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Optimization of the Effect of Electropolishing's Current Density and Time on Roughness, Microstructure and Corrosion Resistance

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Abstract

The surface roughness of medical, pharmaceutical, food and beverage equipment in direct contact with materials and products plays an important role in product quality, hygiene, equipment corrosion and ease of cleaning. The high surface roughness is feared as a place for the accumulation of process residues, products, and nesting of microbes such as pathogenic bacteria that degrade product quality. The purpose of this research is to investigate the parameters of the electropolishing process, namely the electric current density and the time of the electropolishing process. The electrolyte solution is a mixture of 35% sulfuric acid and 51% phosphoric acid with the electropolishing process temperature being maintained at 50°C, using a stainless steel as cathode, and the material being processed is AISI 316L. Characterization of electropolishing results include roughness, microstructure, and corrosion resistance.

***Keywords**: AISI 316L; electropolishing; Microstructure; potentiodynamic; roughness*

1. INTRODUCTION

1.1. Background

Nutritional food, habits and body conditions are strongly correlated with disease and lead to the need for drugs. Referring to the production system, drug production requires a process system and processing system as well as the qualifications and competencies of human resources. The process system is a series of processes and their operating parameter conditions, which are a road map for producing drugs. While the processing system consists of a series of equipment to accommodate the process system, which converts raw materials or semi-finished materials into final products that are ready for use. Until 2019, Indonesia has around 200 pharmaceutical industries with total sales reaching IDR 80 trillion with a growth rate of the pharmaceutical industry reaching 14.10% per year [1]. Constraints faced by the pharmaceutical industry are generally still imported raw materials [2,3] and equipment as well as limited competence and qualifications of human resources.

Medical equipment, drug production, food and beverage have low roughness requirements, ranging from 0.14μm and shiny [4]. This roughness value is related to the ease of cleaning the equipment. In an effort to achieve this roughness value, the initial material is being mechanically polished and followed by electropolishing. The electropolishing process can reach areas where mechanical polishing cannot or its difficult.

AISI 316 is the most widely used austenitic Cr-Ni stainless steel for medical equipment because of its high corrosion resistance, good formability and weldability [5]. The use of this material is widely found in the food industry (cooking equipment, stoves, refrigerators, dairy processing, wine making, storage tanks), the petrochemical, chemical and nuclear industries, dyeing industry, architecture, and medicine. In addition, chemical containers, heat exchangers, mining equipment, and sea nuts, bolts and screws, filtration systems [5,6]. Uses for medical equipment are shown in Figure 1.

Figure 1. Medical equipment [4]

The formation of a thin passive surface layer on AISI 316L causes better corrosion resistance [5]. The presence of aggressive substances in the environment can cause local damage to the passive film and consequently local corrosion i.e., pitting. Pitting corrosion is influenced by internal factors such as chemical composition, surface treatment, passive film and environmental conditions such as temperature, pH, aggressive ion concentration. Process temperature is a factor that greatly affects thermodynamics and corrosion kinetics, ranging from 20 – 100 °C. Electropolishing costs range from \$0.15-\$0.45 dm2/min, becoming one of the driving factors for the growth and development of the domestic electropolishing industry [6].

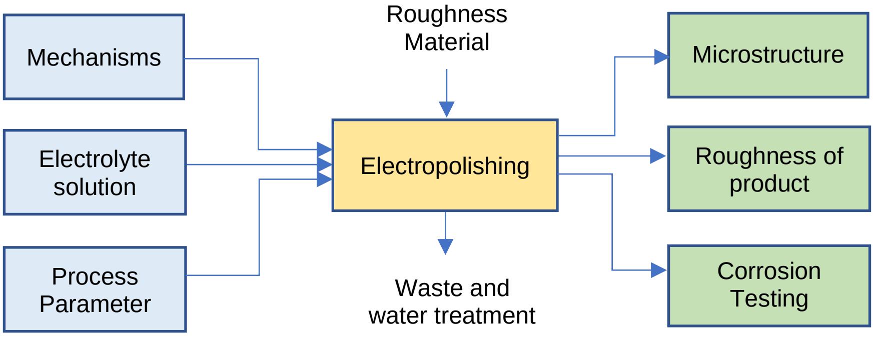


Figure 2. Electropolishing process system

Understanding the electropolishing process system begins with an understanding of the mechanism of the electropolishing process, a mixture of electrolyte solutions, and process parameters to control the electrochemical process of the material surface into a surface that has a smooth and shiny microstructure and better corrosion resistance [7–9]. The smooth and shiny surface makes cleaning easier, while the corrosion resistance increases the life of production equipment and product quality. The simple electropolishing process is illustrated as shown by Figure 2 above.

1.2. Electropolishing Process

The electropolishing process is an electrochemical polishing process that combines an electrolyte solution-electric to produce a smooth and shiny surface, that is easy to clean and has high corrosion resistance [7–10]. Compared with mechanical polishing process are: (1) being able to produce a shiny, clean and microscopically smooth also uniform surface, (2) it can eliminate stains and scratches. The surface roughness of mechanical polishing only reaches 0,51 μm, and then reaches 0,38 μm through the electrolytic polishing process, which is the requirement of 0,25-0,8μm for the process equipment in the pharmaceutical industry [5]. In general, the roughness of pharmaceutical equipment products ranges from 0,35-0.8 μm [3] summarized in Fig. 3.

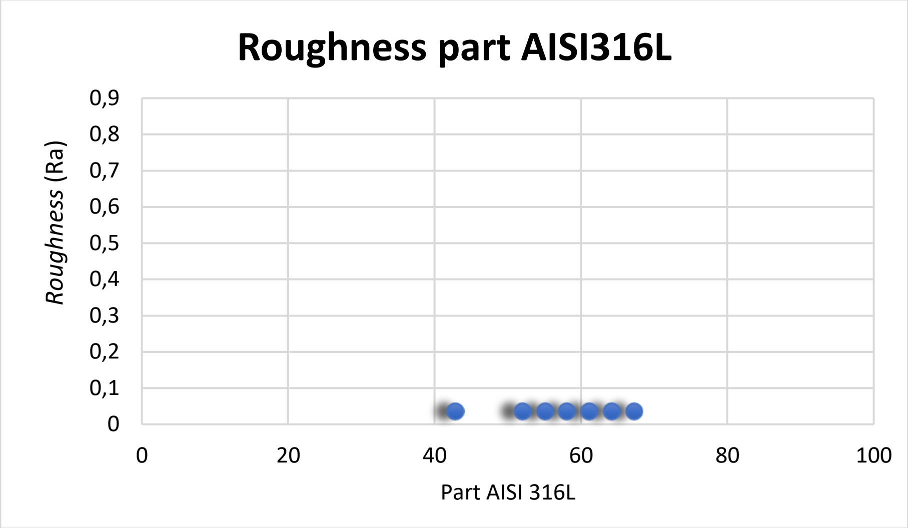


Figure 3. Roughness of various parts AISI 316L [3]

In this study, a compilation of journals is presented, especially related to process parameters and roughness of the electropolishing process for industrial equipment [11,12]. Electropolishing process parameters correlate with roughness characteristics. In other words, to get a low roughness and reproduceable, it is necessary to set the electropolishing process parameter. The basic principle of the electropolishing process circuit and the relationship between electric potential and current density [8] is shown by Figure 4, while the process parameters include electrolyte solution and process parameters current density, temperature, time and cathode. In this process, the pharmaceutical equipment functions as an anode which during the electropolishing process undergoes dissolution on its surface. This dissolution process produces a surface with low roughness (smooth), shiny and microscopically flat [7,8,13].

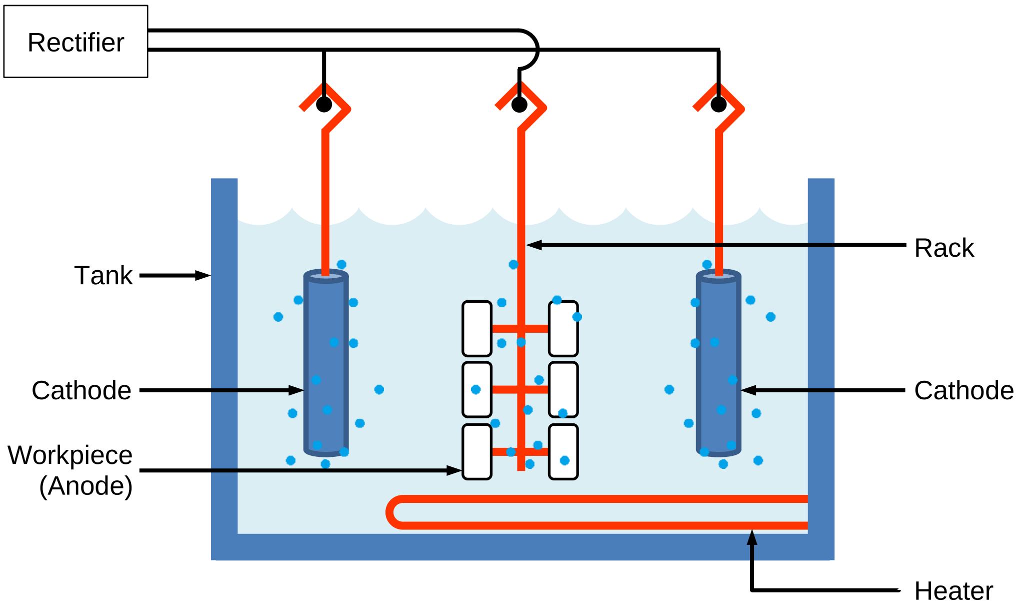


Figure 4. Illustration of Electropolishing circuit

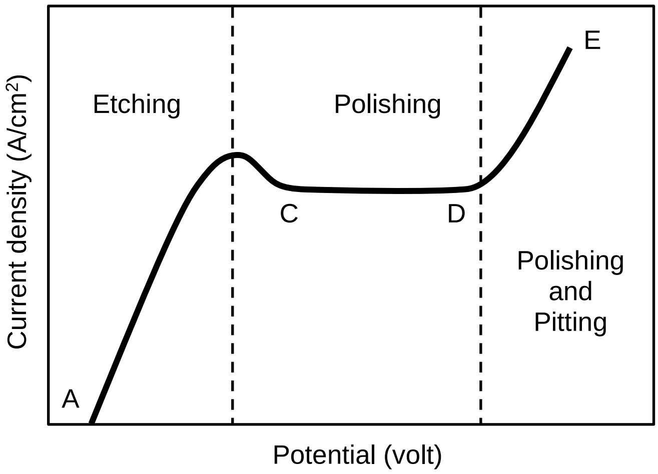


Figure 5. Potential against current density [8]

From figure 5, it has been seen from the beginning that the increase in potential will be proportional to the increase in current density, which is known in the etching process. In addition, when the potential increases, the current density will decrease and stabilize, and the electroplating process will occur at this time. The increase in potential will increase the current density, which indicates that polishing and etching and polishing processes take place [8]. Current limit (I_{limit}) is related to activation energy (Ea), temperature (T K) and rotation disk. When using a rotating disk electrode for electrolytic polishing, the current density limit at the electrolyte temperature can be calculated by equation 5. Therefore, in the electrolytic polishing process, it is necessary to determine the potential range that generates a stable rated current.

$$i_{limit} = k_0 e^{(-\frac{E}{RT})\Omega^{0.5}} \tag{5}$$

Based on equation [5], it is clear that during the electropolishing process there is an increase in temperature. As a result, the electrolyte resistance decreases, the current density decreases, and the viscosity decreases, resulting in a decrease in roughness quality. Of course, the type of electrolyte and the concentration of electropolishing will affect the time required to reduce the roughness. In general, the higher the current density, the shorter electropolishing process time required. Excess time will result in grain and grain boundaries being exposed.

1.3. Mechanism of Electropolishing

It is well known that austenitic chromium and nickel steels (e.g., AISI 304) and chromium-nickel-molybdenum (for example, AISI 316) is widely used in the health, medical, food and beverage industries. In the electrolytic polishing process, the equipment is immersed in a bath with electrolytic polishing process parameters to dissolve the anode and make the surface bright and smooth. Anodic and cathodic reactions in the process of electropolishing or electrochemical dissolution can be described by the following equation [7]:

$$M^0 - ze^- \rightarrow M^{z+} \tag{1}$$
$$2H^+ + 2e^- \rightarrow H_2 \tag{2}$$

The anodic dissolution of metals and their transmission to solution in the form of simple or complex hydrated ions can be described by the following equation (4):

$$Me + xH_2O - ze- \rightarrow Me^{z+} \cdot xH_2O \tag{3}$$
$$Me + xH_2O + yA- - ze- \rightarrow [MeAy]^{z-y} + \cdot xH2O \tag{4}$$

The effects of surface electropolishing of metal elements Stainless steel, aluminium alloy, copper is:

1. Macro-polishing, that is, removing peaks with a height of about 100 μm to make the surface smooth.
2. Micro-polishing, that is, to remove peaks with a height of about 10 μm to make the surface shiny.
3. Passivation refers to the formation of a passivation oxide layer on the metal surface.

1.4. Parameter process

The type of solution that is widely used in the electropolishing process varies from the type and concentration as well as the mixture. Electropolishing solutions are very diverse, an example of an electrolyte composition is presented in Table 1. Based on Table 1, the concentration of sulfuric acid (H2SO4), phosphoric acid (H3PO4), various types of additives are correlated with current density, temperature and electropolishing time [10–26]. Therefore, it is necessary to optimize the electropolishing process parameters.

Table 1. Solution and process parameters of 316L stainless steel electropolishing [7]

No.	Material	Solution			Parameter process		
		H_2SO_4 (% of weight)	H_3PO_4 (% of weight)	Additive (% of weight)	Current density (A/dm^2)	Temperature (ºC)	Time (minute)
1	304 and 316L	35	51	TEA: 3	20	12	55
		40P	60*	ETG: 99-200	35-50	60	1-50
				OSL: 200			
				CAN: 200			
2		50*	50*	-	15	40-75	1-3
			35*	Glycerin 50	75	60-95	1-50
3		H_3PO_4/H_2SO_4 : 2/1-3/2		Glycerin 25	50	30-90	1-10

1. * : % of volume
2. TEA : Triethylamine; ETG : ethylene glycol; OSL: Oxalic acid (gr/dm^3); CAN: acetanilide (gr/dm^3)
3. % of H_2SO_4 purity between 96-97; H_3PO_4 : 85; glycerin : 99; TEA : 99; ethylene glycol : 99

2. METHODS

2.1. Design of experiment

There are 3 types of steel surfaces studied used for cyclic potentiodynamic tests.

a. Surface starting material (AR), i.e., without mechanical/chemical polish treatment
b. Surface polished with mechanical polish (PM)
c. Electro-polished (PM+EP) surface.

A series of electropolishing experiments were carried out to obtain optimal process parameter conditions. The surface roughness of the parts resulting from the electropolishing process will be characterized by X-Ray Diffraction (XRD) to determine the dominant compound, Scanning Electron Microscope (SEM) to determine morphology, and Energy Dispersive X-9 Ray Spectroscopy (EDX/EDS).

Table 2. Design of Experiments

Steel Surface	Material	Parameter Process		
		Current Density	Time (minute)	Potential (volt)
Surface starting material (AR)	Austenitic stainless	0.5	1	2.4
	AISI 316L	1.5	3	3.1
	AISI 304	2.5	6	4.2
Surface polished with mechanical polish (PM)	Austenitic stainless	0.5	3	3.2
	AISI 316L	1.5	6	3.1
	AISI 304	2.5	1	4.1
Electro-polished (PM + EP) surface	Austenitic stainless	0.5	6	2.1
	AISI 316L	1.5	1	3.0
	AISI 304	2.5	3	3.8

The electropolishing experiments was conducted to three materials with the same solutions, H_3PO_4, H_2SO_4, and Amidis. Their percentage of weight is 75, 50, and 30, respectively and the temperature for these experiments was determined in 30^o C for all materials with different types of steel surfaces. It is as depicted in table 2.

2.2. Material

Materials for the pharmaceutical industry process equipment include austenitic stainless, AISI 316L, AISI 304 [4].

2.3. Flowcharts

The series of electropolishing processes as presented in Figure 5. Electropolishing process flow chart consists of 3 stages, namely surface preparation, electropolishing process, and passivation. Surface preparation consists of measuring the initial roughness of the results of mechanical polishing, pickling, and rinsing. The electropolishing process consists of electropolishing, rinsing, drying, and measuring roughness, while the passivation process is carried out with nitric acid solution, rinsing, and drying.

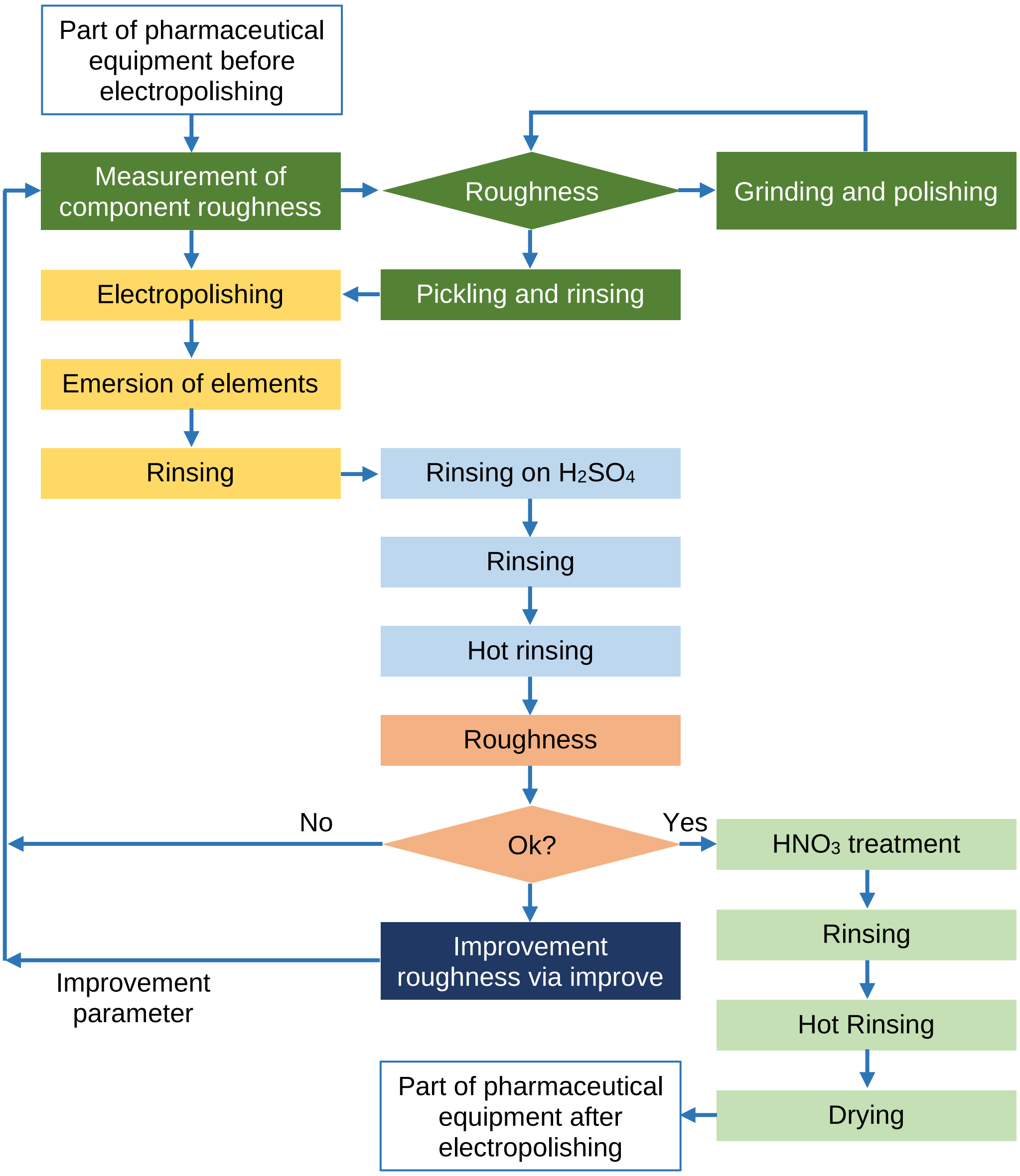


Figure 5. Flowchart of electropolishing process

2.4. Potensiodynamic/Galvanodynamic

Corrosion testing is carried out with potentiodynamics/galvanodynamics which is a corrosion test method that utilizes a polarization technique that provides potential variations at a determined rate by applying a current through a certain electrolyte medium.

In the corrosion testing approach with the potentiodynamic polarization method, several condition parameters are related to the number of electrons associated with the reaction, the atomic weight of the metal being tested, and the time of the test process. In the potentiodynamic test, the atomic equivalent weight parameters of the pure metals tested were Fe/2 (27.92), Al/3 (8.99), Ti/2 (23.95) and SS 304 Fe/2, Cr/3,Ni/2 (25.12) alloys. in the case of this study using AISI 316L austenitic stainless steel material, it can use the atomic equivalent of Fe/2, Cr/3,Ni/2 (25.12).

By knowing this measurement method, we can take advantage of the test data for anodic protection applications on equipment used in chemical processes. References used in the testing process include; ASTM G3 Practice for Conventions Applicable to Electrochemical Measurements in Corrosion Testing, ASTM G5 Reference Test Method for Making Potentiodynamic Anodic Polarization Measurements, ASTM G59: Standard Test Method for Conducting Potentiodynamic Polarization Resistance Measurements, ASTM G61: Standard Test Method for Conducting Cyclic Potentiodynamic Polarization Measurements for Localized Corrosion Susceptibility of Iron-, Nickel-, or Cobalt-Based Alloys. On Figure 4. Illustration of corrosion testing process with potentiodynamic below is shown the testing process and the schematic for the preparation of the tool consisting of a potentiostat instrument, a computer, a hot plate stirrer, a reaction vessel and electrodes.

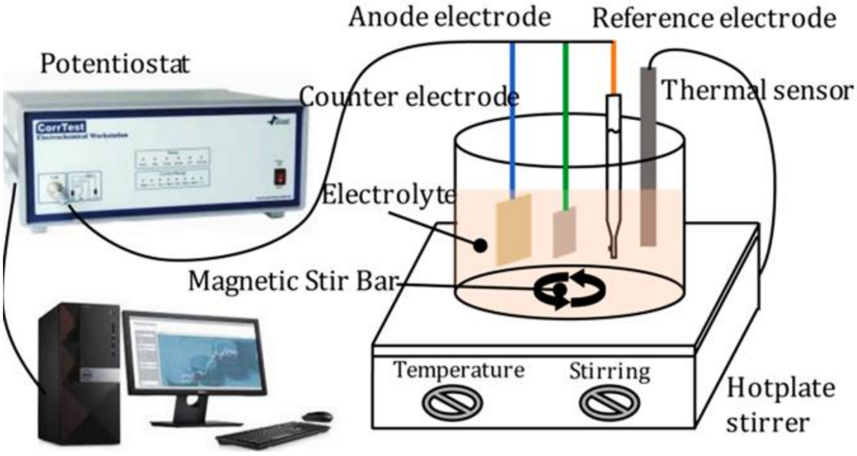


Figure 4. Illustration of corrosion testing process with potentiodynamic

3. RESULT AND DISCUSSION

3.1. Roughness

Roughness AISI 316L material at first 4.559-5.182 μm (Table 7), after mechanical polishing is 0.227-0.2582 μm (Table 8). Furthermore, electropolishing is carried out 0.278-0.388μm (Table 9). Thus, the roughness of the mechanical polish results is still better than electropolishing.

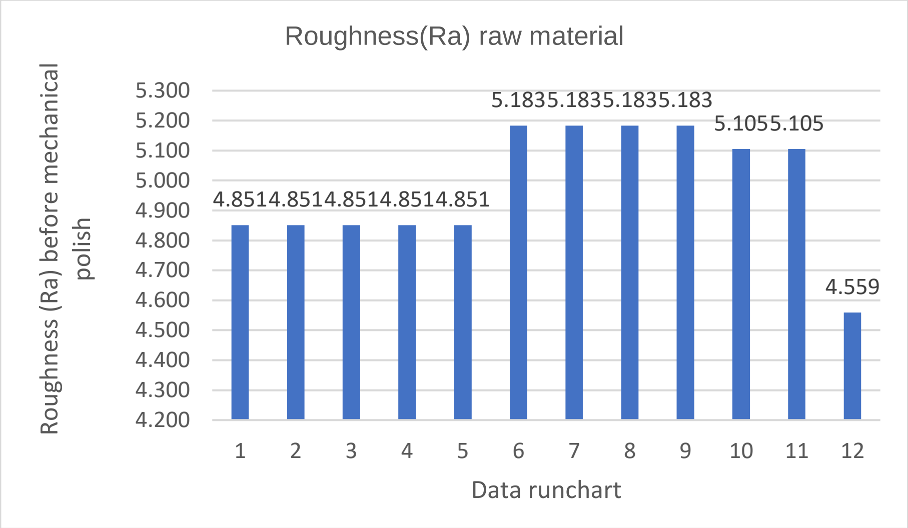


Figure 5. Material roughness of 316L

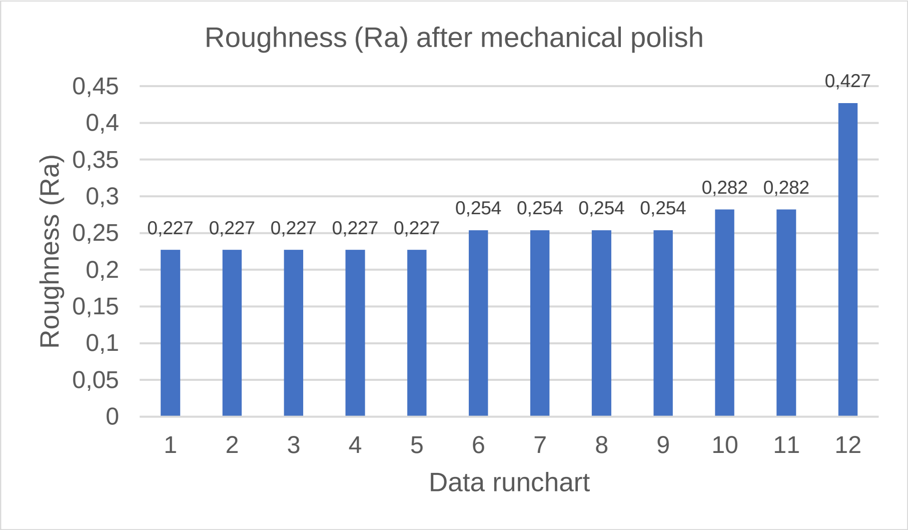


Figure 6. Material roughness of 316L after mechanical polish

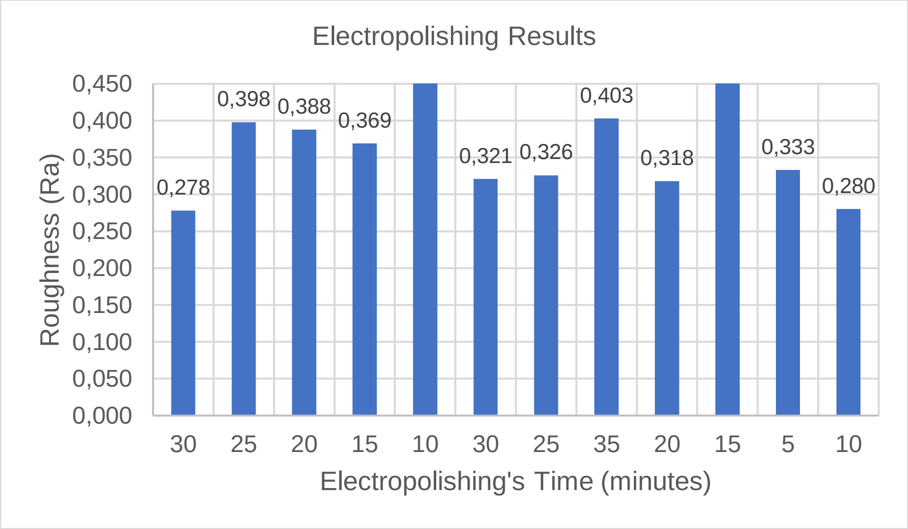


Figure 7. Material roughness of 316L after electropolishing

The roughness of the 316 material initially ranged from 4.2 to 5.2, while the mechanical polish yield ranged from 0.227 to 0.42. With the electropolishing process, it is expected that the roughness of the results will be smaller. It turned out that from the results of electropolishing the roughness increased to in the range of 0.278 – 0.403 Ra as shown in Figure 4, Figure 5 and Figure 6. Based on Figure 2 it can be understood that with an increase in voltage there will be a linear increase with an increase in voltage. Which means that the process that occurs is still in etching, so the roughness value increase.

3.2. Corrosion test with Potentiodynamics

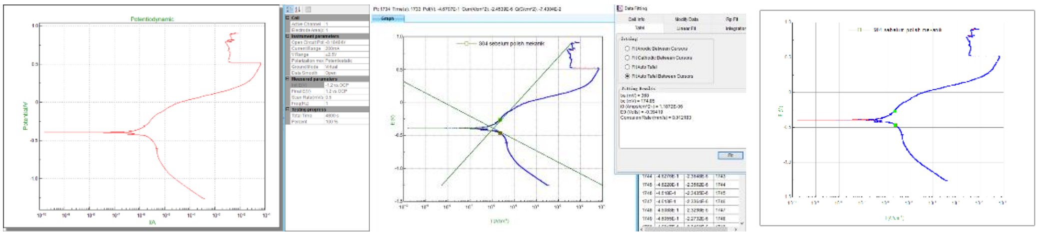


Figure 8. AISI 304 before mechanical polish

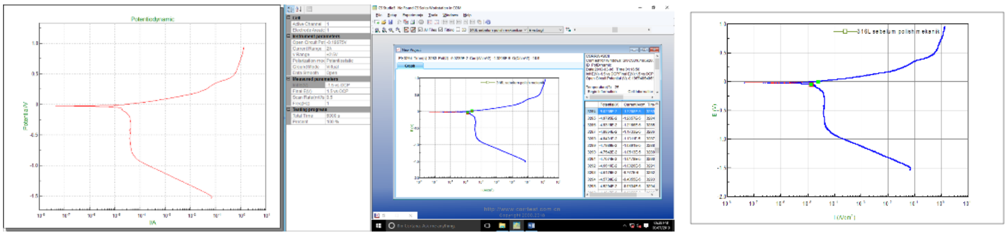


Figure 9. AISI 316L before mechanical polish

Based on Figure 11, the results of potentiodynamic testing of the initial AISI 316L material before mechanical polishing resulted in a corrosion rate of 0.0013046 as shown in Table 3.

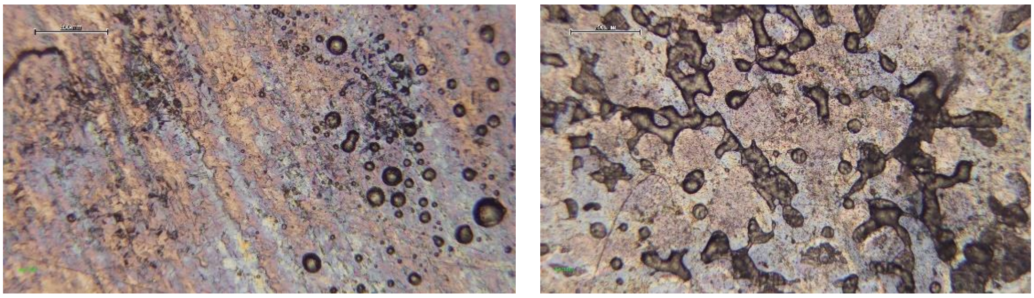
Table 3. Calculation output with corrtest®

ba (mV)	11.642
bc (mV)	11.29
i0 (Amps/cm^2~)	1.2524E-07
E0 (Volts)	-0.026008
Corrosion Rate (mm/a)	0.0013046
Residual	4.2335E-09

3.3. Microstructure test

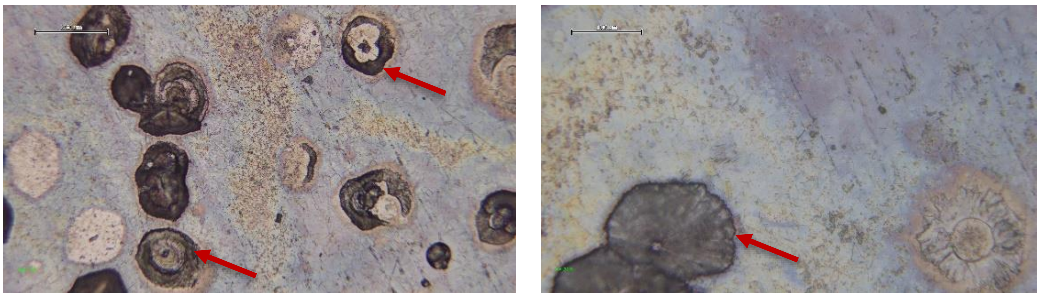
This AISI 316L microstructure shows an austenite phase matrix at

Figure 10. (A, & B), in the figure (C & D) shows the condition of the presence of globular oxide inclusions on the surface resulting from abrasion and mechanical polishing. In this condition, an electropolishing process will be carried out so that the surface of the ASI 316L material that will be used in the pharmaceutical production process is better and meets the FDA or ASME BEP grade.



A. 100x Magnification, showing austenite matrix

B. 100x Magnification, showing austenite matrix



C. 100x Magnificationi, showing globular oxide inclusion

D. 200x Magnificationi, showing globular oxide inclusion

Figure 10. Microstructure of AISI 316L mechanical polish (HF Etch 98%)

3.3. Determining the voltage

To determine the electropolishing area, it is necessary to determine the voltage where the current density is constant (level) or not a function of the voltage as shown in Figure 12. The magnitude of the voltage value to be used for the electropolishing process is close to the area where a small voltage changes results in a large current density change.

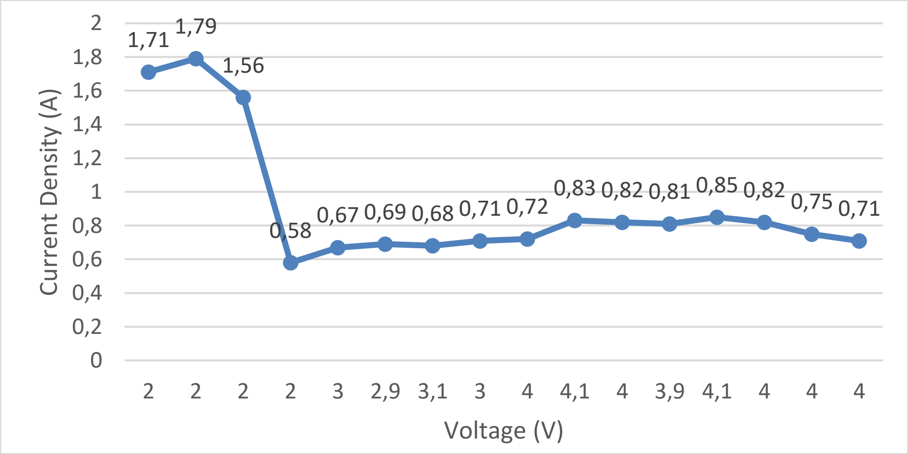


Figure 113. Voltage and current density

4. CONCLUSION

The selection of a voltage that is still at an increasing current will result in a higher roughness during the electropolishing process than the mechanical polish roughness. Research and development of the electropolishing process continues to be carried out to determine the parameters that produce roughness that meets the requirements set by the pharmaceutical, drug, food and beverage industries. Research and development of the electropolishing process can be a vehicle for implementing the MBKM program, increasing the competence of graduates, lecturers and improving the course curriculum. Several courses can be converted into the MBKM Program, including improving quality targets, processing problems, or/or quality development through mastery and development of link-and-match processing technology between universities-industry-government capable of solving industrial and university problems simultaneously. Scale up the research laboratory into a pilot plant and into a commercial electropolishing process plant as an application expansion for the food and beverage medical parts industry. The electropolishing process facility can become a teaching factory for industrial partners and universities and a vehicle for increasing product competitiveness and graduates.

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