Erosion behaviour of alumina ceramic coating on mild steel by the modified composition of phosphate binder

D Idamayanti¹, D Ginanjar¹, B Bandanadjaja¹, W Purwadi¹, and N Lilansa²
¹ Foundry Engineering Department, Politeknik Manufaktur Bandung, Jl. Kanayakan no. 21 Bandung, West Java, Indonesia
² Automation Engineering Department, Politeknik Manufaktur Bandung, Jl. Kanayakan no. 21 Bandung, West Java, Indonesia

E-mail : idamayanti79@gmail.com

Abstract. The Pulverizer pipe made of mild steel had erosion failure due to coal dust impacting, thus its service life also reduces. The ceramic coating overlay on the surface of mild steel is one of the appropriate ways to protect the mild steel from erosion. This research is aimed to perform a ceramic coating over the surface of the mild steel using a dipping method to improve its erosion resistance by using the alumina-phosphate ceramic coating. The coating layer is formed by the reaction between monoaluminum phosphate (MAP) as a binder and Al₂O₃ particles. It transforms into berlinite phase when heated at an elevated temperature. The observation is carried out with the variation of the MAP binder composition Al:P (30:70) and the Al₂O₃/MAP slurry is given at 40/60. Scanning electron microscopy is used to characterize the coating morphology. X-ray diffraction is applied to investigate the ceramic coating phases. The gas erosion jet measures erosion resistance of the ceramic coating. From the test result, it can be concluded that the binder composition influenced the erosion behaviour of alumina ceramic coating, the binder with Al:P (30:70) showed the erosion resistance increasing four times compared to the condition without coating.

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 to 300 ºC. Pulverizer pipe and inner cone which made of mild steel is impinged by coal dust particle and exposed to heat continuously, then the erosion takes place. The failure of the Pulverizer pipe due to coal dust erosion demonstrates in figure 1.

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].
Figure 1. Erosion failure of pulverizer pipe.

Alumina ceramic coated 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 itself. 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 a 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 an 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. 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. They observed the performance of phosphate adhesive which contains Al(HPO$_4$)$_2$ as the major constituent, has a heat-resistance of 1500ºC.

In this study, the variety of mole ratio Al/P as a phosphate binder for alumina ceramic that coated mild steel were 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$_2$O$_3$) as a filler and Al(OH)$_3$ – H$_3$PO$_4$ as a raw material of binder. Alumina (Al$_2$O$_3$) that has a size of 50 – 60 µm was obtained from the local market. All materials that are used come up with a 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 (MAP) binder as the equation (1).

$$3\text{H}_3\text{PO}_4 + \text{Al(OH)}_3 \rightarrow \text{Al(H}_2\text{PO}_4)_3 + 3\text{H}_2\text{O}$$ (1)

The mole ratio of Al/P was varied to find out the influence of the binder composition to the erosion behavior of the coating material as shown in Table 1.

Table 1. The mole ratio of Al/P in the synthesis of monoaluminum phosphate binder.

| 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 %           |
| H₃PO₄     | 55 %                            | 52 %           | 51 %           |
| Aquades   | 33 %                            | 34 %           | 33 %           |

This mixed solution was reacted at 100° - 120° Celsius 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.

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. Heat treatment of the aluminum-phosphate ceramic coating.

2.4. Characterization
Several tools were used to characterize the coating. Optical microscope (OLYMPUS SZ61) was used to figure the coating surface, Scanning Electron Microscope (Hitachi SU3500 at 5kV and 40% spot intensity) 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 as depicted in figure 4. It showed the alumina was high purity.

Figure 4. Elemental analysis of alumina by EDS.

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 to operate at 30 kV and 30 mA scanning at $10^\circ$ to $60^\circ$. 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 [10], reviewed that aluminum hydrogen phosphate or $\text{Al(H}_2\text{PO}_4)_3$ is the phase being expected to be present on this binder which can be reacted with the aluminum oxide particle to form berlinite. The XRD result can be seen in figure 5.
The peak shows other compound aluminum hydrogen phosphate hydrate $\text{Al(H}_2\text{PO}_4\text{nH}_2\text{O)}$. This phase was detected 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. Aluminum hydrogen phosphate hydrate was formed as much as aluminum hydrogen phosphate based on their height of intensity peaks.

![XRD result of synthetic binder.](image)

**Figure 5.** XRD result of synthetic binder.

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 Colonetti research [11]. Colonetti reviewed that crack and porosity influence to the mechanical properties of the coating.

![SEM micrograph of surface morphology of the coating affected by mole ratio of Al/P](image)

**Figure 6.** SEM micrograph of surface morphology of the coating affected by mole ratio of Al/P
a. Al/P (25:75) b. Al/P (28 :72) c. Al/P (30:70).

As seen in figure 6, the surfaces of the coating has a uniform particle distribution, leak and fissures are observed in this coating. The mole ratio of Al/P leads the binder ability to form the compacted ceramic coating. The more the mole ratio of Al in Al/P the denser of the coating becomes.

Wagh et al [10] suggest that the bonding phase is a converted from a very small amount of alumina. $\text{Al}_2\text{O}_3$ particles disperse in the $\text{Al}_2\text{O}_3$-berlinite system and the product is a thin coating of berlinite on the alumina. As depicted in figure 6 (a-c), the increase of P ratio (as $\text{H}_3\text{PO}_4$) creates the amount of porosity due to gas formation when the dipping process. This phenomenon relates with the increase of MAP acidity which will react with mild steel surface and then releases hydrogen gas.
XRD test was used to characterize the compound of the coating with binder ratio Al/P (28:72). Figure 7 shows the main compound of this coating are aluminum oxide, cristobalite and little amount of berlinite in 20º-30º. Berlinite is described as a small peak in a diagram, since it serves only as a binder in the material. He et al [3], and Colonetti et al [11] also detected berlinite at 20º - 30º with a small peak.

3.3. Effect of density to an 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 [10] observed the effect of binder density on the distance and compactness of the coating. Table 2 shows the various density of the binder with a different mole ratio of Al/P.

| Density (g/cm³) |  |
|---------------|---|
| Al/P (25:75)  | 1.6704 |
| Al/P (28:72)  | 1.7029 |
| Al/P (30:70)  | 1.7158 |

Figure 8 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 Al₂O₃ particle [10]. Void on aluminum ceramic coating was filled by the berlinite as synthesized between the binder and the Al₂O₃ particle. The reaction product will fill the space of the particle and make it compact. This condition occurs when the product reaction has less density than the Al₂O₃ particle. Figure 8 (c) shows the product reaction of the binder with molar
ratio Al/P (30:70) has less density than Al$_2$O$_3$ 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 behavior of the alumina-phosphate ceramic coating

Figure 9 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.

![Erosion Rate vs Mole Ratio](image)

**Figure 9.** Erosion testing of alumina-phosphate ceramic coating and uncoated mild steel.

The result indicates that the erosion resistance of the coating steel is four-time than steel without any protection. This molar ratio also has a 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. He et al [3] reviewed the abrasion resistance of the coating on their research which has two-time wear durability compared to steel without coating. The microstructure of the coating after erosion test can be seen in figure 9.

![SEM Micrographs](image)

**Figure 10.** 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 exhibit 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 mole ratio of Al/P in MAP binder influence the properties and erosion behavior of alumina ceramic coating. 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 best 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 binding phase of this coating in this research.

References
[1] Bell T 2018 Steel Applications What Is Steel Used For?
[2] Mbabazi J G, Sheer T J and Shandu R 2004 *Wear* 257 612–24
[3] He L, Chen D and Shang S 2004 *J. Mater. Sci.* 39 4887–92
[4] BMW steel team Alumina Ceramic
[5] Ścieszka S, Grzegorzek W and Żołnierz M 2010 *Sci. Probl. Mach. Oper. Maint.* 4 37–54
[6] Troczynski T and Yang Q 2001 *US Pat.* US 6,284
[7] Chen D, He L and Shang S 2003 *Mater. Sci. Eng. A* 348 29–35
[8] Wagh A S and Dicosola G 2014 All-Inorganic Ceramic Performance Coatings *Ceram. Ind. Mag.*
[9] Chao W, Wen-bin L, Ji-jiang L and Tao S 2007 *Chem. Adhes.* 02
[10] Wagh A S and Jeong S Y 2003 *J. Am. Ceram. Soc* 86 1838–44
[11] Colonetti E, Hobold E and Noni A De 2014 *Ceram. Int.* 40 14431–8

Acknowledgments
The authors acknowledge 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.