Study of Corrosion Resistance of Ferritic Stainless Steel 444 on Artificial Saliva

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Abstract: ***Magnetic dental veneers composed of ferritic and austenitic stainless steels are usually connected by root covers made of different dental metals in the oral cavity. Because the device is designed to serve as a seat or base for the artificial tooth to attach to the root that is already embedded in the gum, the magnetic tooth coating device is subject to mechanical interactions such as friction, which can cause wear, along with corrosive attacks from contact with saliva solution. In this study focuses on assessing the ferritic corrosion resistance of type 444 stainless steel in artificial saliva solutions with different surface roughness. Surface roughness was controlled with 80, 400 and 600 grid sandpaper and then finished with velvet cloth polishing. Tests have been carried out by determining the surface roughness value first, then the corrosion test using the Weight Loss and Linear Polarization methods. The results show that a smoother surface (0.00022 mm / y) provides a lower mean corrosion current compared to a coarser surface (0.00057 mm / y). A rougher metal surface will cause a potential difference on the surface, the rougher the greater the potential difference between the surfaces, this causes the surface to have a tendency to become corroded anode***

Keywords: Corrosion, Stainless Steel, Artificial Saliva

1. Introduction

Almost all steels performed by machining process experience surface roughness that will be visible when viewed with the naked eye and the effect of catalysts on chemical reactions. For example, concerning corrosion, metal surfaces generally oxidize when in the air at room temperature and form a very thin oxidation layer (dull layer). In everyday language, corrosion is known as a case that is considered a common enemy by the public. Corrosion is an electrochemical reaction process that has natural properties and takes place spontaneously. Corrosion cannot be prevented or stopped at all. Corrosion can only be controlled or slowed down to facilitate the process of damage that will occur. Most equipment or materials that are often used in the environment have corrosive properties(1).

Such as the use of metal materials for medical implant purposes in the early 19th century in bone repair. Then, biomaterials in the form of implants (ligaments, vascular grafts, heart valves, intraocular lenses, dental implants, etc.) And medical devices (pacemakers, biosensors, artificial hearts, etc.) It is reported to be widely used to replace and/or restore the function of tissues or organs experiencing trauma or degeneration. Today, metal materials dominate in orthopedic surgery with their application in commercial orthopedic devices(2). Whatever material is used, wear problems can be predicted in any replacement connection and consequently implant loosening as a result of high wear coefficient or low wear resistance from friction(3).

The roughness of the surface of the workpiece affects the corrosive attacks that occur. Rougher workpieces will be easily corrosion-prone compared to metals with smooth surfaces. The parameters of the machining process will affect the roughness of the surface of a workpiece. Surface roughness is a product quality parameter of an ingredient(4).

Based on the explanation above, Used workpieces with various variations of surface roughness resulting from the sanding process to observe the rate of corrosion in artificial saliva solution with pH ±7.

This study was conducted to determine the effect of surface roughness on the corrosion rate of Ferritic Stainless Steel type 444 in artificial saliva solution, it is hoped that this research can add reference material and help overcome corrosion problems that occur in the medical world, especially in the field of dentistry.

Type 444 is a low carbon, low nitrogen, ferritic type stainless steel that provides superior hole and slit corrosion resistance to most stainless steels. Any application requiring superior corrosion resistance and resistance to corrosion cracking chloride voltage is ideal for this alloy. Its current uses include food processing, brewery, and winemaking equipment; hot water tanks, heat exchanger tubes, and automotive components. Ferritic type 444 stainless steel has gained attention as a potential material for use in medicine, as it does not contain much nickel(5)(6).

Artificial saliva acts as a buffer solution or buffers in the medium or as a substitute for saliva function. The use of artificial saliva is important to maintain the pH to stay within the normal range. Metal ions as anodes and H^+ ions as cathodes(7).

This paper will be conducted more research on the corrosion resistance of Ferritic Stainless Steel type 444 in Artificial Saliva solution with different variations of roughness, this study was conducted to determine the rate of corrosion due to different surface roughness.

The purpose of this study is to know and analyze the resistance and rate of corrosion in stainless steel material type 444 and know the influence on the difference of surface roughness to the rate of corrosion. Then in the future, the results of this study can be one of the references to biomaterials in the field of magnetic implant teeth, and can practically be used as a consideration for the field of implant dental manufacturing industry to get a good product and resistant to good corrosion.

2. Methods

The material used in this study is Stainless Steel type 444, Chemical composition testing using Spectrotest tool TXC03 with a testing method according to ASTM E 415-15, test results are shown in Table 1:

Table 1: Chemical composition of SS 444

W (%)	C	Si	Mn	P	S	Cr	Mo	Ni	Cu	Fe
Max	0.03	0.54	0.09	0.03	0.01	18.57	1.71	0.15	0.02	78.5

2.1 Material Preparation

Specimens of test objects are made with the following stages:

1) Provision of metal materials
   The metal chosen is a type of stainless steel type 444, because it contains fewer nickel elements than the type of stainless steel 316L commonly used in the medical world.
2) Cutting spesimens
   Stainless steel metal available on the market in the form of thin plates with a thickness of 2 mm, then cut using a cutting machine with a size of 23 mm x 13 mm as much as 9 pieces.
3) Polishing
   To give differences in roughness on the surface of the specimen, polishing is done using silicon carbide paper with a grid size from 80,400, to 600.
4) Finishing
   Specimens that have reached the stage of sanding then cleaned from the rest of the dirt that is still attached with distillation water and then dried with a cloth.

For the manufacture of saliva a lot of formulas and methods have been done, one of which is by following from previous research, with the composition as follows:

Table 2: Artificial saliva composition for 1L(8)

Larutan	gr/l
NaHPO4	0,26
KSCN	0,33
NaCl	6,7
KH2PO4	0,200
KCL	1,20
NaHCO3	1,50
HCL	Balance
Aquades	Balance

All materials are weighed according to weight and mixed into one measuring glass and then dissolved with distilled water up to 1 liter, stirring the solution using magnetic steering tools in the laboratory metallurgy and physics Department of Mechanical Engineering UNDIP until the solution becomes homogeneous and HCL is dropped little by little, the pH change is controlled using a digital pH meter until the pH solution becomes ±7.

2.2 Roughness Test

Roughness testing was conducted to determine the influence of different roughness on the surface on the rate of corrosion occurred, Tools used Mitutoyo model SURFTEST SJ-210 measurement range of 17.5 mm (X-axis) and 260 µm (Z-axis), where testing was conducted in the Metrology Laboratory of the Department of Mechanical Engineering UNDIP. In this measurement, 9 specimens were tested with each specimen tested at 3 different points. Roughness measurement parameters are measured by calculating the difference between the deviation of the original surface from the ideal surface by the amount of distance. Large deviations can be known that the surface is rough, and vice versa small deviations indicate that the surface is smooth. To measure surface roughness can use a method of direct contact on the surface of the material. The working principle of this method uses a needle that runs along the surface of the material. The needle has its size depending on the accuracy of the surface roughness tool used. Along the way, stylus measurements move up and down following the roughness of the surface.

Figure 1: Roughness test scheme

2.3 Corrosion study weight loss method

Specimen preparation and corrosion test by weight loss method are conducted according to the standards of ASTM G1. Which stainless steel plate is cut with a size of 23 mm x 13 mm as much as 9 pieces, then rubbed using sandpaper starting from the grid 80,400 and 600, before starting immersion specimen must be cleaned first with distilled water to remove the rest of the dirt. Testing was conducted in 3 variations of roughness: 80, 400, 600. Calculation of the rate of corrosion loss of weight by weighing each specimen of the initial weight. Specimens are immersed in an artificial saliva solution (pH 7) using plastic vessel containers, the temperature of the solution is modified according to the temperature in the mouth which is ± 36°C which is controlled automatically using a digital thermostat. Data retrieval is done once a week, each specimen is weighed again using an analytical balance sheet with a precision rate of up to 0.0001 gr so that later get the value of lost weight (W), they can calculate the rate of corrosion with the equation following the standard ASTM G1(9) :

$$CR : \frac{KxW}{AxTxD} \qquad (1)$$

Where Q is notation explanation, n is notation explanation, and D is also notation explanation, etc.
Where CR is notation explanation, K is Constant (mm/y), W is weight lost (gr), A is Surface area exposure (cm^2), T is Length os soaking time, and D is Density (gr/cm^3).

2.4 Electrochemical Corrosion Study (Linear Polarization Method)

The equipment used in this test is a 3-electrode cell connected to the Autolab PG STAT 302 M potentiostat equipment so that the current generated at each given voltage can be recorded by a computer that already has NOVA Autolab software in it. The steps taken in the corrosion rate testing process are as follows:

1) Preparing artificial Saliva Solution as much as 1.5 liters in the middle of pH 7 (±0.1).
2) Preparing potential source equipment, namely a set of PotensiostatAutolab PG STAT 302 M connected with a computer and NOVA software.
3) Put the reference electrode in the form of Ag / AgCl element, auxiliary electrodes in the form of platinum, and work electrodes or test specimens into beaker glasses that already have Artificial Saliva solution in it using clamp pliers. Attempted at the time of laying each electrode does not intersect with each other.
4) In NOVA Software connected with a set of PotensiostatAutolab PG STAT 302 M that serves as a potential source set at -500 mv up to +500 mv.
5) Once all is done set, click the "Start" sign on the bottom left to start testing the corrosion rate. This scanning process aims to find the potential value that will be used to run the specimen testing.
6) Once the potential value is found, next nova software in running on potential -500 mv up to -100 mv. During the running process nova software will automatically display the creation of tafel graphics created gradually.
7) Once the Tafel graph is formed, enter the material density data tested (g/cm^3), the weight of the metal atom (g/mol), and the surface area of the test material (cm^2) in the column in the bottom right corner. Data – this data is used by the software to calculate its corrosion rate.

3. Result and Discussion

3.1 Surface Characterization

Characterization of the surface by determining the roughness of the surface, which has been done grinding and coping using silicon carbide paper with sizes 80, 400, and 600. The roughness measurement results are shown in Table 3. The result of the surface roughness difference data will be used as additional data for corrosion resistance testing.

Table 3: Roughness Test Result Data

No	Grid Polishing	Ra (µm)
1	80	0.624
2	400	0.196
3	600	0.146

Figure 2 shows a comparison curve of surface roughness on Feritic Stainless Steel 444 material after grinding and polishing.

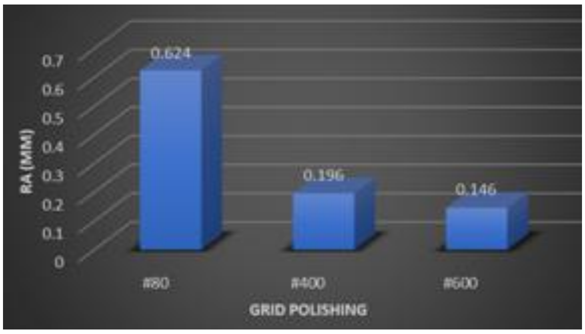

Figure 2: Roughness Test Results

3.2 Corrosion Test Weight Loss Test Method

The corrosion behavior of SS 444 is studied by calculating the loss of specimen weight against the length of dyeing time in the artificial Saliva solution, the pH content of Saliva will change with the dyeing process for one week (7 days), with the temperature of the solution that remains maintained at 36-37 °C by using the heater as a heater and thermostat as an automatic temperature controller. The pattern was observed for cumulative sample weight loss versus dyeing time. Figure 2 shows the relationship between roughness and Ferritic Corrosion Rate Stainless Steel 444 after graphically exposed to artificial Saliva solution. Table 4 shows the corrosion rate (CR) of three different types of surface roughness directly proportional to the addition of testing time (t). The test sample loses weight but the result of corrosion rate will be different every week of data retrieval, because it can be a process of cleaning and preparation of test samples that are not maximal, and the surface roughness that has begun to change.

Table 4: Weight loss corrosion rate test result data

Time (weeks)	Corrosion Rat (mm/y)		
	0.624	0.196	0.146
0	0	0	0
1	$22x\ 10^{-5}$	$19x\ 10^{-5}$	$15x\ 10^{-5}$
2	$19x\ 10^{-5}$	$18x\ 10^{-5}$	$15x\ 10^{-5}$
3	$64x\ 10^{-6}$	$30x\ 10^{-6}$	$18x\ 10^{-6}$
4	$12x\ 10^{-5}$	$91x\ 10^{-6}$	$51x\ 10^{-6}$
5	$23x\ 10^{-5}$	$18x\ 10^{-5}$	$15x\ 10^{-5}$
6	$25x\ 10^{-6}$	$17x\ 10^{-6}$	$13x\ 10^{-6}$
7	$71x\ 10^{-6}$	$32x\ 10^{-6}$	$12x\ 10^{-6}$
8	$70x\ 10^{-6}$	$50x\ 10^{-6}$	$31x\ 10^{-6}$

In figure 3, it can be seen based on the weight loss data that occurred in each sample obtained the highest to lowest corrosion rate, at a surface roughness of 80 (0.624 µm), which is the highest corrosion rate occurred in week 5 of 0.00023 mm/y because the rougher stainless steel metal surface will cause potential differences and tend to be a corroded anode. The rougher surface of metal causes inhomogeneity on the surface, which is one of the main causes of corrosion. Uneven metal surfaces will facilitate the occurrence of charge poles (positive and negative charge), which will eventually act as an anode and cathode in electrochemical reactions. While the lowest corrosion rate occurred in the 7th-week sample surface roughness of 600 (0.146 µm), which is 0.000012 mm/y.

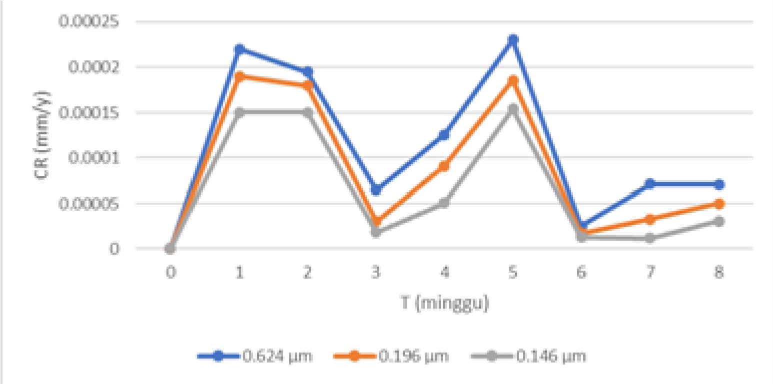

Figure 3: Corrosion rate vs time

At the beginning of dyeing the specimen into the saliva solution, the condition of the specimen surface is still clean so that the corrosion rate is fast and then in the 3rd week when it begins to corrode so that the surface of the specimen begins to be covered with corrosion products causing the rate of corrosion has decreased, this causes the blocking of ion transfer to the electrolyte. This problem is repeated in the 4th week until the next.

Figure 4: Sample specimen before corrosion test

Figure 5: Sample specimen after corrosion test

3.3Electrolysis Method Corrosion Test (Linier Polarization)

From linear polarization test results, the corrosion rate at higher surface roughness (0.624 µm) is directly proportional to the increasing corrosion rate compared to the smaller roughness (0.146 µm), which proves that the difference in roughness on the surface will greatly affect the corrosion rate in Stainless Steel type 444 materials. In the sense of the word, the smoother the surface, the lower the corrosion rate, and vice versa. The test result data is presented in table 3.3, and figure 3.4 shows the relationship between roughness and Ferritic Corrosion Rate Stainless Steel 444 after exposure to artificial Saliva solution is presented graphically using the linear polarizing method.

Rougher stainless steel metal surfaces will cause potential differences and tend to be corroded anodes. The rougher surface of metal causes inhomogeneity on the surface, this facilitates the occurrence of corrosion. Uneven metal surfaces will facilitate the occurrence of charge poles (positive and negative charge), which will eventually act as an anode and cathode in electrochemical reactions. Smooth and clean metal surfaces will cause corrosion difficult to occur because it is difficult for the poles to act as an anode and cathode. Therefore, the rate of corrosion will increase as the roughness of the metal surface increases. This is similar to the results of previous research(4) which states bring when the value of surface roughness increases, then the rate of corrosion will also be greater (directly proportional).

The rougher the metal surface will cause a potential difference on the surface, the rougher the greater the potential difference between the surfaces, this causes the surface to tend to become a corroded anode.

Table 5: Corrosion rate test result data (Linier Polarization)

Sample	Ecorr (V)	Icorr (A/cm²) x 10^{-8}	CR (mm/y) x 10^{-5}
A1	-0.051	5.5	58
C1	-0.039	4.28	55
B2	-0.036	3.39	39
C2	-0.005	1.41	21
B3	0.0013	1.28	17
C3	0.0400	1.47	26

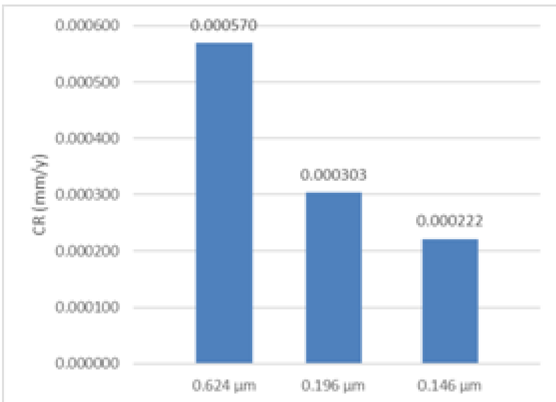

Figure 6: Linear polarizing corrosion rate

4. Conclusion

Based on the results of research and data calculation obtained several conclusions. Surface roughness greatly affects the corrosion rate of Ferritic Stainless steel material type 444. Where the greater the roughness value of Ferritic material Stainless steel type 444, the higher the rate of corrosion that occurs. Because the rougher metal surface will cause a potential difference on the surface, the rougher the potential difference between the surfaces, causes the surface to tend to become a corroded anode. The best result (lowest corrosion rate) obtained from the weight loss method is at a surface roughness of 0.146 μm with a corrosion rate of 0.000012 mm/y. The best result (lowest corrosion rate) obtained from electrolysis method (Linear Polarization) is at surface roughness of 0.146 μm with corrosion rate of 0.000222 mm/y, and highest corrosion rate at surface roughness variation of 0.624 μm by 0.000570 mm/y.For further research can add testing using SEM EDX to determine the type and form of corrosion that occurs on the surface of the specimen so that in the future it can be used to determine corrosion prevention or control measures.

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