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Relationship between the process parameters and the saturation point in electrophoretic deposition

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Abstract

We used the electrophoretic deposition (EPD) process to fabricate a composite with glass frit and investigated the EPD parameters to find the optimum deposition time by understanding the relationship between the process parameters of zeta potential (ZP), pH, deposition yield and saturation point in a slurry. A binder and a dispersing agent were mixed properly with glass frit (0.2–25 μm, d_{50} = 8.77 μm) in an ethyl alcohol medium for the preparation of the slurry. The pH and ZP were in an inverse relationship to each other due to the generation of H_3O^+ ions with the addition of the dispersing agent in the slurry. The acidic nature of the slurry resulted in a decrease of the pH and an increase of the ZP. Otherwise, the pH increased with the addition of the glass frit in the slurry because H_3O^+ ions were absorbed on the glass frit. Therefore, the OH^- ions correspondingly increased. The saturation point of EPD was strongly correlated with the variation of the pH in the slurry; this is caused by a chemical reaction between the ethyl alcohol and the ions that make up the glass frit. An adjustment of the pH variation and the saturation point in the slurry can be established with respect to the optimum deposition time in the slurry.

Keywords: Electrophoretic deposition; Glass powder; pH; Saturation point; Zeta potential

1. Introduction

The electrophoretic deposition (EPD) method has been widely applied for the coating and infiltration in ceramic areas because the process is economically viable considering the lower cost of the raw materials and considering that the processing timing is fast comparing to other deposition techniques [1–3]. The EPD provides suitable means of fabricating ceramic–matrix composites that are reinforced with fibers [3–5] and for fabricating porous layers as membranes [6], laminated structures [7] and thermal barrier coatings [8] due to their advantages of less limited substrate shapes and the capability to scale-up to high rates of production.

A number of the EPD have been used to fabricate the composite using glass frit [9–11] but noticed that the optimum deposition time for controlling the deposition yield and the layer was rarely considered [11,12]. Reasons for setting the deposition time were not mentioned as well [9,10,13–15]. In addition, the adjustment of the voltage during the deposition process is very difficult if used to control the porosity and the shape of the deposition layer [10,11]. Therefore, understanding the relationship among processing factors in EPD method can contribute to acquire optimum condition of infiltration and coating for fabricating glass composite.

To explain the origin of the potential drop over the deposit during the EPD process for an efficient process operation and the optimum deposition time, a number of studies have investigated the origin of ion transport and related theories using an electrostatic double layer [16–19]. Some suggest that ions move with particles in a slurry and that this is related to the thickness of the electrostatic double layer and to the magnitude of the surface potential of the particles. Furthermore, several papers have focused on other parameters, such as the behavior of differently sized powders during the EPD process [20], the effect of the suspension's composition [21], and the concentration of the material [22] for an effective fabrication or deposition. However, given that these studies were based on the deposition of single-crystal material such as Al_2O_3 [16–19,21] and MgO [22], the saturation point of glass frit during the EPD

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process was not directly addressed. As glass frit is a multi-component system which contains more than one oxide element, it is important to analyze the parameters of the deposition of glass frit when it is used in the EPD process.

The main purpose of this study is to find a correlation between the process parameters for an optimum deposition time during the EPD process regardless of the raw materials or the purpose of the deposition. We investigated the internal state of a type of slurry by specifically preparing the slurry, and we succeeded in the deposition using the optimum slurry condition. The optimum deposition time during the EPD process is established considering the saturation point of the deposition yield, which occurs under the optimum slurry condition. The internal characteristics of the slurry were investigated to evaluate the effect of the raw materials that make up the slurry, in this case glass frit, a dispersing agent and a binder, all of which were tested at various ratios. The establishment of the optimum deposition time was determined from the relationship among the pH, zeta potential (ZP), and the saturation point of the deposition and the leaching ions.

2. Experimental procedure

The glass frit used for EPD process was $7SiO_2$–$9B_2O_3$–$18Al_2O_3$–46ZnO–7MgO–$4.5Na_2O$–8.5CaO system in this study. The approximate size of the glass frit was 0.2–25 μm (d_{50} = 8.77 μm), and a dispersing agent (DISPERBYK-103, BYK Additives and Instruments, Germany) and a binder (Butvar B-98, Solutia, USA) were dispersed in ethyl alcohol (95.0%, Sigma–Aldrich, USA) with the glass frit to improve the dispersibility and adhesive property. The slurry was dispersed properly using an ultrasonic bath (JAC-2010, Kodo Technical Research Co. Ltd., Korea) for 5 min, with a subsequent dispersion process conducted by means of mixing with a magnetic stirrer for 30 min to improve the dispersion of the glass frit in the slurry. EPD experiments were performed at room temperature at a constant voltage of 80 V using a DC supply (OPE-1501DI, ODA Technologies Co. Ltd., Korea). At this voltage, the current was maintained at 0.1–0.2 mA. The distance between the stainless-steel square-shaped electrodes was 1.5 cm.

To investigate the effect of the raw materials and the internal characteristics of the slurry, glass frit, a dispersing agent and a binder were added to ethyl alcohol at various ratios while simultaneously measuring the pH and ZP with a Zeta-meter (Zetaprobe analyzer, Colloidal Dynamics Inc., USA). The optimum slurry condition was chosen after considering the deposition yield and the layer characteristics of fifteen types of slurry conditions. The optimum slurry was used to find the relationship between the process parameters so as to determine the optimum deposition time in the EPD process. The deposition weight was continuously measured every 30 min during the process to obtain the maximum deposition yield and saturation point. The pH was measured by a portable pH meter (CP-401, Elmetron, Poland) for continuous measurements from inside the EPD bath to analyze how the pH varied. An ICP-OES device (Optima 7300DV, PerkinElmer Inc., USA) was used to examine the amount of leaching ions by comparing the results 5 min and 300 min after the slurry was manufactured to understand the cause of the saturation point and the variation of the pH during the EPD process.

3. Results and discussion

Two results able to predict the relationship between the parameters and the optimum deposition time were obtained by the EPD experiments. The first is the characteristics of slurry as influenced by the effect of the raw materials, and the second is the variation of the slurry characteristics, in this case the pH, ZP and deposition yield during the EPD process. The glass frit was deposited on the cathode which had a positive ZP value due to the presence of H_3O^+ ions. The H_3O^+ ions are greater in number compared to the OH^- ions in the slurry because the dispersing agent, which is acidic, generated H_3O^+ ions to create an acidic slurry with a positive ZP.

The pH and ZP with the variation of the glass frit content showed an inverse relationship (in Fig. 1). This result was in accordance with several previous studies [4,23,24]. The ZP shows a tendency to decrease with an increase in the frit content, whereas the pH increases inversely (neutralization). The increased pH with the addition of glass frit is related to the frit absorbing the H_3O^+ ions. Therefore, the OH^- ions increased relatively and the pH became neutral. In contrast, the ZP, which is the potential difference, was lower than before because the number of absorbed H_3O^+ ions on each piece of glass frit was decreased by the increase in the surface area due to the addition of the glass frit. This weakens the surface charge layer based on the electric double layer [25]. Fig. 2 shows this procedure, in which the number of adsorbed ions decreased after the addition of the glass frit, leading to the variation of the pH and the ZP.

The binder does not affect the pH, as shown in Fig. 3, because the binder is not associated with the ion generation and the adsorption in the slurry. However, Van der Waals force arises due to the long linear polymer, which is generated by the binder [26,27]. Hence, the glass frit is dispersed by the Van der Waals force. Eventually, the number of ions that are adsorbed on each piece of glass frit can be reduced, as shown in Fig. 4,

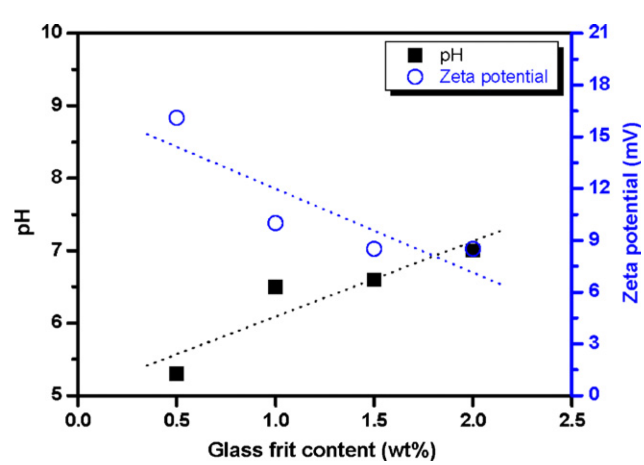

Fig. 1. Relationship between the pH and ZP with the glass frit content.