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Short Communication

Electropolishing of Additive Manufactured 17-4 PH Stainless Steel Using Sulfuric Acid

Jun-Kai Chang^{1,2}, Chia-Yu Lee³, Yu-Chih Tzeng^{4,7}, Ming-Hsien Lin^{5,7}, Ming-Der Ger^{5,7}, Chun-Hsiang Kao⁶, Chih-Peng Chen⁶, Kuo-Kuang Jen⁶, Shun-Yi Jian^{5,7,*}

¹ Research and Development Department, Chung Yo Materials Corporation, Gangshan district, Kaohsiung City 820, Taiwan

² Laser and Additive Manufacturing Technology Center, Industrial Technology Research Institute, Liujia district, Tainan City 734, Taiwan

³ Graduate School of Defense Science, Chung Cheng Institute of Technology, National Defense University, Taoyuan, Taiwan

⁴ Department of Power Vehicle and Systems Engineering, Chung Cheng Institute of Technology, National Defense University, Dasi, Taoyuan 335, Taiwan

⁵ Department of Chemical & Materials Engineering, Chung Cheng Institute of Technology, National Defense University, Dasi district, Taoyuan City 335, Taiwan

⁶ Missile and rocket systems research division, National Chung-Shan Institute of Science and Technology, Taoyuan City 32546, Taiwan

⁷ System Engineering and Technology Program, National Chiao Tung University, Hsinchu City 300, Taiwan

*E-mail: <u>ftvko@yahoo.com.tw</u> (S. Y. Jian)

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The sulfuric acid electropolishing system was built to reduce the surface roughness of the additivemanufactured 17-4 PH stainless steel. The extra 10 V power was applied to electropolish the AM specimen to reach the desired surface roughness. The roughness was reduced to 6.76 μ m and additional corrosion resistance was given. The corrosion potential became noble and the corrosion current density decreased to approximately 10⁻⁶ A/cm². The post treatment of the electropolishing process accurately improved both the surface roughness and the corrosion resistance, demonstrating the electropolish process is a useful application in AM objects.

Keywords: Additive Manufacturing; Electropolish; 17-4 PH stainless steel; Roughness

1. INTRODUCTION

17-4 precipitation-hardening (PH) steel is known as martensitic matrix steel. Using appropriate heat treatment, the mechanical properties can reach the required standard, and therefore 17-4 PH steel

has been applied in different fields such as nuclear power plants, automobiles and aviation industries [1-3]. Recently, the requirement of the manufacturing process of 17-4 PH stainless steel has been raised to a precision level, and traditional manufacturing is unable to reach such high accuracy [4].

Additive manufacturing (AM) technology, also termed 3D-printing, has been widely applied in the metal industrial field because of the reduced cost, mold consumption and creation of complex geometric shapes with good accuracy [5]. The objects are built layer by layer using metal powders as materials and laser beams as the heating source. The metal powders are melted with a high-energy laser beam, followed by rapid solidifying at a large cooling rate to build the component layer-by-layer object. The common metal AM technology is classified into (1) powder feeding systems such as directed energy deposition (DED) and (2) powder bed systems, such as electron beam melting (EBM) or powder bed fusion (PBF) according to the difference in melting heat source [6-10].

Though AM resolves the accuracy and limit of the metal materials, the surface roughness is still an obstacle because of the mutual and continuous melting and solidifying during the AM process. The undulated surface is a stress concentration and directly affects the wearing and fatigue properties while using [10-11]. Therefore, post treatment is necessary in AM objects. Grinding, lapping and mechano-chemical polishing are commonly used in the post treatment of the metal. Surface polishing is the process of smoothing metals and alloys to a bright, smooth, mirror-like surface. However, there are limits to the object shape and accuracy, and the restrained stress would lead to unnecessary and extra obsession [11]. Electrochemical polishing is normally carried out in concentrated acid media such as phosphoric acid, sulfuric acid, perchloric acid, acetic acid and their mixture solutions. Suitable electropolishing electrolytes for different metals can be found in the works of Mctegart [12] and Shigolev [13]. Instead of water, methanol is sometimes used as a solvent [14-15]. Therefore, electropolishing (EP) is an approach to reduce surface roughness in AM objects [16-18]. The process includes putting a cathodic electrode in the solutions and exerting direct power to cause a reaction between the object surface and solutions to remove the rough surface and efficiently reduce the roughness [18-22]. This work used sulfuric acid as the electrolyte to build the EP system and study the further effect and application of the EP process for AM 17-4 PH stainless steel.

2. EXPERIMENTAL

2.1. Materials and electropolishing parameters

In this study, 17-4 PH stainless steel was chosen as the AM specimen. Table 1 lists the element content of the 17-4 PH stainless steel. The EP parameters are also shown in Table 1.

In this work, sulfuric acid was used because the previous studies identified the passive film would form on SS316L or CoCrFeMnNi high entropy alloy during the polarization process [23-24]. The code for each specimen in this study was given according to whether EP was conducted or not, e.g. EP-AM meant the AM specimen conducted the EP process, but AM meant it was not.

17-4 PH stainless steel composition											
Element	Cr	Ni	Cu	Mn	Ta	Mo	Nb	С	Si	Fe	
Content (wt.%)	16.98	5.42	3.41	0.46	0.016	0.76	0.25	≦0.07	≦1	Bal.	
Electropolishing parameters											
Electrolyte			Operating voltage (V)					Operation time (min)			
10 wt.% H ₂ SO ₄ solution			10				5				

Table 1. 17-4PH stainless steel chemical compositions and related electropolishing parameters

2.2. Microstructural characterizations

The surface morphology of the AM and EP-AM specimen was characterized by a scanning electron microscope (SEM, JEOL JSM-IT100) in secondary electron mode (SE) to present the microstructural characterizations of the surface. In addition, the energy dispersive spectroscopy (EDS) equipped in SEM was used to analyze the chemical compositions with an acceleration voltage of 15 kV. X-ray diffraction (XRD, Bruker D2 PHASER) was also used to identify the crystallization of the specimen before and after the EP process. Moreover, surface roughness was measured by digital 3D white light interferometry (Chroma 7503).

2.3. Electrochemical properties measurements

The electrochemical properties of the effects of the EP specimen were examined by a potentiostat (VERSASTAT4, Princeton Applied Research) in a solution with 3.5 wt.% sodium chloride (NaCl) at 25 °C. The three-electrode electrochemical cell with a saturated calomel electrode (SCE, +0.24 V vs. SHE at 25 °C) as the reference electrode, a platinum plate of 16 cm² as the counter electrode, and the working electrode with an exposure area of around 1.77 cm² were used for all electrochemical corrosion tests.

Prior to the potentiodynamic polarization test and electrochemical independence spectrum (EIS) measurement, each specimen was immersed in the test solution to reach the steady open circuit potential (OCP), whereby potential fluctuation was less than 5 mV within the previous 300 s. The polarization curve was measured by sweeping the potential from -200 mV to +500 mV with a scan rate of 1 mV / s, relative to the steady OCP [25-29]. The electrochemical impedance spectroscopy (EIS) was acquired using a sinusoidal wave with amplitude of 5 mV over the frequency range between 10^{-2} Hz and 10^5 Hz [30].

3. RESULTS AND DISCUSSION

Figure 1 shows the OCP records of the AM specimen in 10 wt.% H_2SO_4 solution. Initially, the potential was approximately +0.44 V (v.s. SCE), and the potential sharply grew with increased immersion time. The final steady potential was larger than +0.49 (v.s. SCE). With such noble potential,

it seems the passive film formed on the surface AM specimen during the immersion and the distinguished film exhibited effective resistance in the H_2SO_4 solution. Previous studies [27, 29] showed the H_2SO_4 solution reacted with the noble elements such as Cr and Ni to form dense and continuous protective film and enhance corrosion resistance.



Figure 1. The OCP of the AM specimen immersed in 10 wt.% H2SO4 solution.



Figure 2. The potentiodynamic polarization curves of the AM specimen in 10 wt.% H₂SO₄ solution.

Figure 2 shows the potentiodynamic polarization result of the AM specimen immersed in 10 wt.% H₂SO₄ solution after a steady potential with a vibration of less 5 mV for 300s. In the cathodic reaction, an obvious passivation region was recorded while the corrosion current density (*i*_{corr}) maintained as the extra positive potential was applied. The phenomenon demonstrated the polarization in the cathodic region provided not only a reduced-roughness surface, but also passive protection resistance on the AM specimen. Table 2 lists the Tafel curve parameters in the polarization result of Figure 2, the corrosion potential (*E*_{corr}) was 0.48 V(v.s. SCE) and the corrosion current density was 9.56×10^{-6} A/cm².



Table 2. The potentiodynamic polarization results in Figure 2

Figure 3. The XRD diagram of the AM and EP-AM specimens.

Following the EP process, Figure 3 shows the XRD crystallization result. The initial AM specimen showed the surface covered with Cu and Fe-Cr-Ni phases. During the additive manufacturing process for the powder laser melting process, the 17-4 PH powders melted via laser and re-solidification. During the re-solidification process, the elements precipitated in different order due to the melting points. Compared to these elements in the 17-4 PH stainless steel, Cu has the lowest melting point, which led to Cu finally solidifying during the additive manufacturing process [23-24]. Moreover, the cooling rate of the additive manufacturing was too rapid, e.g. 106~108 K/s, causing

clear element precipitation rather than solid soluting in the matrix. After the EP process, the XRD patterns were similar to the initial AM specimen. However, the intensity of Cu signals was relatively larger than the initial one, which seemingly means during the EP process, Cu was not etched and restrained on the surface after the EP process.

Figure 4. The SEM surface morphology of (a) initial AM 17-4 PH stainless steel and (b) the enlarged part in (a); (c) EP-AM and (d) the enlarged part in (c).

Figure 5. The quantification of surface roughness mapping of the AM and EP-AM specimens, respectively.

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Figure 4 and Figure 5 demonstrate the surface morphology analyzed by SEM under a 3D white light microscope. In Figure 4(a) and (b), the AM surface observation shows the powders existing on the surface, meaning the laser power causes the scattering of powders and the laser energy is unable to make the powder fully melt. The powders therefore remained in solid form. Figures 4(c) and (d) show the surface of the EP-AM specimen, whereby the EP process effectively reduces the amounts of powder-shape surface and improves fluctuation of the surface. Further, using the 3D white-light reflection on the surface to qualify the roughness to study the effect of the EP process. The surface roughness of the AM specimen was 11.69 μ m, and the roughness was reduced to 6.76 μ m, meaning the EP process accurately improves the roughness regardless of the precipitation of the Cu phase on the surface. In very basic terms, two kinds of effects may occur during the electropolishing process, smoothing and brightening. The former refers to the elimination of surface roughness of more than 1 μ m, and the latter to the elimination of surface roughness less than 1 μ m [15, 31-32].

Figure 6. The OCP of AM and EP-AM specimens immersed in 3.5 wt.% NaCl solution.

Figures 6-8 display the electrochemical tests of the EP-AM compared to the initial AM. Tested in the 3.5 wt.% NaCl solution, the OCP of the EP-AM was nobler than the AM according to Figure 6, meaning the EP process made the AM surface.

Table 3. The potentiodynamic polarization parameters in Figure 7

	AM	EP-AM
E_{corr} (V v.s. SCE)	-0.51	-0.39
i_{corr} (A/cm ²)	3.68×10 ⁻⁵	1.35×10 ⁻⁵

Moreover, the potentiodynamic polarization results in Figure 7 and Table 3 demonstrate corrosion resistance greatly improved after the EP process.

Figure 7. The potentiodynamic polarization curves of AM and EP-AM specimen in 3.5 wt.% NaCl solution.

Figure 8. The EIS curves of AM and EP-AM specimen in 3.5 wt.% NaCl solution.

The corrosion potential became noble and the corrosion current density neatly reduced to one order. Similar results are also shown in the EIS result, with the enlarged impedance near 2500 ohms,

which has an initial impedance of approximately 500 ohms, as shown in Figure 8. From the electrochemical measurements, the EP process provides additional corrosion resistance [33-34]. Besides, some electropolishing processes form a thin, roughness-reducing and protective oxide (or salt) film on the surface. The passive film can improve corrosion resistance and extend the life of the specimen [28-30, 34-35].

4. CONCLUSIONS

In the present work, the sulfuric acid EP process was built to improve on the poor roughness from additive manufacturing. The following are the conclusions of this work:

1. The roughness of the AM specimen improved from 11.69 to 6.76 μ m via the EP process.

2. The corrosion resistance increased, contributing to the passive film in the sulfuric acid during the EP process, which is similar to the cathodic treatment.

3. The sulfuric acid system in the EP process provided not only reduced roughness, but also assisted corrosion resistance.

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