment, causing damage and death of aquatic creatures [3].

Furthermore, due to the reasons that some of corrosion inhibitor and coating materials used are very toxic, can cause pollution to the environment and harmful to human health, it is a wise decision to utilize the use of eco-friendly material in combating corrosion. Eco-friendly material can be considered as a material that has no contribution towards pollution and not harmful to the environment and human health. These actively demonstrate that eco-friendly material is safe to be used widely. Moreover, it is a cost saving material that made from a waste product such as banana peel and also from a natural resource such as from plant. Recycling and inventing the waste product of green material will benefit in lessen the environmental impact by decreasing the amount of waste production. This will help to achieve zero waste production. Eco-friendly material is a renewable resource and readily available. Besides, it is free from heavy metals and toxic compounds which the use of heavy metals and toxic compounds will raise the environmental concerns and health issues such as environment pollution and cancer disease.

This review presented the results based on the effectiveness of a few eco-friendly materials in inhibiting corrosion on both ferrous and non-ferrous metals in various environments. It also can be a source of reference to current and future researcher in studying the application of green waste product in corrosion mitigation methods with focusing on the inhibition efficiency on metals, besides, a reference to industrial companies as well. Moreover, this review can develop the awareness on the need of inventing waste product of green material and natural resource as corrosion protection on metallic structure, thus decreasing the pollution in the environment and human health issue. The eco-friendly material discussed were rice husk waste, eggshell waste, banana peel waste and henna that can be found abundantly in the world as a green waste product.

## 2. Previous Research

A summary of literature review on a few eco-friendly materials was presented in Table 1 with focusing to the effectiveness in term of inhibition efficiency (IE) of the materials in inhibiting corrosion on metal based on past research studies. Numerous articles journal has been selected for this review and the selected articles journal are related and relevant to the review topic. The eco-friendly materials reviewed were rice husk, eggshell, banana peel and *Lawsonia Inermis* or also known as henna. The selection of these eco-friendly materials was made due to that there were many research studies done on these materials and these materials were proven to be effective in corrosion protection on metal surface with higher inhibition efficiency, whether as green corrosion inhibitor, enhancement in coating or even as an enhancement in composite material as to enhance the corrosion resistance characteristic. Moreover, these selected eco-friendly materials were identified to be a waste product either from agriculture industry, food industry and others. Hence, many researchers from around the world were interested in doing research regarding the effectiveness of these eco-friendly materials in combating corrosion on metals and replacing the toxic substances used in corrosion mitigation. The example of toxic substances used in corrosion mitigation are mainly from inorganic corrosion inhibitors such as chromates, zinc and lead oxide while phosphates cause pollution in water. A long-term usage can pose threats to social health. In the same time, the use of sodium nitrite inhibitor is considered as not economical because a high concentration is needed for each time to use. A high concentration in the range of 300-500 mg/L is needed for sodium nitrite inhibitor and this portrays that it is not economical [28]. While, with using a green material as a corrosion inhibitor, it is still be considered as appropriate if a high concentration will be used because it causes no toxicity and pollution.

Based on Table 1, the main focus of this review would be pointed to the inhibition efficiency showed by these eco-friendly materials in inhibiting corrosion on various metal surface in a few electrolytes. To obtain the inhibition efficiency value, a few corrosion test methods were done by past researchers. The test methods involved both gravimetric method such as weight loss measurement and electrochemical method such as linear polarization resistance measurement, potentiodynamic polarization measurement and electrochemical impedance spectroscopy measurement. These tests were conducted as to test the corrosion resistance of the metal when the eco-friendly materials were used either as an inhibitor or being incorporated into a coating. Weight loss measurement involved the immersion test of the metal substrate into a solution that contain inhibitor, or the metal substrate has been coated earlier and tested the corrosion behaviour of the metal substrate in a particular electrolyte. Meanwhile, linear polarization resistance measurement, potentiodynamic polarization measurement and electrochemical impedance spectroscopy measurement involved the use of three electrodes namely working electrode (the tested metal substrate), reference electrode and counter electrode in a particular electrolyte. All these corrosion measurement methods will make it easier for past

researchers to calculate the inhibition efficiency of the eco-friendly materials as a corrosion resistance enhancement on ferrous and non-ferrous metal surface. The material concentration and temperature for all corrosion tests used in past studies were considered as to relate with inhibition efficiency value obtained.

Inhibition efficiency will reveal on how well the material would be to combat or inhibit corrosion as to protect the metal from degradation or damage caused by corrosion process in any environments. The inhibition efficiency of a material is influenced by the absorption process on the metal surface where the absorption process will result in the formation of a protective layer that prevent a direct contact between the metal surface and corrosive environment. The absorption of the green material on the metal surface can be in various types of namely as physical absorption, chemical absorption as well as mixed absorption where mixed absorption involves both physical and chemical absorption. The higher percentage value of inhibition efficiency would recommend an excellent corrosion resistance behaviour of the metal towards corrosion in particular environment while a lower percentage value of inhibition efficiency would recommend an unsatisfied corrosion resistance behaviour of the metal towards corrosion in particular environment. In Table 1, the summary was done in sequence where the first summary was rice husk, followed by eggshell, banana peel and henna. Meanwhile, n.m. in Table 1 represents the shortcut to not mentioned. At the same time, HCl represents hydrochloric acid, NaCl represents sodium chloride (seawater), H<sub>2</sub>SO<sub>4</sub> represents sulfuric acid and HNO<sub>3</sub> represents nitric acid.

**Table 1 - Summary of previous research findings**

Reference/ year	Material	Parameter	Substrate	Environment	Maximum IE (%)
[4]/2020	Rice Husk (inhibitor)	Concentration :5-20%  Temperature: n.m.	Steel Reinforced Bar (in concrete)	Seawater (NaCl)	85.04
[5]/2019	Rice Husk (rice husk + acrylic coating)	Concentration: n.m. Temperature: n.m.	Mild Steel	Acid (HCl) and Seawater (NaCl)	45.38 (acid) and 77.75 (seawater)
[6]/2019	Rice Husk (inhibitor) + Iodine ion (50 ppm)	Concentration: 250-1250 ppm  Temperature: 313-353 K	Mild Steel	Acid (H <sub>2</sub> SO <sub>4</sub> )	65.48 (at 353 K) and 78.88 (at 313 K)
[7]/2018	Rice Husk (inhibitor)	Concentration: 10-25 ppm Temperature: Room Temperature	Mild Steel	Acid (H <sub>2</sub> SO <sub>4</sub> )	96.60
[8]/2017	Rice Husk (inhibitor)	Concentration: 0.10-0.25 g/L  Temperature: 313-333 K	Mild Steel	Acid (HCl and H <sub>2</sub> SO <sub>4</sub> )	82.65 (at 313 K) 87.50 (at 333 K) (HCl) and 74.64 (at 333 K) 94.24 (at 313 K) (H <sub>2</sub> SO <sub>4</sub> )
[9]/2017	Rice Husk (inhibitor)	Concentration: 5-25 mL  Temperature: 25°C	Brass	Acid (HNO <sub>3</sub> )	99.10
[10]/2017	Rice Husk (inhibitor)	Concentration: 0.003-0.030 M Temperature: n.m.	Carbon Steel (SAE 1045)	Seawater (NaCl)	88.40
[11]/2017	Rice Husk (inhibitor)	Concentration: 0.003-0.030 M	Carbon Steel (SAE 1045)	Distilled water	98.40

		Temperature: n.m.			
[12]/2016	Rice Husk (inhibitor)	Concentration: 175-1500 ppm Temperature: n.m.	Carbon Steel	Seawater (NaCl)	86.00
[13]/2015	Rice Husk (inhibitor)	Concentration: 0.1-0.5% v/v Temperature: 303-333 K	Mild Steel	Acid (HCl and H <sub>2</sub> SO <sub>4</sub> )	96.00 (HCl) and 86.00 (H <sub>2</sub> SO <sub>4</sub> )
[14]/2014	Rice Husk (inhibitor)	Concentration: 5-25 ppm Temperature: n.m.	Aluminium (6061)	Acid (HCl)	67.00
[15]/2013	Rice Husk (sodium silicate inhibitor)	Concentration: 500 ppm Temperature: Room Temperature	Copper (99.9%), Aluminium Alloy (6061) and Carbon Steel (SAE 1045)	Acid (HCl)	27.00 (copper), 99.00 (aluminium) and 100 (carbon steel)
[16]/2013	Rice Husk (inhibitor)	Concentration: 5-20 ppm Temperature: 25°C	Carbon Steel (SAE 1045)	Distilled water	99.50
[17]/2013	Rice Husk (inhibitor)	Concentration: 25-175 ppm Temperature: 25°C	Carbon Steel (SAE 1045)	Acid (HCl)	99.00
[18]/2019	Egg Shell (inhibitor)	Concentration: 2-10 g Temperature: n.m.	Austenitic Stainless Steel (Type 904L)	Acid (H <sub>2</sub> SO <sub>4</sub> )	99.45
[19]/2019	Egg Shell (zinc + egg shell) (coating)	Concentration: 5-20 g/L Temperature: n.m.	Mild Steel	Seawater (NaCl)	99.92
[20]/2018	Egg Shell (inhibitor)	Concentration: 2-10 g Temperature: n.m.	Austenitic Stainless Steel (Type 316)	Acid (H <sub>2</sub> SO <sub>4</sub> )	94.74
[21]/2018	Egg Shell (inhibitor)	Concentration: 2-10 g Temperature: Room Temperature	Austenitic Stainless Steel (Type 904L)	Seawater (NaCl)	100
[22]/2018	Egg Shell (inhibitor)	Concentration: 2-10 g Temperature: 15-35°C	Austenitic Stainless Steel (Type 316)	Acid (H <sub>2</sub> SO <sub>4</sub> )	86.99 (at 35°C) and 90.53 (at 15°C)
[23]/2010	Egg Shell (inhibitor)	Concentration: 0.05-0.5 %	Mild Steel	Acid (H <sub>2</sub> SO <sub>4</sub> )	97.00

[24]/2021	Banana Peel (50%) + Orange Peel (50%) (inhibitor)	Temperature: 303-353 K Concentration: 500-2000 ppm Temperature: Room Temperature	Bright Steel	Acid (HCl)	79.19
[25]/2020	Banana Peel (inhibitor)	Concentration: 0.1-4 g/L Temperature: 298.15-308.15 K	Mild Steel	Acid (HCl)	81.65 (at 308.15 K) and 90.00 (at 298.15 K)
[26]/2019	Banana Peel (inhibitor)	Concentration: 100-500 ppm Temperature: 25°C	Mild Steel	Acid (HCl)	87.44
[27]/2019	Banana Peel (inhibitor)	Concentration: 300-500 ppm Temperature: 25-60°C	Mild Steel	Acid (HCl)	64.50 (at 60°C) and 88.99 (at 25°C)
[28]/2018	Banana Peel (inhibitor)	Concentration: 2-6 g/L Temperature: n.m.	Carbon Steel AISI 4041	Acid (HCl)	78.59
[29]/2018	Banana Peel (inhibitor)	Concentration: 200-400 mg/L Temperature: 26°C±1°C	Mild Steel	Acid (HCl and H <sub>2</sub> SO <sub>4</sub> )	90.00 (HCl) and 87.00 (H <sub>2</sub> SO <sub>4</sub> )
[30]/2017	Banana Peel (inhibitor)	Concentration: 0.03-0.25 % (gm/mL) Temperature: Room Temperature	Mild Steel	Acid (HCl)	92.92
[31]/2015	Banana Peel (inhibitor)	Concentration: 100-300 mg/L Temperature: 26°C±1°C	Mild Steel	Acid (HCl)	92.00
[32]/2012	Banana Peel (inhibitor)	Concentration: 0.005-2.00% v/v Temperature: 30°C	Mild Steel	Acid (HCl)	96.08
[33]/2021	Henna (polycaprolactone + henna) (coating)	Concentration: 1% w/w Temperature: 37°C	AZ31 Magnesium Alloy	Hank's solution (saline electrolyte)	98.30
[34]/2020	Henna (inhibitor)	Concentration: 0.05-2.50 g/L Temperature: 25°C	API 5L Gred B (carbon steel)	Hydrogen sulfide solution	92.00
[35]/2019	Henna	Concentration:	Mild Steel	Well water	96.00

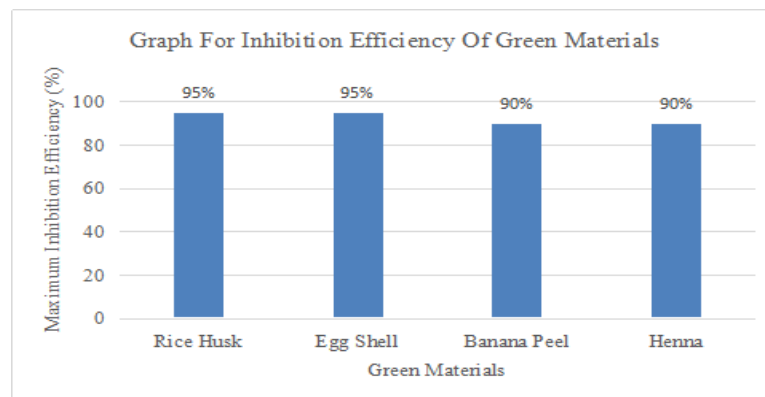
	(inhibitor)	2-10 mL Temperature: n.m.			
[36]/2017	Henna (inhibitor)	Concentration: 200-1000 ppm Temperature: n.m.	Aluminium Alloy 5083	Seawater (NaCl)	92.00
[37]/2017	Henna (anti-corrosion paint + henna)	Concentration: 4-10 % Temperature: Room Temperature	Aluminium Alloy 5083	Seawater (NaCl)	96.25
[38]/2017	(coating) Henna (acrylic resin + henna) (coating)	Concentration: 0.1-0.4 wt/vol % Temperature: n.m.	Aluminium Alloy 5083	Seawater (NaCl)	99.90
[39]/2013	Henna (inhibitor)	Concentration: 100-3000 mg/L Temperature: 20-60°C	Carbon Steel	Acid (HCl)	67.73 (at 60°C), 88.42 (at 20°C) and 92.72 (at 25 °C)
[40]/2012	Henna (inhibitor)	Concentration: 100-500 ppm Temperature: n.m.	Aluminium Alloy 5083	Seawater (NaCl)	88.00
[41]/2012	Henna (inhibitor)	Concentration: 0.2-1.6 g/L Temperature: 28°C	N80 API Steel	Mud acid solution (HCl + ammonium bifluoride)	92.62
[42]/2012	Henna (polymerized vinyltrimethox ysilane + henna)	Concentration: 0.012-0.1 % Temperature: 37°C ± 1°C	316L Stainless Steel	Simulated body fluid	92.53
[43]/2012	(coating) Henna (inhibitor)	Concentration: 10-500 mg/L Temperature: 30-45°C	Mild Steel	Acid (HCl)	44.20 (45°C) and 93.14 (30°C)
[44]/2009	Henna (inhibitor)	Concentration: 0.2-2.0 g/L Temperature: 25-60°C	Mild Steel	Acid (HCl)	37.95 (at 60°C) and 92.59 (at 25°C)

### 3. Discussion on Previous Findings

Egg shell waste is a waste product from food industries or from domestic waste and can be found easily around the world. According to Sanni et. al. [21], eggshell is made up of 95% of calcium carbonate, 1% of calcium phosphate and other organic matters. It has been mentioned by Aigbodion et. al. [19] that due to the chemical compositions and wide availability of eggshell, it makes eggshell as a potential source for coating and composites. It will enhance the properties of the coating and composites, especially in corrosion resistance behaviour. Next, banana peel waste is a waste product either from food industries or from domestic waste and can be found abundantly in most Asian countries, especially in Malaysia where in Malaysia, the banana peel waste can be collected easily from roadside vendors that sell banana fritters. Banana peels were remained unused and thrown in the environment, causing serious environmental problems when rotting [31]. When banana peels undergo rotting process in the landfill, it will produce methane gas. The production of methane gas has contributed to a major portion in the greenhouse gases towards global warming

[29]. Moreover, Tiwari et. al. [29] and Ji et. al [31] stated that the presence of bioactive compounds like catechin, ascorbate, gallic acid and epigallocatechin gallate have made banana peels as a suitable material to be studied for the inhibition capability. Furthermore, rice husk waste is a waste product from agriculture industries and most of Asian countries have paddy field. Rice husk is the shell from rice that being produced during de-husking of the rice and rice plant absorbs silica from soil and then assimilates the silica into its own structure during the growth process [45]. There is a high percentage of silica concentrated in the rice husk with 80-85% and silica is a remarkable ingredient to enhance coating properties such as scratch and abrasion resistance as well as silica is highly chemical durable in various media [45]. Next, henna or scientific name as *Lawsonia Inermis* is a natural source from plant and most of it was used in beauty industries as colouring agent to finger and toe nails. Henna is consisting of four main constituents that named as Lawsone, tannic acid,  $\alpha$ -D-Glucose and gallic acid that can combine with metal cations and be absorbed on the metal surface as in order to protect from corrosion attack on the metal [46]. All these four materials were capable in inhibiting corrosion on any metallic structure in various environments.

Generally, much researches were done mainly by Asian countries. This is due to the wide availability of these green materials in Asian countries. According to Table 1, it can be clearly seen that rice husk, eggshell, banana peel and henna acted as a potential natural material in corrosion inhibitor and coating for corrosion protection purpose. All green materials recorded a remarkable result for the inhibition efficiency through all past studies. Based on the corrosion test results for all green materials, the mode of inhibition efficiency values recorded for rice husk and eggshell were mainly above 95% while for banana peel and henna were mainly above 90% as shown in Fig. 1. This depicted that all these four green materials work efficiently in protecting the ferrous and non-ferrous metal surface from corrosion attack in various environments. The excellent value for inhibition efficiency obtained was influenced by the high concentration of green material used in all corrosion tests. It has been identified in all past studies that as the concentration of rice husk, eggshell, banana peel and henna was increased, the inhibition efficiency also was increased and eventually, the corrosion rate was decreased. A high value of inhibition efficiency will suggest that there is the existence in formation of the inhibitor surface complex (complex compound) that provided a barrier protection, hence, less corrosion attack on metal surface by corrosive medium [15, 36]. The formation of the complex compound on the metal surface has an insoluble characteristic, which means that the compound produced will not easily soluble in electrolyte and able to protect the metal surface from destruction caused by corrosion attack. Moreover, when corrosion rate was decreased, it showed that these green materials had successfully protected the metal surface from corrosion attack in a particular environment, either in seawater solution, acid solution, hydrogen sulfide solution, simulated body fluid or even in distilled water and well water. Various ferrous metals and non-ferrous metals were used in past studies as to investigate the corrosion resistance of the metal when being protected by the green materials that used either as inhibitor or being incorporated into coating.



**Fig. 1 - Maximum inhibition efficiency of green materials**

Basically, rice husk was proven to be effective in protecting various metals such as mild steel, brass, carbon steel, aluminium and copper while egg shell was proven to be effective in protecting metals such as austenitic stainless steel and mild steel. In a coating system, the addition of white rice husk ash (more silica content and less carbon content) as a filler in epoxy paint has contributed to the enhancement of the paint properties, for instance, scratch and wear resistance as well as the elongation [47]. Obviously, if the scratch resistance is higher, it means that the metal substrate that being coated will not easily exposed or in direct contact with moisture, thus prevented from corrosion attack since the coating itself does not easily break down. It has been identified in past studies that silica in rice husk was responsible to hinder corrosion on metal surface, thus, protect metal from deterioration caused by corrosion process. In 2016, Ahmaed et. al. [45] have mentioned that silica is considered as a well-known and an efficient compound in corrosion protection as well as an enhancement in coating properties. In 2017, it has reported that the nano-size silica has a high surface area and since it has a high surface area, it provides a high absorbance and thus increased the formation of the protective film on the brass surface in nitric acid solution [9]. The brass was eventually being

protected from corrosion attack in acid environment. Previously in 2013, Awizar et. al. reported that nanosilicate that produced from nanosilica has successfully protected the carbon steel (SAE 1045) in distilled water from corrosion attack by the formation of a thin protective film on the metal surface [16]. Next, it has been identified in past studies that eggshell contains a high amount of calcium carbonate and according to [48], the contain of calcium carbonate in eggshell is made up of 94-95%. It was believed that calcium carbonate was responsible to forestall corrosion attack on metal surface either as inhibitor or enhancement in coating. According to [48] as well as [49], The calcium carbonate precipitation from eggshell can be formed as a coating on grey cast iron and mild steel as to forestall the diffusion of oxygen that contribute to corrosion through electrophoretic deposition (EPD) method [48,49]. Jafar et. al. [49] reported that the coating of eggshell had provided a good adhesion strength on metal and Bakar et. al. [48] have stated that coating is one of the most efficient methods to reduce corrosion attack on metals. Eggshell waste was proven to be effective against corrosion attack on austenitic stainless steel (Type 904) in acid environment, posed a higher inhibition efficiency value of 99.45% from weight loss measurement [18]. Meanwhile, Aigbodion et. al. [19] have reported that the coating on mild steel that was made from incorporated the zinc and eggshell powder has successfully protected the mild steel from corrosion attack in simulated seawater solution, posed a high inhibition efficiency value of 99.92% from potentiodynamic polarization measurement. These have showed that calcium carbonate in eggshell was able to protect stainless steel and mild steel from degradation caused by corrosion attack. Meanwhile, a coating that made from incorporated of zinc and eggshell powder has able to increase the thermal stability as well as decreased the decarburization occurrence in mild steel [19]. In short, the presence of silica in rice husk and calcium carbonate in eggshell have provided a maximum protection on metal surface in various environments.

Furthermore, banana peel was proven to work best in protecting metals such as mild steel and carbon steel, while henna was proven to work best in protecting metals such as magnesium, carbon steel, mild steel, aluminium, N80 API steel and stainless steel. Vani et. al. [24] found out that incorporating a balance extract of banana peel (50%) and orange peel (50%) as a corrosion inhibitor has successfully inhibited corrosion attack on bright steel in HCl solution with 79.19% through weight loss measurement in 24 hours of immersion. Tannin, a complex polyphenol compound from banana peel extract was absorbed on metal surface and form a protective thin layer on metal surface, thus help to protect the metal from corrosion attack [24,28]. The thin layer formed on metal surface cannot be seen with human naked eyes. Banana peel has the potential to be a corrosion inhibitor that is safe to be used and proven to retard corrosion on metal surface. Tannin in banana peel has the potential to lower the corrosion rate and suitable to be used as a corrosion inhibitor [28]. Besides, a study by Ji et. al. [31] has showed that compounds like gallic acid and catechin have contributed to protect the mild steel from corrosion attack in acid medium while a study by Tiwari et. al. [29] has mentioned that protonated gallic acid compound has contributed to protect the mild steel in acid medium from corrosion attack. Banana peel can act as a good green corrosion inhibitor that prevent corrosion attack on mild steel, for instance, cracking and pitting corrosion by the absorption of the inhibitor molecules on mild steel surface in HCl solution [25]. As for henna, the increasing henna percentage used in coating has depicted an excellent inhibition ability on metal surface [50]. This eventually protects the metal from corrosion phenomenon. The use of henna in a coating has portrayed a remarkable output where all inhibition efficiency values recorded was above 90% as can be seen in Table 1. As for the direct use of henna as an inhibitor, the inhibition efficiency values recorded was mainly above 90%. This indicates that henna is suitable to be incorporated in a coating as to improve the corrosion resistance characteristic as well as henna is suitable to be used as a green inhibitor in an electrolyte. Next, from past studies, it was found that lawsone was the responsible constituent in henna that inhibited corrosion attack on metal surface. Lawsone was the main constituent in henna that was responsible for the inhibition effect on aluminium alloy 5083 tested in sea water medium [51]. Meanwhile, Ostovari et. al [44] reported that lawsone was the main constituent of henna that was responsible in protecting the mild steel in HCl medium from corrosion attack, posed the inhibition efficiency value of 92.59%. In short, the presence of tannin, gallic acid, catechin and protonated gallic acid in banana peel as well as lawsone in henna have provided an extra protection on metal surface in various environments.

Moreover, besides a high concentration of green materials used, temperature also will influence the inhibition efficiency values obtained in all corrosion tests. Even though it was proven that increase the concentration of green materials will benefit in such that the inhibition efficiency will increase, but the drawback lies in the temperature. It has been identified in all past studies that an increase of temperature up to a certain level (maximum temperature) will decrease the inhibition efficiency of green materials. It was discovered that green materials used in past studies work well in room temperature. This is a good sign that explains the use of green materials should be encouraged and the materials work best in room temperature. Even though rice husk works very well in room temperature, unfortunately, it reported that as the temperature was increased up to 80°C, the inhibition efficiency of rice husk was decreased (65.48%) [6]. However, the good sign is that the inhibition efficiency was still above 50% which showed that it still works well in protecting the metal from corrosion attack. Besides that, it reported that rice husk inhibitor works best from 40°C (313 K) until 60°C (353 K) with 82.65% (313 K) to 87.50% (353 K) of the inhibition efficiency values for mild steel in HCl solution [8]. In addition, the rice husk inhibitor performs best in 40°C (313 K) until 60°C (353 K) for mild steel in H<sub>2</sub>SO<sub>4</sub> solution, although there is a reduction in the inhibition efficiency from 94.24% (313 K) to 74.64% (353 K) [8]. For the value of 74.64%, it was still above 50% of inhibition efficiency and this showed that the rice husk was still able to protect the mild steel in H<sub>2</sub>SO<sub>4</sub> solution efficiently. At the temperature of 30°C until 60°C, the



inhibition efficiency values recorded for the use of 0.5% of rice husk concentration were all above 80.00% for mild steel in HCl solution while in H<sub>2</sub>SO<sub>4</sub> solution, the inhibition efficiency values recorded for the use of 0.5% of rice husk concentration were all above 50.00% and 86.00% was the highest inhibition efficiency recorded for mild steel in H<sub>2</sub>SO<sub>4</sub> solution [13]. Eggshell also perform well in room temperature [22]. As the temperature was at 35°C and with a total of 96 hours of immersion time in weight loss measurement for austenitic stainless steel (Type 316), the inhibition efficiency recorded was 86.99% in acid medium which showed that the eggshell concentration works well until 35°C [22]. Previously, Rajalakshmi et.al. reported that when the temperature in the range of 30°C to 50°C, the inhibition efficiency recorded was in the range of 83-88% for mild steel in H<sub>2</sub>SO<sub>4</sub> medium. This has clearly indicated that eggshell works best as an inhibitor until the temperature of 50°C [23]. However, the drawback is that when increasing the temperature for more than 50°C, the inhibition efficiency of eggshell was decreased. In short, rice husk and eggshell work best in room temperature up until certain temperature where keep increasing the temperature will tend to decrease the inhibition efficiency.

Next, banana peel works efficiently in room temperature and up to 40°C, but decrease after 40°C. One study in 2020 by Kumar et. al. has showed that banana peel portrayed the inhibition efficiency of 81.65% at 35°C for mild steel in acid medium [25]. Meanwhile a previous study by Gunavathy et. al mentioned that after a total of 24 hours of immersion from weight loss measurement for mild steel in acid medium at 30°C, the inhibition efficiency recorded was 89.40% where during the first 5 hours of immersion, the inhibition efficiency recorded was the highest with 96.08% [32]. Within that, it has been reported that at 25°C, the inhibition efficiency values recorded for banana peel were 90.00%, 87.44% and 88.99% respectively for mild steel in acid medium [25, 26, 27]. Meanwhile, at temperature of 26°C±1°C, [29] have reported that mild steel tested in HCl solution as well H<sub>2</sub>SO<sub>4</sub> solution was protected by the banana peel inhibitor, posed the inhibition efficiency values of 90.00% and 87.00% respectively. On the other hands, it reported that mild steel was protected in acid medium with 92.00% of inhibition efficiency [31]. All these with the meaning that banana peel portrayed an excellent performance in protecting mild steel in acid environment. Moreover, even though it was mentioned that after 40°C where the inhibition efficiency was decreased, there was one study by Rosli et. al in 2019 has showed that at 60°C, the inhibition efficiency recorded was 64.5% in total of 24 hours of immersion from weight loss measurement for mild steel in acid medium [27]. This has indicated that even the temperature was increased up to 60°C, the inhibition efficiency was still above 50% and the banana peel still perform to protect the metal surface from corrosion attack. As for henna, it does works well in room temperature up until 60°C. A research by [43] Dananjaya et. al. reported that henna was effective in inhibiting corrosion attack on mild steel in HCl medium with 93.14% of inhibition efficiency from weight loss measurement at 30°C for four hours of immersion time [43]. Next, Hamdy et. al. has mentioned that in the range of 30°C to 50°C in total of 24 hours of immersion time from weight loss measurement for carbon steel, the inhibition efficiency of henna recorded was slightly decreased from 85.32% to 75.69% and it was still a good inhibition efficiency recorded where the henna concentration still performs the duty of protecting the metal surface from corrosion attack in HCl medium [39]. However, it reported that keep increasing the temperature to 60°C, the inhibition efficiency was lowered to 67.73% [39]. Even though the inhibition efficiency was decreased, it was still above 50% where it showed that henna still work well to protect the metal from corrosion attack. In the application of coating, it reported that a combination of polycaprolactone and henna concentration as a coating on AZ31 magnesium alloy in saline electrolyte at 37°C has successfully posed an inhibition efficiency value of 98.30% [33]. While, mixing an anti-corrosion paint with henna concentration as a coating has able to protect aluminium alloy 5083 in seawater environment, tested in room temperature of Malaysia [37]. In addition, it reported that inhibition efficiency value of 92.53% was recorded when a mixture of polymerized vinyltrimethoxysilane with henna concentration was used as a coating on 316L stainless steel in simulated body fluid at 37°C±1°C [42]. These showed that henna performs well in coating system besides in the use as a direct inhibitor in a solution. In short, banana peel and henna perform well in the protection on metal surface in room temperature up to certain temperature where keep increasing the temperature will eventually decrease the performance.

Basically, based on the result in all past studies for rice husk, eggshell, banana peel and henna, it is understandable that there are studies that will produce the same outputs while there are some studies that will produce different outputs. For instance, among rice husk as a corrosion inhibitor, the result of inhibition efficiency in each past study may be different because different medium with different metal substrate being tested even though the same type of green material being used. Besides, when applying corrosion test, the total of immersion time and day may be different in each past study that cause the inhibition efficiency value to be different. The different of temperature used in all corrosion tests does influence the inhibition efficiency value recorded. Moreover, different concentration of the same type of green material used in past studies also influence the inhibition efficiency value to be different. With all of these reasons, the same situation happened to the inhibition efficiency value of eggshell, banana peel as well as henna. As comparison among the four green materials used in the past studies, due to different active compounds presence in all four green materials that responsible for corrosion protection, it gives influence to the inhibition efficiency value to be different between the four green materials. For instance, rice husk contain silica, eggshell contain calcium carbonate, banana peel contain tannin, gallicocatechin, catechin and protonated gallicocatechin as well as henna contain lawsone. Each of active compound may give different result for the inhibitory effect on metal surface for corrosion protection purpose.

In observing the inhibition efficiency recorded in past studies, it is obvious that the inhibition efficiency was influenced by the absorption process and it can be in various types namely physical absorption, chemical absorption and mixed absorption (physical and chemical absorption). There are also a few absorption models (absorption isotherm) that can be used as to illustrate the absorption mechanism of green material as corrosion inhibitor such as Langmuir, Temkin, Freundlich and others. The absorption model was used as to study the interaction between metal surface and the inhibitor molecules as well as the equilibrium of the adsorption process [36]. Based on the absorption of green material molecules on the metal surface, it will create a protective barrier that will prevent a direct contact of the metal surface with the corrosive environment, hence, the metal is protected from corrosion attack. Physical adsorption stated that inhibition occurs as there is electrostatic attraction between the charged molecules of the inhibitor and metal surface while chemical adsorption stated that a better absorption can happen when the charged molecules of the inhibitor and metal surface forming chemical bonds through sharing of electrons. According to past studies, it has been reported that the adsorption of rice husk, eggshell, banana peel as well as henna inhibitor was mainly in physical adsorption. Ahmad et. al. mentioned the physically adsorbed species can be removed from the metal surface by increasing the temperature and increasing the fluid velocity [1]. This explained on why green material inhibitor work best up to a certain temperature where keep increasing the temperature in exceeding the limit temperature might destroy the complex compound formed on the metal surface. Keep increasing the temperature in exceeding the limit temperature will leave the metal surface unprotected and prone to corrosion attack. Besides that, chemical adsorption will result in a strong binding between the inhibitor molecules with the metal surface and chemical adsorption is more effective process because it is not a reversible process [1]. When it means an irreversible process, the complex compound formed is not easily removed due to a strong chemical bond developed between the inhibitor molecule (adsorbate) and the metal surface (adsorbent).

Basically, a different in charge (positively charged and negatively charged) between metal surface and the green inhibitor molecule will make it possible for the formation of insoluble complex compound on the metal surface. For example, when the metal surface is positively charged, hence the negatively charged of the green inhibitor molecule will be absorbed on the metal surface. The same thing happens when the metal surface is negatively charged where positively charged of the green inhibitor molecule will be absorbed on the metal surface. The determination on what type of absorption would the inhibitor portrayed is as stated in Table 2 [10, 11, 30, 36]. Gibbs free energy value can be used as to characterize the interaction strength and stability of adsorbed layer of the inhibitor molecule on the metal surface [11] with regarding to the types of adsorptions. A positive value of Gibbs free energy will indicate that the adsorption process occurs non-spontaneously while a negative value of Gibbs free energy will indicate that the adsorption process occurs spontaneously in retarding corrosion process. Besides, the absorption of the green inhibitor molecules on the metal surface is also facilitated by the presence of heteroatoms such as oxygen and nitrogen in their structure. The heteroatoms are responsible for the inhibitory effect which help to better the absorption of the green inhibitor molecules to the metal surface. Furthermore, as according to past studies, most of the research had reported and agreed that rice husk, eggshell, banana peel as well as henna inhibitor followed the Langmuir adsorption isotherm. Langmuir adsorption isotherm works on the assumption that the adsorption process does take place only at the specific homogeneous sites of the metal surface which the adsorption forms a monolayer (a single layer) on the metal surface [8]. This model mentioned that there is no interaction between the adsorbed molecules. However, in a case of rice husk, there are studies reported that rice husk inhibitor follows the Temkin adsorption isotherm with mixed type of absorption. Temkin adsorption isotherm works on the assumption that there is a molecular interaction between adsorbed molecules [30]. The existence of different type of adsorption isotherm is depending on a few factors [11]. The factors are type of inhibitor, concentration of inhibitor used, corrosion medium used, temperature as well as methods of measurement being carried out.

**Table 2 - Indicator on choosing the types of absorption**

<b>Type of absorption</b>	<b>Indicator</b>
Physical	$\Delta G^{\circ}_{ads} = -20 \text{ kJ mol}^{-1}$ or lower
Chemical	$\Delta G^{\circ}_{ads} = -40 \text{ kJ mol}^{-1}$ or more negative than $-40 \text{ kJ mol}^{-1}$
Mixed	$\Delta G^{\circ}_{ads} = -21 \text{ kJ mol}^{-1}$ to $-39 \text{ kJ mol}^{-1}$

To sum up everything that has been stated so far, all past studies suggested that rice husk, egg shell, banana peel and henna can be used as a corrosion protection method for many types of metal in various environments. The green materials can act as a green inhibitor or as an enhancement in a coating. The presence of green material in a coating was aimed to be acted as a natural corrosion inhibitor that will help the coating to combat corrosion on metals surface. It enhances the coating protection properties, especially in corrosion resistance property. As for the direct use of inhibitor concentration in an electrolyte, the green inhibitor molecules will be absorbed on the metal surface, creating a

protective layer (insoluble complex compound) on the metal surface, thus, protect the metal surface from corrosion attack. The inhibition efficiency of more than 75% in most of the cases can be obtained by using green corrosion inhibitors [24]. It is a good sign that green materials can be used in protecting metallic structures in various industries, all over the world. Moreover, with considering of safety and environmental issues around the world, the development of corrosion inhibitors from natural materials and bio-based resources which are non-toxic, effective and eco-friendly is essential [52]. Nevertheless, the key points here are increasing the concentration of a green material will increase the inhibition efficiency, increasing the concentration of a green material will decrease the corrosion rate and green materials work very best in room temperature until certain temperature where keep increasing the temperature might lower the inhibition efficiency to certain level. For the green material concentration, until the maximum concentration is obtained where it means that the absorption of inhibitor molecules on metal surface is maximum, an increase of the concentration of green material will no longer affects the effectiveness of the inhibitor. Moreover, it cannot be assumed that it is not relevant to implement the use of natural product or waste material in combating corrosion because it has been proven that the implementation of natural product or waste material such as henna, banana peel, eggshell and rice husk have showed an excellent corrosion protection behaviour on metal with higher inhibition efficiency through various corrosion tests. Hence, it is relevant to implement the use of natural product or waste material as corrosion protection on metals, especially in small factories that involve small industries. The repair costs can be saved significantly and spend to other needs.

#### 4. Conclusion

As conclusion, all four green materials discussed have showed a higher inhibition efficiency, mostly was above 90%. Besides, all four green materials discussed have also showed good outcomes when incorporated into a coating. These have showed that all four materials can be used widely as in order to fight corrosion attack on any metal surfaces in various environments because the use of these four materials have proven that it increased the corrosion resistance of metal structures, especially in acidic medium. The presence of silica in rice husk, calcium carbonate in eggshell, tannin, gallic acid, catechin and protonated gallic acid in banana peel as well as lawsone in henna were proven to be a responsible agent in protecting various metal surface from deterioration caused by corrosion process in various environments. As the world is facing environmental pollution, it is very practical and imperative to utilize green materials, either from waste product or even natural resource such as from plants as to decrease the pollution level. Preserving the well-being of the environment is the responsible of human beings. If industry in the world can implement the use of green material as in corrosion protection method, the repair cost can be save as well as the zero waste production in the world could be achieved.

This review is hoped to provide valuable knowledge and information for further investigations in the study of green materials as to be used in corrosion mitigation methods. It is hoped that with this review, all industries can start to pay attention to the effectiveness of green materials in combating corrosion problems. It is suggested that more research projects in the future are hoped to be continuously done as to test the effectiveness of green materials from time to time as corrosion mitigation methods, thus, providing the new or current data regarding the research outcomes, for example, the inhibition efficiency, corrosion rate and others related. It is also suggested that a review for other green materials besides eggshell, banana peel, rice husk and henna should be done as to provide a wide range of view for other green materials in inhibiting corrosion on metal in various solutions.

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