

THE EFFECTIVENESS OF WATERMELON RIND EXTRACT AS CORROSION INHIBITOR IN STAINLESS STEEL ORTHODONTIC WIRE

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ABSTRACT

Objective: To determine the effectiveness of watermelon rind extract as corrosion inhibitor in stainless steel orthodontic wire.

Methods: This research was a laboratory experimental research with post-test control group design. Samples used in this experiment were 40 pieces of stainless steel orthodontic wire that were divided equally into four groups with artificial saliva as the medium, one control group without extract addition, and three treatment groups with various concentrations of watermelon rind extract, namely 200 ppm, 600 ppm and 1000 ppm. Measurement of the corrosion rate was carried out by using an eDAQ potentiostat and the result of data analysis were presented in tables and diagrams.

Results: The highest corrosion rate occurred in control group, while in treatment groups the corrosion rate decreased significantly, with inhibitor effectiveness raised along with the increasing concentration. The most effective concentration was 1000 ppm with inhibitor effectiveness value of 46.12%.

Conclusion: Watermelon rind has the inhibiting effect on stainless steel orthodontic wire's corrosion rate.

Keywords: Corrosion, Orthodontic wire, Watermelon rind

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INTRODUCTION

The term malocclusion was first invented by Guilford, it is defined as an abnormality that causes disfigurement or impedes function and requires treatment. The malocclusion might be associated with one or more of the following: malalignment of individual's teeth in each arch, malrelationship of the dental arches relative to the normal occlusion: in anteroposterior, vertical, or transverse planes. The orthodontic treatment aims to adjust the position of teeth to the right dental arch [1].

Stainless steel, Ni-Ti and Ni-Cr are the most common materials used in orthodontic wire fabrication. However, several studies show that nickel and chromium as the main compounds of these alloys have proven to be allergenic. Humidity, temperature and normal flora of the oral cavity could create a destructive environment for the orthodontic wire [2]. In a long term, saliva as electrolytes could react to these alloys and causes electrical induction and corrosion, which leads to the occurrence of metal ions release to the oral cavity [3]. This may result in pathologic reaction which appears as local hypersensitivity with symptoms similar to those of bacterial gingivitis. Some research have reported the cytotoxic and carcinogenic characteristics of nickels which are related to asthma and hypersensitivity [2]. Metal ions release also affects the orthodontic treatment mechanically since corrosion escalates the friction on wire surfaces which then reduces the free-sliding movements required in orthodontic treatment [4].

Corrosion process cannot be avoided, but the rate could be slowed down. One way to reduce the corrosion rate is by applying organic inhibitor. Organic inhibitor is the best option for reducing corrosion rate in orthodontic treatment because of its safe, natural, and biocompatible characteristics [5]. Watermelon rind (*Citrullus lanatus*) is generally considered as organic waste and often gets thrown away despite its potential as organic inhibitor. Watermelon rind contains several heteroatoms and covalent-bonded atoms like oxygen and nitrogen, which allow the ionic interaction with metal surface, thus could be functioned as corrosion inhibitor [6]. Odewunmi et. al stated that watermelon rind also contains *L-citrulline* which could decrease corrosion rate due to its antioxidant property [7]. Numerous studies have proven the effectiveness of watermelon rind as an economic bio sorbent in solutions containing

nickel (Ni), cobalt (Co) [8], chromium (Cr) [9] and cadmium (Cd) [10]. This is caused by functional groups consisted in watermelon rind, which are hydroxyl (cellulose) and carboxyl (pectin) that could easily bond with metal ions [11].

An experiment done by Hera and Johnsons has proven that corrosion rate of stainless steel orthodontic wire is relatively faster than other kinds of orthodontic wire [12]. Hence, the purpose of this study is to determine the effectiveness of watermelon rind extract as corrosion inhibitor in stainless steel orthodontic wire.

MATERIALS AND METHODS

Material

This research was a laboratory experimental research using the post-test control group design. This research was done from May to June 2017 in several laboratories in Hasanuddin University, which are Phytochemical Laboratory and Biopharmaceutical Laboratory of the Faculty of Pharmacy, Biochemistry Laboratory and Integrated Chemistry Laboratory of the Faculty of Science.

Method

First, watermelon rinds were chopped into small pieces and rinsed under running water before dried with herbal dryer machine for two days in temperature of 50 °C. After that, maceration extraction method were done by soaking the watermelon rind in 70% ethanol for two days, then the solution was filtered with filter paper and evaporated using rotary evaporator to obtain the extract of watermelon rind. Four solutions were then made by diluting watermelon rind extract in artificial saliva solution with various concentration of watermelon rind extract: 0 ppm (control group), 200 ppm, 600 ppm and 1000 ppm.

Samples used in this experiment were 40 pieces of rectangular stainless steel orthodontic wire with the size of 17x25 (0.017 "x 0.025") and 65 mm in length for each wire. The wires were divided equally into four groups and were soaked in four solutions with various concentration of watermelon rind that were made before. Measurement of the corrosion rate was then carried out by using an eDAQ potentiostat and EChem software v.2.1.2. The data resulted by the software was then moved into Microsoft Excel and was

processed by making a Tafel graphic to attain the density of corrosion current. The corrosion rates of each wire were then calculated using equation below:

$$R_{mpy} = \frac{0.13 I_{corr} E}{\rho}$$

Where:

R_{mpy} : Corrosion rate (milli inch/year)

I_{corr} : Density of corrosion current ($\mu A/cm^2$)

E: Equivalent weight of the material (g)

ρ : Material density (g/cm^3)

RESULTS AND DISCUSSION

The average corrosion rate of stainless steel orthodontic wires after soaked in solutions containing various concentration of watermelon rind extract are shown in table 1.

Table 1: Difference between average corrosion rates of stainless steel wires soaked in control solution and watermelon rind solution with concentration of 200 ppm, 600 ppm and 1000 ppm

Treatment groups	N (%)	Corrosion rate of stainless steel wire (mpy)	Normality test	Comparison test
		mean±SD	p-value*	p-value**
Control	10 (25%)	2.630 x10 ⁻⁵ ±0.018 x10 ⁻⁵	0.100	0.000
200 ppm	10 (25%)	2.107 x10 ⁻⁵ ±0.013 x10 ⁻⁵	0.001	
600 ppm	10 (25%)	1.717 x10 ⁻⁵ ±0.019 x10 ⁻⁵	0.011	
1000 ppm	10 (25%)	1.417 x10 ⁻⁵ ±0.022 x10 ⁻⁵	0.035	

*Shapiro Wilk test: p>0.05; normal data distribution, **Kruskal Wallis test: p<0.05; significant

Table 1 shows that the highest corrosion rate was found in control group, with average corrosion rate of 2.630x 10⁻⁵ mpy, whilst the lowest corrosion rate was found in 1000 ppm group with average corrosion rate of 1.417x 10⁻⁵mpy, the average corrosion rate in 200 ppm group reached 2.107x 10⁻⁵mpy and 600 ppm group reached the corrosion rate of 1.717x 10⁻⁵ mpy.

The results of Saphiro Wilk's normality test are also shown in table 1 which shows the value of p>0.05 only in control group while other groups show the value of p<0.05. This means that data in control

group is distributed normally, otherwise the data in other groups are not. Ergo, the result data does not meet the requirement of parametric test where all of the data must distribute normally. Kruskal Wallis' non-parametric test was used instead for this reason, resulted the value of p=0.000 (p<0.005). This shows that there were significant differences of corrosion rate between each sample groups. However, to know further comparison between each of the groups, the post hoc analysis was done in which the results are explained in table 2.

Table 2: The post hoc test results of average stainless steel wires corrosion rates after soaked in control solution and watermelon rind solution with concentration of 200 ppm, 600 ppm and 1000 ppm

Treatment group (i)	Comparison (j)	Mean difference (i-j)	p-value
200 ppm	Control	-0.523 x10 ⁻⁵	0.000
	600 ppm	0.390 x10 ⁻⁵	0.000
	1000 ppm	0.690 x10 ⁻⁵	0.000
600 ppm	Control	-0.913 x10 ⁻⁵	0.000
	1000 ppm	0.300 x10 ⁻⁵	0.000
1000 ppm	Control	1.213 x10 ⁻⁵	0.000

*Post Hoc Test: Mann Whitney test: p<0.005; significant

Table 2 shows the results of the post hoc test of the average stainless steel wire's corrosion rate in watermelon rind extract solution with concentration of 200 ppm, 600 ppm, 1000 ppm, and in control group. Based on Mann Whitney's post hoc analysis results, there was a significant difference between average corrosion rate in control group and all other groups. There were also significant differences

between 200 ppm group with 600 ppm and 1000 ppm groups, also between 600 ppm group and 1000 ppm group. This suggests that soaking the stainless steel wire in watermelon extract solution has an effect on inhibiting corrosion rate. The effectiveness of corrosion inhibition of each concentration is represented by its effectiveness value (table 3).

Table 3: Calculation results of the corrosion inhibitor effectiveness value of 200 ppm, 600 ppm and 1000 ppm watermelon rind extract

	Average corrosion rate in control group (X_a)	Average corrosion rate with inhibitor (X_b)	Difference ($X_a - X_b$)	Inhibitor effectiveness value (EI %)
200 ppm	2.630 x10 ⁻⁵	2.107 x10 ⁻⁵	0.523 x10 ⁻⁵	19.89%
600 ppm	2.630 x10 ⁻⁵	1.717 x10 ⁻⁵	0.913 x10 ⁻⁵	34.71%
1000 ppm	2.630 x10 ⁻⁵	1.417 x10 ⁻⁵	1.213 x10 ⁻⁵	46.12%

Table 3 shows the inhibitor effectiveness value of watermelon rind extract with concentration of 200 ppm, 600 ppm, and 1000 ppm. The highest effectiveness value was found in 1000 ppm concentration group with effectiveness value of 46.12%, the lowest effectiveness value was found in 200 ppm concentration group with effectiveness value of 19.89%, whereas in 600 ppm concentration group the effectiveness value of the inhibitor reached 34.71%. The

comparison of effectiveness values of these three treatment group is represented by the bar chart in fig. 1.

It can be seen in fig. 1 that the inhibitor effectiveness value increased along with the increasing of concentration. This shows that the concentration of watermelon rind in the solution influences the corrosion inhibition effect, and the most effective watermelon rind

extract solution in this study is the solution with concentration of 1000 ppm.

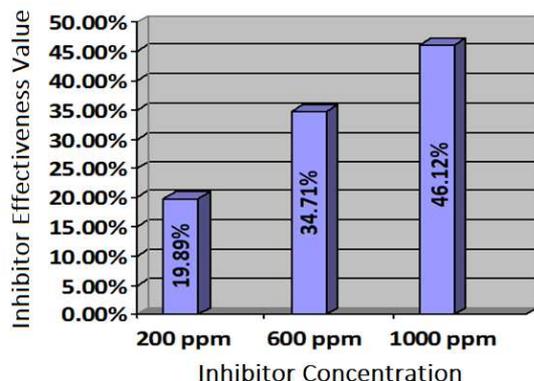


Fig. 1: Bar chart shows the comparison of the inhibitor effectiveness values of watermelon rind extract with concentration of 200 ppm, 600 ppm, and 1000 ppm

The results showed that the average corrosion rate in control group was 2.630×10^{-5} mpy, whereas the lowest corrosion rate was found in the group with 1000 ppm of watermelon rind concentration, reaching 1.417×10^{-5} mpy of corrosion rate. The corrosion rate of the sample group with 200 ppm of watermelon rind extract reached 2.107×10^{-5} mpy, and sample group with 600 ppm of watermelon rind solution reached 1.717×10^{-5} mpy. From these results, it can be concluded that the immersion of stainless steel orthodontic wire in a solution of watermelon rind extract has an influence in inhibiting the corrosion rate. There were also significant differences between 200 ppm group with 600 ppm and 1000 ppm groups, and between 600 ppm group and 1000 ppm group. This indicates that the amount of watermelon rind extract concentration contained in the solution influences the corrosion inhibition effect.

Extracts of natural materials, especially compounds containing N, O, P, S, and atoms with free electron pairs can serve as ligands, i.e. ions that have a pair of electrons or more that can be donated [13]. Watermelon rind contains pectin that consists of carboxylic and hydroxyl groups with free electron pairs, thus metal cations may be attracted and bind to the pectin, forming a pectin-metal complex [14]. Characterization analysis with FT-IR by Lakshmiopathy *et al.* showed that watermelon rind contains the -OH, -C=O, -COO and -C-O groups which are functional groups that could easily bond with metals [10]. Watermelon rind also contains L-citrulline, an antioxidant that consists of heteroatoms (nitrogen and oxygen) and an aromatic ring in its chemical structure. The chemical arrangement of L-citrulline enables the occurrence of ionic interactions with metal surfaces and can function as a corrosion inhibitor [7].

Corrosion inhibitors are substances or mixtures that in low concentration and in aggressive environment inhibit, prevent or minimize the corrosion. Generally, inhibitor works by chemical adsorption (chemisorption) or by combination between inhibitor ions and metal surfaces, which then forms a protective thin film with inhibitor effect. The inhibitor could also work by leading a formation of a film by oxide protection of the base metal, or by reacting with a potential corrosive component present in aqueous media and generating a complex [15]. According to Yatiman, because the surface of the covered metal is proportional to the concentration of the inhibitor, the concentration of the inhibitor in the solution could greatly affects the effectiveness of the inhibitor. The greater the concentration of the inhibitor in solution, the better the corrosion inhibiting effect [16].

Hussin and Kassim stated that the addition of an appropriate concentration of inhibitor will effectively inhibit the corrosion process, but if the applied concentration is excessive it can cause the inhibitor molecules on the metal surface to be pulled back into the

environment of the solution, which means the protective layer formed on the steel surface would be decreased and the corrosion rate that occurs would be increased instead [17].

In this study, the application of watermelon rind extract as corrosion inhibitor with concentrations of 200 ppm, 600 ppm, and 1000 ppm are still effective in inhibiting the stainless steel orthodontic wire's corrosion rate. The results indicated that the greater the concentration of the solution, the greater the inhibitor effectiveness value. The inhibitor effectiveness value was 19.89% in 200 ppm group, 34.71% in 600 ppm group, and the highest effectiveness value reached 46.12% in 1000 ppm group. This shows that the most effective concentration used in this study is 1000 ppm.

Although the results proved that the addition of watermelon rind extract of 1000 ppm in the solution was effective in obstructing the stainless steel wire's corrosion, the effectiveness value of 46.12% is still relatively low compared to the effectiveness value of other natural inhibitors in previous studies. In research conducted by Haryono G, Sugiarto B, Farid H and Tanoto Y by adding various kinds of plant extracts in sea water media to inhibit corrosion in iron, the effectiveness value of 57.84% was obtained with the use of coffee extract, 63.75% with tobacco extract and 87.22% with pine resin extract. The study also used gambir leaf extract, but was not considered effective in inhibiting corrosion since the effectiveness value only reached 11.34% [13]. Another experiment executed by Priyotomo G and Nuraini L by adding 6000 ppm of bilimbi leaf extract into an acid hydrochloric solution has showed decreasing corrosion rate of carbon steel up to 94.5% [18]. A study done by Hamdani S and Elta MS using papaya leaf extract to inhibit the corrosion rate of carbon steel schedule 40 grade B ERW showed the effectiveness value of 78.49% in freshwater medium and 78.63% in sea water medium [18]. Low inhibitor effectiveness value of watermelon rind extract in this experiment is possibly because 1000 ppm as the largest concentration used in this study is not the optimum concentration of watermelon rind extract as corrosion inhibitor.

CONCLUSION

Based on the results of the experiment which has been done, the researcher concluded that watermelon rind extract has the inhibiting effect on stainless steel orthodontic wire's corrosion rate. Watermelon rind extract with concentrations of 200 ppm, 600 ppm and 1000 ppm showed significantly increasing effectiveness value along with the increasing of the concentration of inhibitor. The most effective concentration in this research is 1000 ppm with effectiveness value of 46.12%.

Authors suggest that in the future, further research with more variation of concentration could be done to attain the precise optimum concentration of watermelon rind as an organic corrosion inhibitor. Similar experiment with watermelon rind extract also needs to be done on other types of metals that are often used in orthodontic treatment. Different trial methods are also needed to obtain more information and to understand more about the exact working mechanism of watermelon rind extract as corrosion inhibitor.

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AUTHORS CONTRIBUTIONS

All the authors have contributed equally

CONFLICT OF INTERESTS

There are no conflict of interest in this study

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