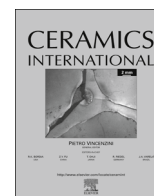




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# Preparation processes and characterizations of alumina-coated alumina support layers and alumina-coated natural material-based support layers for microfiltration

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## ABSTRACT

Recently, porous ceramic membranes have become a subject of significant interest due to their outstanding thermal and chemical stability. To reduce the high manufacturing costs of these porous ceramic membranes, recent research has focused on the utilization of inexpensive natural materials. However, there have not been any well-established direct comparisons of the membrane properties between typical alumina-based membranes and novel natural material-based membranes. Therefore, we compared alumina-coated alumina support layers (with average pore sizes ranging from 0.10  $\mu\text{m}$  ~ 0.18  $\mu\text{m}$ ), alumina-coated diatomite-kaolin composite support layers (with an average pore size of 0.12  $\mu\text{m}$ ), and alumina-coated pyrophyllite-diatomite composite support layers (with an average pore size of 0.11  $\mu\text{m}$ ) via the dip-coating method and subsequent heat treatment ranging from 1200 °C–1400 °C for 1 h. The pure water permeability of the alumina-coated diatomite-kaolin composite support layer and the alumina-coated pyrophyllite-diatomite composite support layer was found to be approximately  $2.0 \times 10^2 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$ , which is similar to that of an alumina-coated alumina support layer. Therefore, we suggest that the average pore size of an alumina-coated natural material-based support layer can be effectively controlled while exhibiting acceptable water permeability.

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## 1. Introduction

In recent years, porous ceramic materials have received great attention [1] because of their unique properties, such as low density [2], low thermal conductivity [3,4], and low dielectric constant [5]. Among the potential applications of porous ceramic materials, porous ceramic membranes [6–8] are the most feasible. In porous ceramic membranes, it is important to maintain precise control of the average pore size, mechanical strength, and permeability of the porous ceramic membranes while minimizing the material processing costs. Unfortunately, there are some limitations when reducing the processing costs; for example, only simple alterations to the processing conditions can be made while using common, yet expensive, high-purity ceramic materials, such as  $\alpha$ -alumina,  $\gamma$ -alumina, zirconia, and silicon carbide.

In this study, we employed two types of natural materials as a porous ceramic support layer prepared from (1) a diatomite-kaolin

composite (diatomite as the matrix and kaolin as a strength enhancer) and (2) a pyrophyllite-diatomite composite (pyrophyllite as the matrix and diatomite as a pore former). These materials have been selected because they are inexpensive, natural, and abundant. The reasons for investigating the diatomite-kaolin and the pyrophyllite-diatomite composite support layers are as follows:

First, diatomite is a sedimentary rock resulting from the siliceous fossilized skeletons of diatoms that are composed of rigid cell walls called frustules [9–12]. The mechanical strength of a diatomite support layer is not acceptable for a free-standing membrane, therefore, we introduced kaolin to tailor the pore characteristics of the diatomite support layer. Previously, we reported possible approaches for fabrication of a diatomite-based support layer that allowed fine-tuning of the pore characteristics [13–22].

Second, pyrophyllite is an environmentally friendly clay material that is abundant especially in the Jeollanam-do Province of Korea. Pyrophyllite is a hydrated aluminosilicate with a composition of  $\text{Al}_2\text{O}_3 \cdot 4\text{SiO}_2 \cdot \text{H}_2\text{O}$ . The crystals of the pyrophyllite possess a plate-like appearance and they are similar to talc in structure.

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Because the Al–OH bonds in pyrophyllite may also possess the capacity for fluoride adsorption [23], pyrophyllite was recently considered to be a promising new candidate material for utilization in porous ceramic membranes [24,25]. Because the permeability of a pyrophyllite support layer is not acceptable for microfiltration, we introduced diatomite to enhance the permeability of the pyrophyllite support layer [26,27].

There are practical limitations in reducing the pore size of a natural material-based support layer under 0.5  $\mu\text{m}$  due to the coarse particle size, the inherent pores inside the particles, and the irregular shape of natural materials. Most harmful bacteria, such as *Escherichia coli*, *Staphylococcus aureus*, *Salmonella*, *Legionella*, and *Pseudomonas aeruginosa*, are larger than 0.4  $\mu\text{m}$  in size [28,29]; therefore, to remove bacteria in water treatment applications, the average pore size of a feasible porous ceramic membrane must be less than 0.4  $\mu\text{m}$ . With this knowledge, we attempted to reduce the pore size of the tested natural material-based support layers by depositing an alumina coating layer over the support layer.

In previous reports, we measured only the presence of a coating layer over diatomite-based support layers by analyzing the pore size distributions and the microstructure [16–18]. Although the reliability of the coating layer, the average pore size and the permeability of the porous ceramic membrane are important factors, few studies have been conducted that discuss the reliability of the coating layer with experimental results.

Therefore, the present study investigates the following important aspects: First, we determined whether the alumina coating layer could be deposited over the diatomite-kaolin composite support layer and the pyrophyllite-diatomite composite support by the dip-coating method and the subsequent heat treatment. Second, we investigated whether the average pore size of the diatomite-kaolin composite support layer and the pyrophyllite-diatomite composite support layers could be reduced to less than 0.4  $\mu\text{m}$  by depositing a layer of alumina coating. Finally, we investigated whether the alumina coating layer deposited over the diatomite-kaolin and the pyrophyllite-diatomite composite support layers could be reliably operated during the microfiltration test.

## 2. Materials and methods

$\alpha$ -Alumina (AM-210, Sumitomo Chemical Co. Ltd., Japan), diatomite (Celite 499, Celite Korea Co. Ltd., Korea), and pyrophyllite (Korea Powder Co. Ltd., Korea) were used for the preparation of the porous ceramic support layers. The average particle sizes of the powders were determined using a particle size analyzer (LSTM 13 320 MW, Beckman Coulter, USA). The average particle sizes of the as-received alumina, diatomite, and pyrophyllite were 4.80, 7.43, and 6.95  $\mu\text{m}$ , respectively. Diatomite and kaolin were used for the preparation of the diatomite-kaolin composite support layer. Pyrophyllite, diatomite, and sodium borate (Shinyo Pure Chemical Co. Ltd., Korea) were used for the preparation of the pyrophyllite-diatomite composite support layers.

To extrude the alumina support layers, 17 wt% of methyl cellulose (from Sigma-Aldrich, USA) was added as a binder, and 20 wt% of distilled water was added as a solvent. Similarly, to extrude the diatomite-kaolin composite support layers, 15 wt% of methyl cellulose was added as a binder, and 15 wt% of distilled water was added as a solvent. Finally, to extrude the pyrophyllite-diatomite composite support layers, 17 wt% of methyl cellulose was added as a binder and 50 wt% of distilled water was added as a solvent.

The mixed slurry was aged for 48 h at room temperature and extruded using a double screw extruder (from KTE-50S, Kosentech, Korea). The extruded diatomite-kaolin composite support layer had flat tube-type dimensions (width 50 mm, height 4 mm,

and length 200 mm), and 16 inner holes (width 2 mm and height 2 mm). After extrusion, the specimens were dried for 24 h at room temperature. To burn-off the binder, the dried specimens were heat treated at 400  $^{\circ}\text{C}$  for 1 h and, finally, sintered at 1200  $^{\circ}\text{C}$ –1600  $^{\circ}\text{C}$  for 1 h.

A conventional table-top dip-coater (from E-flex, Korea) was used to deposit an alumina coating onto the alumina support layer, the diatomite-kaolin composite support layer, and the pyrophyllite-diatomite composite support layer. In the coating process, alumina (from AKP-30, Sumitomo Chemical Co. Ltd., Japan), with 0.27  $\mu\text{m}$  of average particle size, 2-propanol, ethanol, glycerol (all from Sigma-Aldrich, USA), as well as distilled water, and an organic binder (Polyvinyl alcohol 500, Junsei Chemical, Japan) were mixed, dip-coated for 10 s, retracted with a withdrawal speed of 1 mm/s, and dried at room temperature for 24 h, before being heat treated from 1200  $^{\circ}\text{C}$ –1400  $^{\circ}\text{C}$  for 1 h. The alumina concentration of the slurry was fixed at 10 wt%.

The flexural strengths of the alumina support layer, diatomite-kaolin composite support layer, and the pyrophyllite-diatomite support layer were measured by a four-point bending test using an Instron 4206 testing system. For the flexural strength test, 3  $\times$  4  $\times$  25 mm specimens were machined and tested on 20 mm outer span four-point fixtures. The pore characteristics of the specimens were investigated by scanning electron micrographs (JSM-5800, JEOL, Japan), and mercury porosimetry was determined using an Autopore IV 9510 system (Micromeritics, USA). Furthermore, the air permeability was measured by capillary flow porosimetry (CFP-1200-AEL, Porous Materials Inc., USA). A specimen (4 cm in diameter and 0.4 cm thick) was fitted between the O-rings, and the insert in the capillary flow porosimeter located in the bottom of the chamber. Finally, the flux was measured automatically by sensors, while the diameter of the motorized valve and the pressure of the regulator were incrementally changed.

The water permeability and reliability of the alumina support layer, the diatomite-kaolin composite support layer, and the pyrophyllite-diatomite support layer were measured using a dead-end microfiltration system (MTS2000, Sam Bo Scientific, Korea) equipped with particle counters (PAMAS-4132, PAMAS, Germany). The operating trans-membrane pressure was maintained at 2.5 bar.

## 3. Results and discussion

Fig. 1 shows the flexural strengths and air permeabilities of the alumina support layer, the diatomite-kaolin composite support layer, and the pyrophyllite-diatomite composite support layer. The flexural strength of the alumina support layer sintered at 1600  $^{\circ}\text{C}$  for 1 h was higher than that of the diatomite-kaolin composite support layer and the pyrophyllite-diatomite layer that were both sintered at 1200  $^{\circ}\text{C}$  for 1 h. However, considering that the air permeability of the natural material-based support layers is similar to that of the alumina support layer, it was worthwhile to investigate the feasibility of the natural material-based support layers when operated properly under low load-bearing conditions.

To eliminate the possibility of a problem related to the alumina slurry composition and/or the dip-coating conditions, it was confirmed that the alumina coating could be deposited on an alumina support layer using the optimized processing conditions. This was done prior to depositing the alumina coating layer on the diatomite-kaolin composite support layer and the pyrophyllite-diatomite composite support layer. Without these preliminary experiments, it is difficult to interpret whether a potential coating problem is caused by a compatibility problem between the alumina-based coating layer and a silica-based support layer or by a non-optimal coating process.

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