Influence of surface roughness on corrosion behaviour of 316L stainless steel in artificial saliva and body fluid

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Abstract. The influence of surface roughness on the corrosion behaviour of 316L stainless steel in artificial saliva and body fluid was investigated in this study. Potentiodynamic polarization and EDAX composition test were used for observing the passive layer activity. The quality difference on steel 316LSS in saliva and blood may cause difference on the formation of passive layer. The result shows that the scratch formation which got rougher caused the uneven distribution of passive layer on the surface which caused the passive layer protection not optimum.

Keywords: corrosion, biomaterial, surface roughness, passive layer

1. Introduction
Biomaterial is commonly known as synthetic material which is used to replace or fix the function of body system continuously or merely have contact with body fluid [1,2]. Biomaterial should be no harmful, nontoxic, and non-carcinogenic response [3]. These requirements eliminate other technical materials that can be used. Besides that, biomaterial should have adequate physical and mechanical characteristics to function as replacement or multiplier in body tissues.

Metal implant corrosion is a negative effect for the body tissues around it [2,4,5]. Corrosion also causes orthodontic wire surface become rough, weaken orthodontic wire use, and the cause the release of metal elements or the compounds which results in colour change of the soft tissues around the wire and allergic [6,7]. Metal surface in human body will be damaged electrochemically because of the interaction with the environment. In this case, the environment is the human body liquid or saliva. The environment contains water, dissolved oxygen, complex organic compound, various chloride ions, sodium, protein, and others [8]. SS-316L has lower hardness value compared to other implant materials like titanium alloy. Thus, it is easy to get scratched and deformed when loaded. However, the metal has resistance to high oxidation in the air and in normal environment temperature. This is because there is passivation phenomenon. Passivation is a phenomenon where thin layer of chrome that protects steel material is formed when it meets oxygen [9,10].
This layer is a transparent chrome oxide compound. Therefore, it has to be found out whether there is thin layer on steel SS316L or not.

This thin layer is a chrome oxide compound which is transparent, very thin and in form of film, insoluble, and keeps the material looks shiny. Each kind of body fluid will lead to different thin layer of 316LSS formed. The quality difference on steel 316LSS in saliva and blood may cause difference on the formation of passive layer. Characterization of passive layer on 316LSS in artificial saliva and blood was conducted in this study. The surface roughness of steel varies in those two kinds of artificial liquid. Characterization of passive layer was conducted with analysis of potentiodynamic polarization test result.

2. Method

2.1 Sample Preparation
Composition of chemical steel 316LSS was %wt: 0.03C, 1.00 Si, 2.00Mn, 0.045P, 0.03S, 12.00Ni, 16.00Cr, 2.00Mo. The specimen roughness was categorized based on polishing process treatment with 100, 500, 1000 and 2000 grid. The scratching was done once and one way. Then, the surface roughness of the polishing grid was measured with surface roughness testing tool of Mitutoyo type SJ-201. Then, the value of polishing grid was equalized with the mean roughness value (Ra) which was in µm. 100 grid was equal with 1.533 µm, 500 grid was equal with 0.900 µm, 1000 grid was equal with 0.547 µm, and 2000 grid was equal with 0.133 µm. The specimen was in size of 40x10x3mm and was soaked in two kinds of solution which were artificial saliva and artificial blood. The composition of artificial saliva was 0.4 g NaCl; 0.4 KCl; 0.78 g NaH₂PO₄. 2H₂O; 0.005 g Na₂S. 9H₂O; 1 g CO(NH₂)₂; 0.81 g potassium chloride with pH 6.75. Artificial blood was represented with Hanks solution with composition of NaCl 0.80 g, CaCl₂ 0.14 g, KC1 0.40g, NaHCO₃ 0.35 g, Glucose 1.00 g, NaH₂PO₄ 0.10 g, MgCl₂.6H₂O 0.10, Na₂HPO₄.2H₂O 0.06, MgSO₄.7H₂O 06 g L⁻¹ with pH 7.4. All the samples which had been soaked in the two kinds of solutions were stored in incubator with temperature 37°C during 7, 14, and 28 days.

2.2 Potentiodynamic Polarization Measurement
Potentiodynamic polarization was run with AUTOLAB PGSTAT 128N. The test was electrochemical testing with 3 electrode cells principle. Ag / AgCl (3 M KCl) was the electrode reference, platina was auxiliary electrode, and the work piece was the working electrode. The test was done along -1V to 1V with Open Circuit Potential and scan rate 0.001 V s⁻¹.

3. Result and Analysis

Based on Figure 1 and Table 1, it can be seen the characteristic of passive layer based on polarization of the steel 316LSS in artificial body fluid. Basically, polarization curve has three zones which are active, passive, and transpassive [11,12]. However, transpassive zone was not visible in the result of polarization. It is because there will not be transpassive reaction of stainless steel with high potential which is more than 1.750 V in human body application.

The low value of Ep towards Ecorr on a material means that the material tends to be passive [13]. The passivation rate is the most important properties in passive layer formation on stainless steel. In Figure 1, there is a specimen with surface roughness of 100 grid with the highest Ep value compared to other specimen grids. This indicates that the material is easy to be corroded compared to smoother specimens. This is caused by the range between E corr and Ep.
Figure 1. The tafel plot of steel 316LSS in artificial body fluid solution with variation of surface roughness a) 100 grid, b) 500 grid, c) 1000 grid, d) 2000 grid

| Variation of surface roughness (grid) | Ep (V)  | Ecorr (V) | Er (V) |
|--------------------------------------|---------|-----------|--------|
| 100                                  | 0.962   | -0.04754  | 0.947  |
| 500                                  | 0.8822  | -0.21643  | 0.8398 |
| 1000                                 | 0.9927  | -0.2488   | 0.9466 |
| 2000                                 | 0.7813  | -0.34768  | 0.7402 |

Figure 2. The tafel plot of steel 316L SS in artificial saliva with variation of surface roughness of: a) 100 grid, b) 500 grid, c) 1000 grid, d) 2000 grid

Figure 2 and Table 2 show that the rougher the surface of steel 316LSS is, the easier the corrosion happens. The highest Icorr value was obtained on the specimen with surface roughness of 100. The lowest value was obtained on the specimen with surface roughness of 2000 grid. These results indicate that the surface roughness with 100 grid causes the corrosion rate to be higher compared to other specimens. With surface roughness grid of 2000 polish, the Icorr value is the
lowest compared to the other specimens. This is caused by the formation of more perfect passive layer compared to other specimens [14].

Table 2. The Polarization zone on steel 316L SS in artificial saliva

| Variation of Surface Roughness (grid) | Ep (V)  | Ecorr (V) | Er (V)  |
|--------------------------------------|---------|-----------|---------|
| 100                                  | -0.1915 | -0.21634  | -0.1898 |
| 500                                  | -0.2349 | -0.32229  | -0.2112 |
| 1000                                 | -0.13449| -0.34566  | -0.13100|
| 2000                                 | -0.17462| -0.34178  | -0.17226|

On potentiodynamic test, it was obtained that the lowest corrosion rate value was at 2000 polishing grid. This proved that the roughness and surface treatment influenced passive layer very much. Thus, the more perfect the passive layer is, the lower the corrosion rate is. The measured reaction between the implant surface and body fluid was very much determined by the characteristics of the material surface. The scratch formation which was getting rougher caused the uneven distribution of passive layer on the surface which indicated that the passive layer protection was not maximum. The specimen corrosion rate value with 100 surface roughness caused more zone with non maximum passive layer formation. This was because there was aeration during the immersion test in circulation of Hanks solution which made the bond between Cr and oxygen formed stronger Cr$_2$O$_3$ because bigger oxigen concentration and agitation will accelerate corrosion. In this case, it was the formation of oxide layer which was Cr$_2$O$_3$.

Table 3. The result of composition analysis with EDAX

| Sample Grid 2000 (day) | %wt |
|------------------------|-----|
|                        | O   | F   | Na  | S   | Cr  | Mn  | Fe  | Co  | Ni  |
| 7                      | 4.65| 12.33| 5.26| 1.55| 13.20| 2.51| 42.35| 13.7| 5.08|
| 28                     | 20.85| 3.08| 32.36| 11.26| 5.78| 1.54| 15.97| 6.27| 2.41|

Figure 3. The result of EDAX test on steel 316LSS with roughness of 2000 grid which was soaked for a) 7 days, b) 28 days in artificial saliva

The scratch formation which was getting rougher led to uneven distribution of the passive layer on the surface which resulted in non maximum passive layer protection. The artificial saliva fluid is
electrolite fluid that can trigger corrosion process. Chloride ion in saliva may damage the oxide layer on the metal surface which causes the metal ion release like iron, nickel, chromium, molybdenum, and titanium which are the important elements of biomaterial [15-17].

Nickel which was contained in 316LSS dissolved in saliva liquid. Thus, the duration of time when metal had contact with liquid influenced the metal ion release. Nickel ion highly tends to release regarding with the element structure at atomic level. Nickle atom is not related strongly with intermetallic compound [18]. This was proofed by EDAX test which was conducted with specimen with 2000 grid which was soaked for 7 days and 28 days in artificial saliva as shown in Figure 3 and Table 1. The data shows the decrease on the amount of nickel ion and chromium ion on steel 316LSS.

The biomaterial is related with the composition of each metal. The biomaterial composition is a factor which influences the metal ion release with released main ion which are iron, chromium, and nickel. The release of metal ion is influenced by biomaterial composition, the degree of saliva acidity, and the soaking time. The chromium functions as protector to corrosion and nickel functions to add strength, flexibility, and improve the corrosion resistance. In acid condition, the amount of ion H+ will get bigger. Thus, it is corrosive and can oxidate metal. This cause the metal in saliva more corrosive compared to if it is in body fluid. Metal ion which is released during the corrosion process is alloy chemical reaction which can influence quality, aesthetics, physical shape, and weaken the metal strength [2,19,20].

4. Conclusion
The surface of biomaterial 316LSS, which is rougher, will prompt the corrosion rate to increase. This is caused by the scratch formation, which is getting rougher causes uneven distribution of passive layer on the surface. Thus, passive layer protection is not maximum. Artificial saliva liquid is electrolite liquid, which can lead to corrosion process. Chloride ion in saliva and damage the oxide layer on the metal surface, which prompts the release of nickel and chrome ion in line with the soaking time addition.

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