The application of alumina-phosphate ceramic coating on steel for pulverizer pipe

Dewi Idamayanti[1], Dia Ginanjar[2], Beny Bandanadjaja[3], Wiwik Purwadi[4], Noval Lilansa[5]

[1-4] Foundry Engineering Department, Politeknik Manufaktur Bandung
[5] Automation Engineering Department, Politeknik Manufaktur Bandung

[1]idamayanti79@gmail.com, [2]diagin96@gmail.com , [3]benybj@yahoo.com, [4]wiwikpurwadi@yahoo.com, [5]noval_l@polman-bandung.ac.id

Abstract. Pipe pulverizer is a part of a coal mill machine that has a function to direct the coal dust from the grinding mill to the turbine combustion chamber of the steam power plant. The pipe material is mild steel. The process of sulfidation and erosion of coal dust can cause a decrease in pulverizer pipe performance, thus its service life also reduces. The pipe deterioration can be anticipated from the beginning of the pipe manufacturing so that it can avoid production time due to maintenance. The ceramic coating overlay on the surface of the mild steel is one of the best ways to protect the steel from erosion and sulfidation. This research is aimed to perform a ceramic coating over the surface of the mild steel to improve its erosion resistance by using the alumina-phosphate ceramic compound. The coating technique using a dipping method. The coating layer is formed by the reaction between monoaluminum phosphate (MAP, $Al(H_2PO_4)_3$) as a binder and aluminum oxide (Al_2O_3) particle. It transforms to berlinite layer on the mild steel surface when heated in the phase transformation temperature. The observation of the coating performance is carried out with the variation of the binder composition Al:P 25:75, 28:72, 30:70 and the Al2O3/MAP slurry is given at 40/60. Optical emission spectrometry (OES) is applied to analyze the chemical composition of the mild steel, energy dispersive spectrometry (EDS) and scanning electron microscopy (SEM) is used to characterize the contents of the particle, analyze the morphology and also the microstructure of the coating. X-ray diffraction (XRD) is applied to investigate phase of the binder compound of monoaluminum phosphate. Erosion resistance properties of the coating are characterized by the gas erosion jet test. From the test result, it can be concluded that the erosion resistance of mild steel with an alumina-phosphate ceramic coating increasing four times compared to the condition without coating. Its performance is mainly influenced by the binder composition.

Keyword : ceramic coating, alumina phosphate binder, low carbon steel, pulverizer pipe

1. Introduction

The pulverizer pipe is a part of the coal mill pulverizer system which has a function to deliver coal dust to a combustion chamber. Mild steel is commonly used for the pulverizer pipe material due to its mechanical strength, machinability, weldability, formability and reasonable cost. Many energy projects also rely on the amount of steel for pipelines and electric power turbine component[1]. Unfortunately mild steel does not have good erosion resistance against solid particles such as coal dust. During coal grinding at the working chamber of a coal mill pulverizer, the process releases a certain amount of heat which raises the temperatures up to 300 ^{o}C. Pulverizer pipe and inner cone which made of mild steel is impinged by coal dust particle and exposed to heat continuously, then the erosion take place. The failure of the pulverizer pipe due to coal dust erosion describe in figure 1.

Figure 1. Erosion failure of pulverizer pipe

The erosion on mild steel is particularly induced by repeated plastic deformation of coal dust particle impact. Mbabazi et al [2] observed that the erosion rate of mild steel was affected by the impingement angle. The highest erosion rate of mild steel is 6.5 $mg.Kg^{-1}$ the impingement angle reached an inclination between 25-30 deg.

Ceramic coatings are widely used for the protection of base metal or cement components in chemical, power, and refractory industries against high temperature corrosion and oxidation and for the minimization of wear or erosion [3]. Alumina ceramic layer on steel is used to provide better performance and help in decreasing erosion that is caused by pressurized air containing entrained abrasive solid coal particles such as oxides and minerals. When the angle of impingement is less than 30 deg, wear produced is closely similar to abrasion but if the angle of impingement is greater than 30 deg to surface, wear produced is released by impact erosion [4]. Ścieszka et al [5] observed the erosion resistance of mild steel coated-alumina compared to high alumina ceramic by solid particle erosion test using pulverized coal dust from a power plant. The result explained that the erosion resistance of alumina-coated mild steel is four times higher than the high alumina ceramic its self. Troczynski et al [6] have already investigated the behavior of alumina ceramic coating using phosphate binder for the variety of application, including high-temperature corrosion protection, wear resistance, dielectric properties, non-sticky surface, bioactive ceramic, thermal barrier ceramics, and others. Chen et al [7] expressed in their paper, phosphate inorganic binder plays role in many applications i.e. repairing and joining of material.

This research focused on the use of alumina ceramic coating with phosphate binder on mild steel due to their superior properties and their suitability to the working conditions of pulverizer pipe. It is preferred over the sintering method which requires a process at elevated temperature. The alumina phosphate ceramic coating consists of alumina particle in which aluminum phosphate used as a binder [8]. Aluminum phosphate binder is formed by reacting inorganic mineral or oxide with an acid-phosphate solution [8]. It is a sort of inorganic binders used in refractory ceramic coating systems, which has been investigated and applied in the thermal spray coating systems. It is reported that refractories bonded with aluminum phosphate exhibit high strength, high-temperature stability, and abrasion resistance. Wang et al [9] reviewed that phosphate adhesive could be made in low temperature curing with high shear strength and excellent electrical properties. He observed the performance of phosphate adhesive which contained $Al(HPO_4)_2$ as the major constituent, has a heat-resistance of 1500^{o}C.

In this study, the influences of the variety of the phosphate binder in alumina particles that coated mild steel are observed. Furthermore, it was characterized using XRD, SEM-EDS and erosion testing then compared with mild steel without any protection. The results will be recommended for erosion protection of pulverizer pipe.

2. Experiment Work

2.1 Materials and specimen preparation

The materials used for ceramic coating were alumina (Al_2O_3) as a filler and $Al(OH)_3$ – H_3PO_4 as raw material of binder. Alumina (Al_2O_3) that has size of 50 – 60 µm was obtained from the local market with technical grade material. All materials that are used come up with technical grade. It is consist of phosphoric acid (85%), aluminum hydroxide ($Al(OH)_3$), methanol and aquades without further purification.

The specimen used in the investigation was mild steel of 5 mm thickness. The mild steel sheet was cut to a size of 25 mm x 25 mm for ceramic coating. Furthermore, the mild steel specimen was cleaned by methanol to remove scale, corrosion, and other contaminants.

2.2 Synthesis of binder

The phosphoric acid and aluminum hydroxide were synthesized to achieve monoaluminum phosphate binder as the equation 1.

$$3H_3PO_4 + Al(OH)_3 \rightarrow Al(H_2PO_4)_3 + 3H_2O \;\ldots\ldots(1)$$

Various moles was added to find out the influence of the binder toward the mechanical properties of the coating material with composition as shown in the table 1.

Table 1. monoaluminum phosphate composition

Materials	Various moles of $Al(OH)_3$: H3PO4		
	Al:P 25 : 75	Al:P 28 : 72	Al:P 30 : 70
$Al(OH)_3$	12 %	14 %	16 %
H_3PO_4	55 %	52 %	51 %
Aquades	33 %	34 %	33 %

This mixed solution was reacted at 100° - 120° Celcius to achieve the final product reaction of monoaluminum phosphate. Furthermore, the reaction was dried at the ambient temperature. Figure 2 showed the whole process of this reaction.

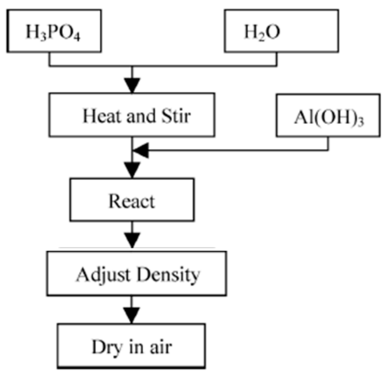

Figure 2. Flowchart of synthesizing monoaluminum phosphate

2.3 Synthesis of alumina phosphate ceramic coating

The alumina phosphate ceramic coating was synthesized from aluminum oxide particle and monoaluminum phosphate binder with 40 : 60 of the ratio [3] and applied to the mild steel which has already cleaned with methanol to remove the scale and grease, by dipping technique at the constant time. To gain the hardness of the aluminum coating, it was dried at the various temperature and illustrated in figure 3.

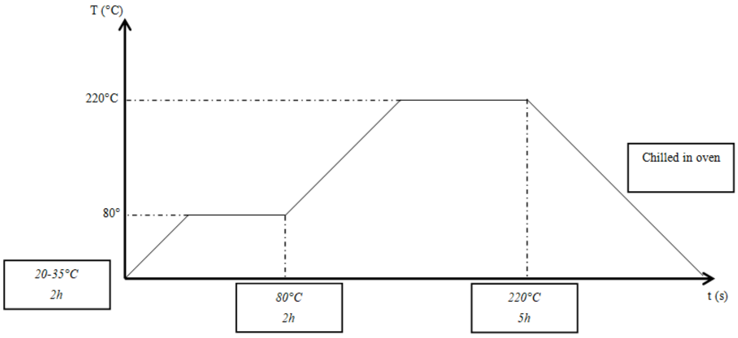

Figure 3. Flowchart of aluminum coating heating

2.4 Characterization

Several tools were used to characterize the coating, Optical Emission Spectrometer (Spektris ARL 3460) is used to determine the composition of the steel, Optical microscope (OLYMPUS SZ61) to figure the coating surface, Scanning Electron Microscope (Hitachi SU3500) to observe the microstructure of the coating and interface bonding between steel and coating. Energy Dispersive Spectrometer (EDS) was applied to check the alumina particle composition. The density of the coating was counted by Pycnometer (Pyrex, 5 mL). X-Ray Diffraction (SmartLab X-Ray Diffractometer) was used to characterize the binder and operating at 30 kV and 30 mA scanning at 10° (2θ) to 60°. Erosion tester (TR-470) used to characterize the erosion properties of the coating. This test according to ASTM G76-02 (standard test method for conducting erosion test by solid particle impingement using gas jets). After the test piece being placed at a jig, it will undergo impact load of alumina particle from the nozzle with speed of 30 m/s until 70 m/s, 90° angle and particle feed rate 2 gr/min for 10 minutes.

3. Result and Analysis

3.1 Monoaluminum Phosphate Binder

He et al [3], Emmerson, et al and Wagh [12], reviewed that *aluminum hydrogen phosphate or* $Al(H_2PO_4)_3$ is the phase being expected to be present on this binder. Because it can be reacted with the aluminum oxide particle to form berlinite. The XRD result can be seen in figure 4.

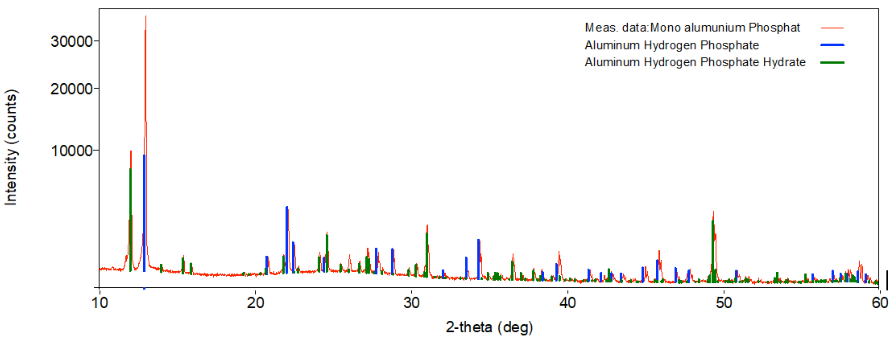

Figure 4. XRD result of synthetic binder

The peak shows other compound aluminum hydrogen phosphate hydrate $Al(H_2PO_4)\ nH_2O$. This phase was read because the binder aluminum still consists of water, due to the heating process before. The

XRD result can be the basis of this research to continue the synthesized between binder and the aluminum oxide particles to achieve coating material.

3.2 Characterizations of alumina-phosphate ceramic coating

Observation of the coating surface was conducted to find the agglomeration of the particle, crack, and the impurity from other material. This observation is important explained by E.Colonetti research [14]. Colonetti reviewed that crack can give an influence to the mechanical properties of the coating.

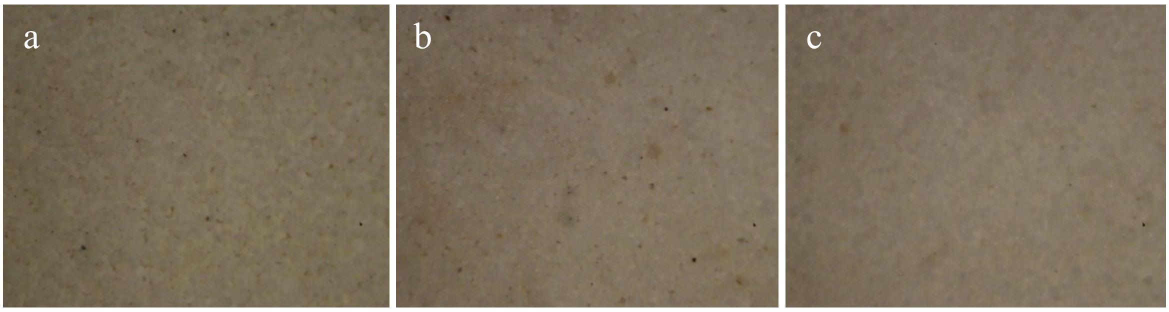

Figure 5. Surface morphology of the coating, a. Al/P (25:75) b. Al/P (28 :72) c. Al/P (30:70)

As seen in fig 5 The surfaces of the coating had a uniform particle distribution, leak and fissures are not observed in this coating. The dark spot on the coating can be can be considered as an impurity or an ash when the coating is heated.

XRD test was used to characterize the compound of the coating with binder ratio Al/P (28:72).

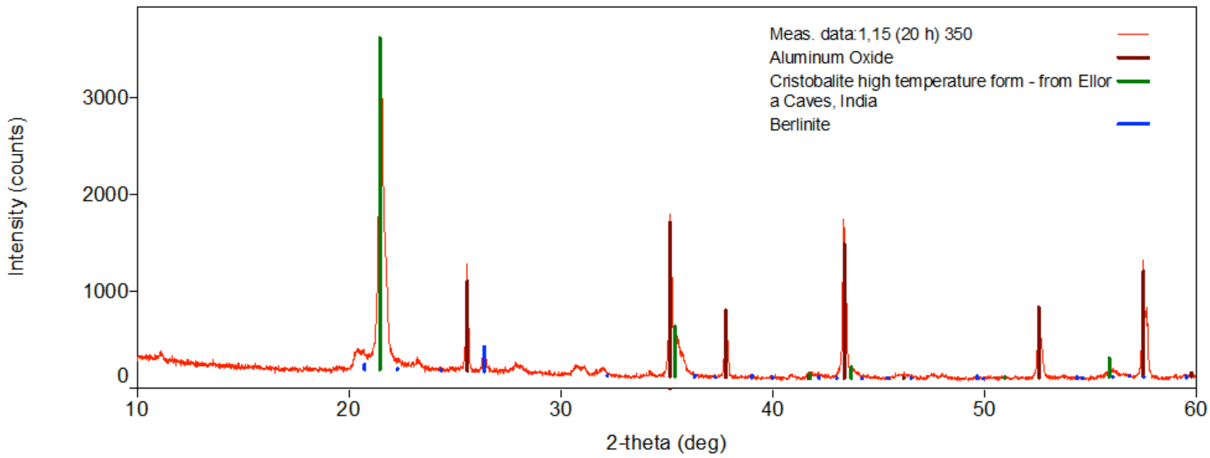

Figure 6. XRD result of alumina-phosphate ceramic coating contained Al/P binder (28:72)

Fig 6 shows the main compound of this coating are aluminum oxide, cristobalite and little amount of berlinite in 20°-30°. Berlinite is described as a small peak in diagram, since it serves only as filler in the material. Liping He [3], and Colonetti [14] also detected berlinite at 20°- 30° with a small peak.

3.3 Effect of density to alumina-phosphate ceramic coating

Characterization of coating morphology was conducted to prove the effectiveness of the binder to the alumina ceramic coating. Wagh et.al [12] observed the effect of binder density to the distance and compactness of the coating. Table 2 shows the various density of the binder with different molarity and fig 7 shows the SEM test result.

Table 2. Density of the alumina-phosphate ceramic coating

	Density (g/cm^3)
Al/P (25:75)	1,6704
Al/P (28 :72)	1,7029
Al/P (30:70)	1,7158

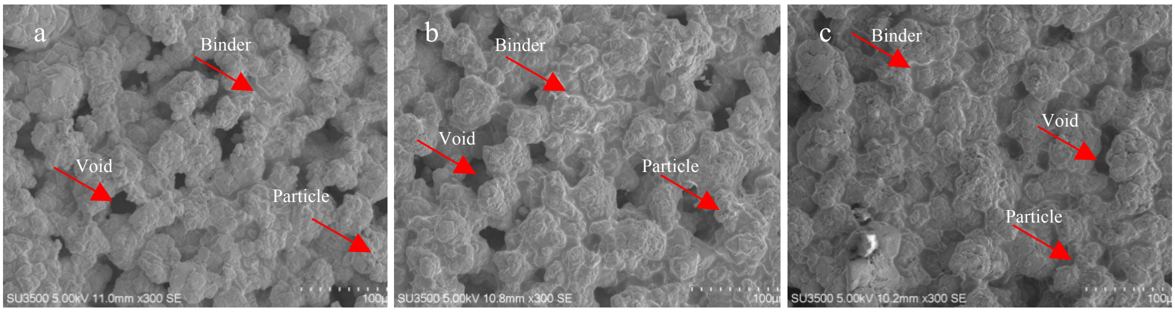

Figure 7. SEM micrograph on magnification x300, a. Al/P (25:75) , b. Al/P (28:72) dan c. Al/P (30:70).

Fig 7 shows in addition to the effect of the density, the void between the particle can also be caused by the distribution and size of the Al_2O_3 particle [12]. Void on aluminum ceramic coating was filled by the berlinite as synthesized between the binder and the Al_2O_3 particle. The reaction product will fill the space of the particle and make it compact. This condition occurrs when the product reaction has less density than the Al_2O_3 particle. Fig 7 (c) shows the product reaction of the binder with molar ratio Al/P (30:70) has less density than Al_2O_3 particle and make the coating more compact compared to other molar ratios. The compactness of the coating is important which related to the mechanical properties of the coating.

3.4 Erosion resistance of the alumina-phosphate ceramic coating

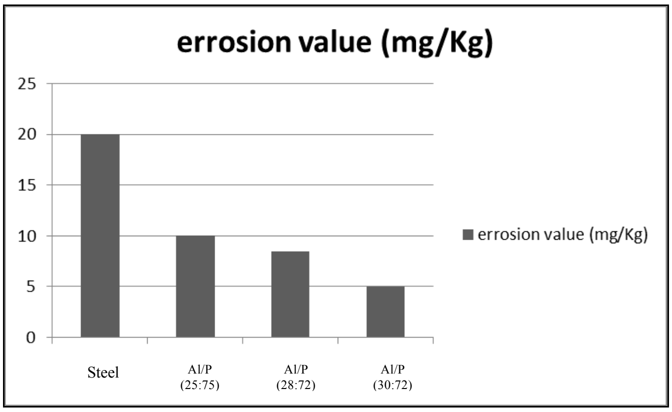

Figure 8. Erosion testing of alumina-phosphate ceramic coating

Fig 8 shows the result of erosion test, as described in the diagram, it can be seen that coating with the molar ratio of Al/P (30:70) has 5mg/Kg loss material, compare to the steel without any protection that has only 20 mg/Kg loss material. The result indicates that the erosion resistance of the coating steel is four-time than steel without any protection. This molar ratio also has higher result than other ratios, it might be caused by the compactness of the coating and the void effect between the particle as the SEM test shows before. Liping He reviewed the abrasion resistance of the coating on his research which has two-time wear durability compared to steel without coating.[3] the microstructure of the coating after erosion test can be seen in fig 9.

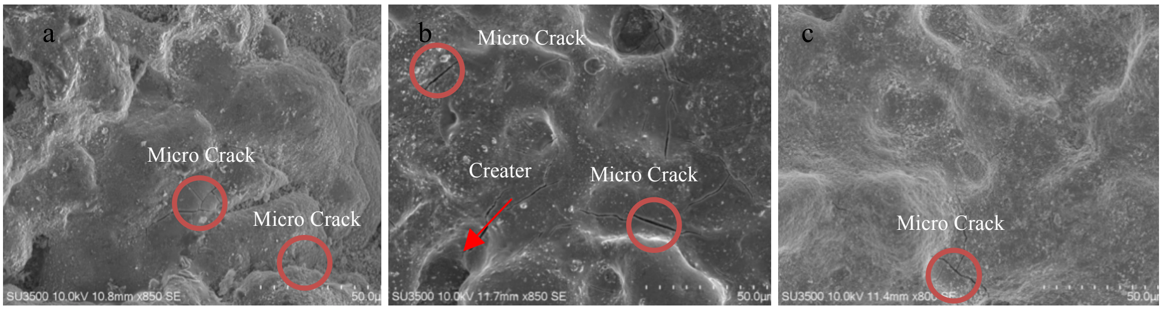

Figure 9. SEM micrograph on magnification x850 after erosion testing
a. Al/P (25:75) , b. Al/P (28:72) dan c. Al/P (30:70)

The microcrack was found on the coating after the erosion test, in which the crack can develop due to the impact from erosion particle and also an indication of mechanical properties of the binder. Mol ratio Al/P (30:70) has a less amount of crack, and it is related to the erosion test result of this mol ratio which exibit less weight loss during the test compared to other ratios. A crater was observed in the mol ratio Al/P (28:72) this crater can happen through the test.

4. CONCLUSION

The aluminum phosphate ceramic coating was successfully coated to the steel and increase erosion resistance of the steel four times than steel without any protection. The optimum mole ratio of this aluminum phosphate ceramic coating was obtained in Al/P (30:70) and heated at 220°C to form aluminum phosphate or berlinite as the best binding phase of this coating in this research.

Acknowledgments
The authors would like to say thanks to RISTEKDIKTI which funded this research as a part of *HIBAH PUSN* research grant. Authors also would say thanks to POLMAN Bandung an all colleagues for the support in all sector.

References

[1] The Balance. 2018. *The Five Sectors for Steel Applications*. [ONLINE] Available at: https://www.thebalance.com/steel-applications-2340171. [Accessed 04 June 2018].
[2] Mbabazi, J. G., Sheer, T. J., & Shandu, R. (2004). A model to predict erosion on mild steel surfaces impacted by boiler fly ash particles. *Wear*, *257*(5-6), 612-624
[3] He, L., Chen, D. & Shang, S. Journal of Materials Science (2004) 39: 4887. https://doi.org/10.1023/B:JMSC.0000035329.15101.77
[4] 92% Alumina Ceramic in Coal Mill Pulverizer, Alumina Ceramic Pipe Liner, High Alumina Ceramic for Wear and Abrasion Resistance. 2018. *92% Alumina Ceramic in Coal Mill Pulverizer, Alumina Ceramic Pipe Liner, High Alumina Ceramic for Wear and Abrasion Resistance*. [ONLINE] Available at: http://www.bmwsteels.com/alumina-ceramic.html. [Accessed 28 June 2018]
[5] Ścieszka, S., Grzegorzek, W., & Żołnierz, M. (2010). Simulative tribo-testing of erosive wear for coal impact mills. *Tribology• Reliability• Terotechnology Diagnostics• Safety• Eco-Engineering Tribologia• Niezawodność• Eksploatyka Diagnostyka• Bezpieczeństwo• Ekoinżynieria*, 37.
[6] Troczynski, T., & Yang, Q. (2001). *U.S. Patent No. 6,284,682*. Washington, DC: U.S. Patent and Trademark Office.
[7] Chen, Z., Zhang, L., & Zhou, K. C. (2009). Research progress of phosphate inorganic binder for high temperature resistance [J]. *Materials Science and Engineering of Powder Metallurgy*, *2*(14), 74-82.

[8] . 2018. . [ONLINE] Available at: https://www.ceramicindustry.com/articles/94152-all-inorganic-ceramic-performance-coatings. [Accessed 03 June 2018].
[9] Wang, C., Liu, W. B., Liu, J. J., & Su, T. (2007). Study on Heat-resistant Phosphate Adhesive [J]. *Chemistry and Adhesion*, *2*, 004.
[10] Lv, F. T., Wang, L. G., & Wang, E. Z. (2013). The preparation of phosphate bonding agent and its application in ceramic coating. In *Advanced Materials Research* (Vol. 821, pp. 1256-1260). Trans Tech Publications.
[11] Formanek, B., Szymański, K., Szczucka-Lasota, B., & Włodarczyk, A. (2005). New generation of protective coatings intended for the power industry. *Journal of Materials Processing Technology*, *164*, 850-855.
[12] Wagh, A. S., & Jeong, S. Y. (2003). Chemically bonded phosphate ceramics: I, a dissolution model of formation. *Journal of the American Ceramic Society*, *86*(11), 1838-1844.
[13] Cotell, C. M., Sprague, J. A., & Smidt, F. A. (1994). ASM handbook. *Surface engineering*, *5*, 784-804.
[14] Colonetti, E., Kammer, E. H., & Junior, A. D. N. (2014). Chemically-bonded phosphate ceramics obtained from aluminum anodizing waste for use as coatings. *Ceramics International*, *40*(9), 14431-14438..