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DIRECT ELECTROPHORETIC DEPOSITION OF ALUMINA PARTICLES ONTO POROUS CERAMICS FOR MEMBRANE FILTER APPLICATION

Jun-ichi Hamagami, Kiyoshi Kanamura, Takao Umegaki,
Norihiko Fujiwara*, Masafumi Ito* and Shinji Hirata*
Department of Applied Chemistry, Graduate School of Engineering, Tokyo Metropolitan University
1-1Minami-Osawa, Hachioji, Tokyo 192-0397, Japan
Fax: +81-426-77-2821, e-mail: hamagami-junichi@c.metro-u.ac.jp
*Research and Development Dept., Araco Co., 1-34 Kotobuki, Toyota, Aichi 471-0834, Japan
Fax: +81-565-28-2535, e-mail: Masafumi-itou@arnotes.araco.co.jp

An electrophoretic deposition (EPD) process using water, followed by sintering, was applied to the formation of a separation layer of ceramic membrane filter. In the EPD process, the substrates (electrode) on which powders in suspension were deposited, are confined to electrically conductive materials, such as metal, because of the principle of EPD process. In this work, we select nonconductive porous Al_2O_3 tube or plate as substrate in the EPD process. By placing the porous ceramic substrate between two electrodes in the EPD suspension, the Al_2O_3 powders suspended in water were deposited directly onto the ceramic substrate. The thickness of the separating layer can be controlled by the EPD processing parameter such as powder concentration of suspension, applied electric field strength or current density, and deposition time. The electrophoretically fabricated Al_2O_3 composite specimens functioned as a ceramic membrane filter. This was confirmed from the result of a filter test using pond water involving bacteria. This fabrication technique provides a new processing route with cost-effectiveness and shortening in the time for the formation of separating layer in ceramic membrane filter.

Key words: electrophoretic deposition (EPD), fabrication process, ceramic membrane filter, alumina, aqueous suspension

1. INTRODUCTION

Development of ceramic or polymeric membrane filter for water purification is important from the viewpoint of water quality improvement of daily life drainage and industrial effluent [1,2]. At present, the polymeric filter is the mainstream, but it is inferior to low temperature operation (typically less than 200 °C) and the chemical durability owing to its organic nature. On the other hand, the ceramic membrane filter is more excellent than the polymeric filter in operation temperature and chemical stability. However, the ceramic filter is limited for the membrane permeability of the water. To solve the above problem, the development of the thin film fabrication technology for the separating layer in the ceramic membrane filter is required. Therefore, various chemical and physical processes have been applied to the fabrication for the separating layer in ceramic filter in order to obtain the high filter performance [1-3].

An electrophoretic deposition (EPD) process, followed by sintering, is one of promising methods for fabrication of thin or thick films and composite materials in ceramic field [4-16]. The advantages of the EPD process have relatively simple apparatus, mass production, low cost, short fabrication time, and environmental friendly process by using water as the dispersion solution [5]. The process of EPD is considered that the charged particles in a suspension are migrated to electrode (substrate) by the potential gradient between electrodes, and then the particles are deposited and aggregated on the electrode. In above EPD principle, the substrate materials should have high electron conductivity, such as metal and alloy. When ceramics without the electron conductivity are used as the substrate in the EPD process, a pre-coating on the surface is required to provide a electron conductivity of the surface [16]. One of the purposes of this study is to propose the EPD technique that is a deposition of suspended powders directly onto the porous ceramic substrate without electron conductivity.

In this paper, we propose a new processing route for the fabrication of separating layer in ceramic membrane filter by using aqueous EPD process combined with sintering [17]. We select a tube-type Al_2O_3 ceramic membrane filter with double-layered structure. The EPD conditions, such as powder concentration in the suspension, applied constant current strength, and deposition time were examined to form the separation layer with the uniform thickness. Furthermore, the filter test of the electrophoretically fabricated specimen was carried out using bacteria-containing water in the natural pond.

2. EXPERIMENTAL PROCEDURE

2.1 Materials

Commercial α-Al_2O_3 powder (Sumicorundum AA-07, Sumitomo Chemical Co., Ltd.) with an average particle size of 0.78 μm was chosen to produce the separating layer in ceramic membrane filter. Since the powder was manufactured by CVD process, it is almost spherical and its particle size distribution is narrow (0.5-0.8 μm), as shown in Fig. 1. Suspension media was a commercial distilled water (pH 5.7) for high performance liquid chromatograph (HPLC) purchased from Wako Pure Chemical Industries, Ltd. The Al_2O_3 powder and the water were used in the EPD process

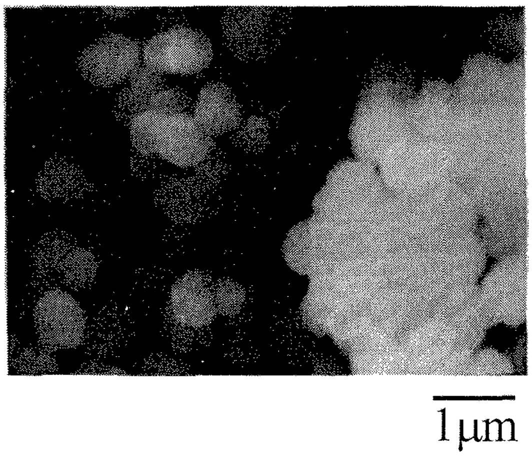

Fig. 1. Scanning electron micrograph of typical Al_2O_3 powders (Sumicorundum AA-07) used as dispersed particle for EPD suspension.

without further purification. Before EPD process, aqueous suspension was prepared by dispersing the Al_2O_3 powders in the water. The concentration of dispersed powders was 1 to 5 mass %. Porous Al_2O_3 tube of 70 or 240 mm long, 10 mm outer diameter, and 2 mm wall thickness was used as substrate (supporting body for ceramic membrane filter). The porous ceramic tube by sintering at 1500 °C had 35% porosity and permeable to water.

2.2 EPD setup and conditions

Figure 2 shows schematic diagram of EPD cell used for the tubular porous ceramic substrate. The porous Al_2O_3 tube is placed between two electrodes. Platinum (Pt) wire (1 mm in diameter) and cylindrical Pt mesh were used as inner and outer electrodes, respectively. The Pt wire sets approximately center of the Al_2O_3 tube. The Pt mesh was in cylindrical shape (diameter became 30 mm). The distance between the electrodes was set 15 mm. Before EPD process, the Al_2O_3 powder was sufficiently dispersed by ultrasonic cleaning machine and magnetic stirrer. The electrodes were connected to

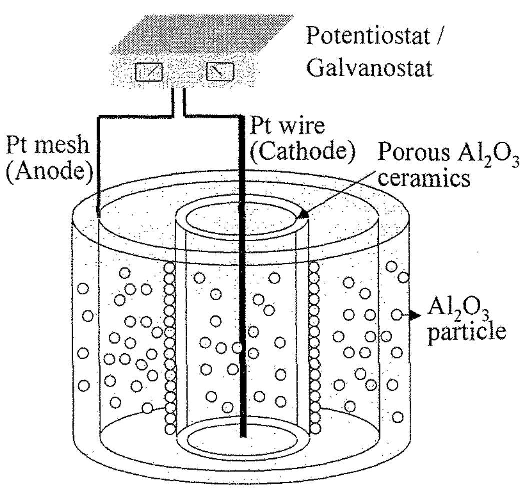

Fig. 2. Schematic illustration of EPD cell for tubular porous ceramic substrate. The substrate was placed between electrodes in aqueous suspension.

Potentiostat/Galvanostat equipment (HA-301, Hokuto Denko Co.). The EPD was carried out under a constant-current method. The current was varied from 10 to 20 mA, and the deposition time was varied from 15 to 30 s. The suspension was continuously stirred using a magnetic stirrer to maintain the uniformity of the solution during deposition.

After EPD forming, the specimens was sintered at 1450 °C for 1 h in air in order to enhance adhesion not only between the electrophoretically deposited particles but also the coated layer and the substrate.

2.3 Characterization

Scanning electron microscope (SEM: JSM-5310, JEOL) was used to examine the surface morphology of the obtained green and sintered specimens and thickness from cross-sectional observation. Maximum pore size and the pore size distribution of the sintered specimen was measured by Perm-Porometer (CFP-1200AEXL, Porous Materials Inc.) [18]. Bacteria-containing water in the natural pond was used to determine the filtration characteristic of the electrophoretically fabricated tubular ceramic specimens.

3. RESULTS AND DISCUSSION

The Al_2O_3 particles suspended in water were deposited onto the tubular porous Al_2O_3 substrates by the EPD process. In the experimental setup, the deposition occurs the outer surface of the ceramic tube. After the EPD process, the formation of the film consisting of aggregated spherical particle (0.78 nm size) through electrophoresis was observed with SEM. It is well known that solid oxide powders in aqueous solution possess a pH-dependent surface electrical

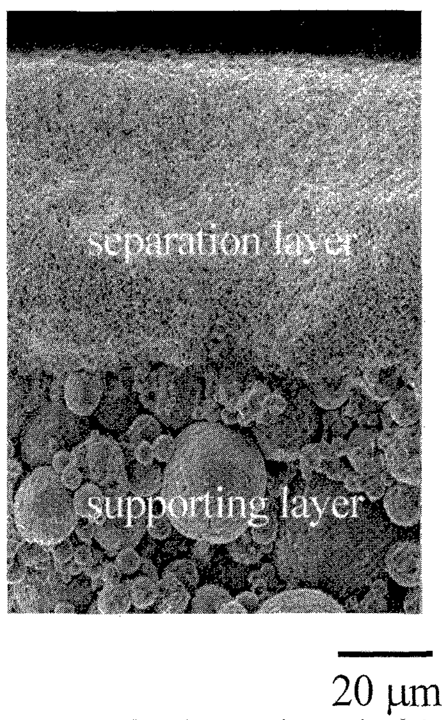

Fig. 3. Scanning electron micrograph of the cross-sectional view of electrophoretically formed Al_2O_3 layer on porous Al_2O_3 substrate sintered at 1450 °C for 1 h in air. The film was formed outer surface of porous Al_2O_3 tube.

charge. The adsorption of protons onto the surface sites of Al_2O_3 is believed to be potential-determining ions. Al_2O_3 powders in water (pH 4.9 in this experimental) have positive surface charge such as $AlOH_2^+$ in the acidic pH region. Therefore, the positively charged Al_2O_3 powders were migrated to cathode electrode by applying the electric field difference between two electrodes in EPD process. More detailed investigation is required for elucidating the deposition mechanism onto porous ceramic substrate by electrophoresis.

Figure 3 shows the scanning electron micrograph of the cross section of outer surface of the tubular Al_2O_3 ceramic specimen consisting of double-layered structure.

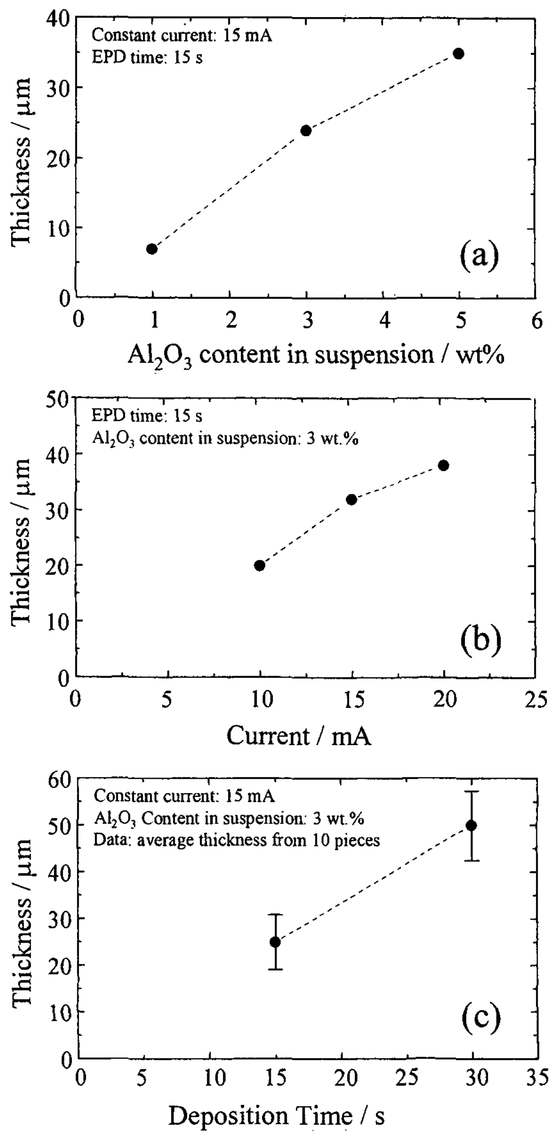

Fig. 4. Thicknesses of the Al_2O_3 thin film, which was prepared using a constant-current EPD in aqueous suspension followed by sintering at 1450 °C for 1 h, as a function of (a) Al_2O_3 content in EPD suspension, (b) current, and (c) deposition time of EPD. The dimensions of porous Al_2O_3 tube were length of 240 mm, diameter of 10 mm, and wall thickness of 2 mm. Other experimental parameter in EPD method is shown in the Figure (a) to (c).

This specimen was obtained by the EPD forming and sintering at 1450 °C for 1 h in air. The top layer is the separation layer prepared by a constant-current EPD process from aqueous suspension, and the bottom layer is the porous Al_2O_3 supporting body (70 mm in length, 10 mm in diameter, and 2 mm in wall thickness) as substrate. The porous Al_2O_3 substrate with permeability is composed of large Al_2O_3 powders less than 10 μm size and many open pores, shown in Fig. 3. On the other hand, a fine Al_2O_3 layer with uniform thickness was formed onto the tubular porous Al_2O_3 substrate.

The effect of Al_2O_3 powder concentration in the suspension (1-5 mass %), applied current strength (10-20 mA), and deposition time (15-30 s) for a constant-current EPD process on the thickness of Al_2O_3 film is shown in Fig. 4 (a) to (c). The substrate was the porous Al_2O_3 tube (240 mm in length, 10 mm in diameter, and 2 mm in wall thickness). The film thickness was measured after sintering at 1450 °C. In this experiment, the Al_2O_3 film obtained by EPD process was formed at the outer surface of the porous Al_2O_3 tube. From the results, the thickness of the Al_2O_3 film prepared by using EPD forming and sintering increases lineally with increasing the powder concentration in suspension, applied current, and time in EPD process. It can be stated that the Al_2O_3 film thickness can be easily controlled by the change of above parameters of EPD process. The adhesion strength between Al_2O_3 support and separating layer is superior for practical use. In fact, it did not exfoliate by the mechanical cutting.

Characteristics as a porous body were evaluated by bubble point method. Figure 5 shows pore size distributions of (a) tubular porous Al_2O_3 substrate and (b) electrophoretically fabricated Al_2O_3 separating layer formed on the outer surface. The thickness of layer was 25 μm. The porous substrate had 0.75 μm of diameter at maximum pore size distribution and 1.2 μm of mean flow pore diameter. The Al_2O_3-layer coated on substrate had 0.14 μm of diameter at maximum pore size distribution and 0.17 μm of mean flow pore diameter. This ceramic membrane filter is classified into microfiltration (10 nm < pore size < 10 μm).

Filtration characteristic of the ceramic membrane filter prepared by means of the EPD combined with sintering was investigated by using bacteria-containing water from the natural pond. Figure 6 shows number of general bacteria before and after filtration using the tubular ceramic specimen having film thickness of 25 μm. Before filtration, the initial number of general bacteria was 27000 counts dm^{-1}. By passing the bacteria-containing water through the filter, the bacteria in filtered water could not be detected. It is revealed from above result that the electrophoretically fabricated tubular ceramic filter is effective for a water purification.

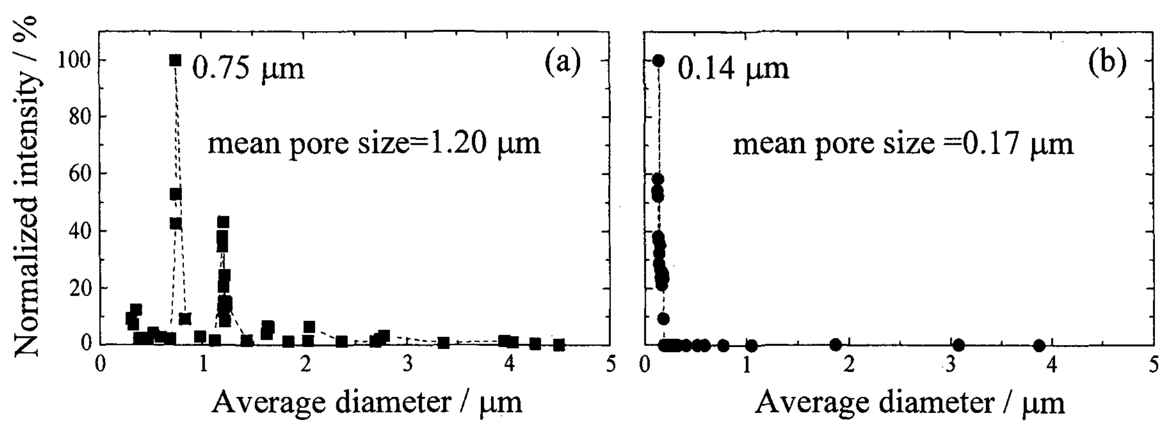

Fig. 5. Pore size distributions of the specimens of (a) porous Al_2O_3 tube used as substrate and (b) Al_2O_3 layer formed on the substrate prepared by using EPD forming followed by sintering at 1450 °C for 1 h. The data was obtained by bubble point method. The thickness of electrophoretically fabricated Al_2O_3 film was approximately 25 μm.

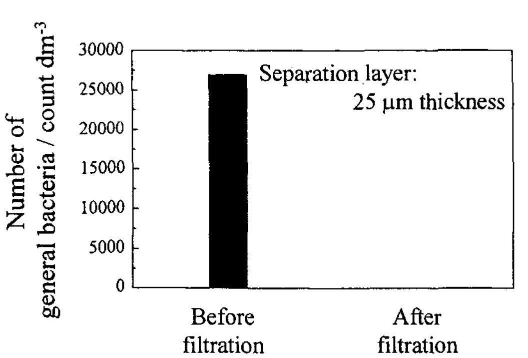

Fig. 6. General bacteria removal before and after filtration of Al_2O_3 ceramic membrane filter prepared by EPD forming and sintering.

4. CONCLUSION

Separating layer of a ceramic membrane filter with double-layered structure was fabricated by using a constant-current electrophoretic deposition (EPD) process followed by sintering. Alumina (Al_2O_3) particles suspended in water were directly deposited onto porous Al_2O_3 tube without electron conductivity by electrophoresis. The thickness of separating layer was easily controlled by changing the powder concentration in the EPD suspension, applied current strength, and deposition time in EPD process. Thus obtained Al_2O_3 ceramic composites act as ceramic membrane filter for water purification. This system provides cost-effective and short-time processing route for the fabrication of separating layer for ceramic membrane filter.

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