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The Prototype of Porous Ceramics for Heavy-Metal Filtration

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ABSTRACT

The efficiency of $AgNO_3$ filtration was tested on barium zirconium titanate $Ba(Zr_{0.05}Ti_{0.95})O_3$ porous ceramic, which was prepared with the tape casting and uniaxial pressing method. The relationship between electrical resistance and filtration efficiency agreed well with the appropriate pore size and percentage of porosity that should be obtained in $AgNO_3$ filtration ceramics.

Keywords: $AgNO_3$, filtration, $Ba(Zr_{0.05}Ti_{0.95})O_3$, porous ceramics

INTRODUCTION

The expectation of this paper is to prepare the porous ceramics with a high effective adsorption by using Barium zirconium titanate as the filter material. $Ba(Zr_{0.05}Ti_{0.95})O_3$ is one of the lead-free ferroelectric ceramics which have been studied for use as dielectric and piezoelectric devices (Maiti *et al*., 2006). The advantage of piezoelectric behavior would be benefit for filtration processing when the particles penetrate into the pores and cause pore clogging (Şan and Özgür, 2007).

Silver nitrate ($AgNO_3$) filtration is the focus of this work because it is a hazardous waste and is unfriendly to the environment. It is used to coat X-ray film and is extracted during the photo development process.

In addition, the porous ceramics used in this work were prepared using the tape casting process, an economical method of producing thin ceramics (Bitterlich *et al*., 2002). Basically, tape casting involves the dispersion of ceramic powders in a solvent. The addition of binder, plasticizer, and dispersant was used to control the rheology and quality of the green tapes.

In the present work, different amounts of plasticizer and dispersant were used during the slurry preparation. A comparison of filtration efficiency between bulk ceramic and tape casting ceramic was carried out through electrical resistance.

EXPERIMENTAL PROCEDURE

Ceramic powder preparation

The $Ba(Zr_{0.05}Ti_{0.95})O_3$ powder was prepared according to the conventional solid-state route using $BaCO_3$, ZrO_2, and TiO_2 as the starting materials. The powders were mixed and ball-milled for 24 h using ethyl alcohol as the milling media. Calcination of the mixed powder was carried out at 1200 ^oC for 4 h. A phase analysis of the calcined powder was made by x-ray diffraction (XRD; Philips PW 1729) with Cu-k_α radiation.

Slurry formulation

Tape casting slurries were prepared from three different formulas, as shown in Table 1. The slurries were prepared by mixing BZT powder with ethanol and 2-butanone, which were used together as the solvent. Carboxy methyl cellulose (CMC) was selected to use as the binder agent. Polyethylene glycol (PEG) and tri-ethanolamine were employed as plasticizer and dispersant, respectively.

Table 1 Composition of tape casting slurries

Formulas	I	II	III
BZT powder (vol.%)	16	16	16
Ethanol (vol.%)	15	15	15
2-butanone (vol.%)	31	31	31
CMC (vol.%)	15	15	15
PEG (vol.%)	8	15	8
Tri-ethanolamine (vol.%)	15	8	8

Porous ceramic preparation

The porous ceramics were prepared employing two processes: tape casting and uniaxial pressing. The taped products were carried out by casting the slurries on a tape casting machine with a moving blade on a polypropylene carrier film. The blade gap was adjusted to 3 mm. After drying at room temperature, the green tapes were cut into rectangles 10 x 10 mm^2 and 3 mm thick before and sintered at 1450 ^oC for 2 h with a heating/cooling rate at 5 ^oC/min.

The pellet ceramics were carried out using BZT powder mixtures of CMC, with a weight ratio of 3 : 1. The mixture was pressed into a pellet with a diameter of 16.5 mm and 5 mm thick under 4 MPa pressure for 30 sec. The binder burnout of the green ceramics took place at a heating rate of 5 ^{o}C/min up to 500 ^{o}C, followed by sintering at 1450 ^{o}C for 2 h with a heating and cooling rate of 5 ^{o}C/min.

The microstructure of the porous ceramics was observed using scanning electron microscopy (SEM) with a JEOL JSM-6380LV. Open porosity and bulk density were determined by the Achimedes method with distilled water as the liquid medium.

$AgNO_3$ filtration and electrical resistance

The porous ceramics were immersed in 1 molar of $AgNO_3$ solution for 4 days before being measured to find their resistances. The resistance of the samples was measured using a digital multi-meter (CD800a Sanwa). The relationship between filtration efficiency in porous ceramics and electrical resistance could be described as when $AgNO_3$ crystal continue filled inside the porous affects to increase the electrical current. On the other hand, the high amount of $AgNO_3$ crystal which is trapped by porous ceramics, the electrical resistance will be decreased.

RESULTS AND DISCUSSION

Phase analysis

Figure 1 shows the X-ray diffraction pattern of the BZT calcined powder and sintered ceramic. They was good agreement with the tetragonal system based on the X-ray diffraction pattern of $BaTiO_3$ (JCPDS file no. 05-0626), which showed splitting of the (100)/(001) reflection at $2\theta \sim 22^{o}$, splitting of the (200)/(002) reflection at $2\theta \sim 45^{o}$, and splitting of the (102)/(210) reflection at $2\theta \sim 52^{o}$, as well as splitting of the (211)/(112) at $2\theta \sim 57^{o}$. The secondary phases were not detected in either the ceramic powder or sintered ceramics.

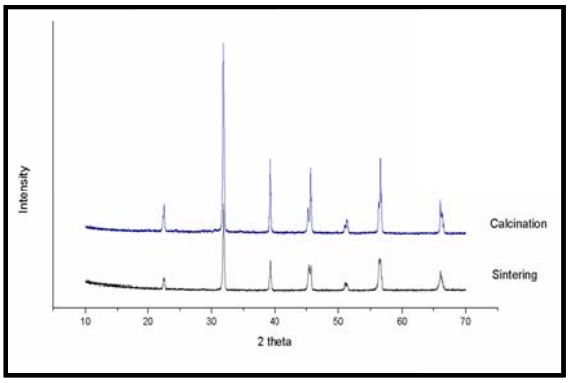

Figure 1 X-ray diffraction patterns of $Ba(Ti_{0.95}Zr_{0.05})O_3$ calcined powder and sintered ceramics.

Tape appearance

The appearance of taped products was different due to the composition of the slurries, as shown in Table 1. The formula I slurry, where the plasticizer content was higher than in other formulas, was easily poured. The dried tapes were homogeneous, crack free, and peeled away from the support easily. Conversely, for formula II and III, the plasticizer content was lower than that of formula I, suggesting that their rheologies were nearly the same compared with that of formula I. The appearances of the dried tapes, however, were significantly different. The dried tapes of formula II were covered with liquid, which resulted from a high amount of the plasticizers. As a result, a long time was spent for drying. Additionally, the dried tapes were difficult to peel away from the support and residual from the adhesion was apparent.

The slurry, which was prepared from formula III was homogeneous and required only a short time for drying. The dried tapes were smooth and crack free. However, the dried tapes exhibited a strong bonding with the support. Consequently, the tape casting which was prepared from formula III did not achieve in peeling from the support.

Microstructure of porous ceramic

Figure 2 and 3 show the SEM micrograph of the surfaces of the porous ceramics, which were prepared from tape casting and uniaxial pressing processes, respectively. The porosities were uniform and interconnected to each other. There were many large holes dispersed in the ceramic body which

resulted from evaporation of a group of binders. The pore size of the tape ceramics had values ranging from 9 – 15 μm, which was smaller than that of the pressed ceramics (Table 2). This indicated that the tape casting method can improve the particle arrangement results in particle agglomerate reduction. Table 2 shows the density, open porosity, and linear shrinkage of porous ceramics. The density of the taped products were higher than that of the pellet ceramics. This result can be explained by the fact that improvement of the particle arrangement in tape casting ceramics can contribute to high density and a low amount of porosity. The linear shrinkage in pellet ceramics were low, which should relate to a high number of liquid phases during sintering, thus resulting in a larger capillary force and higher shrinkage (Jiang *et al*., 2009; Bissett *et al*., 2008).

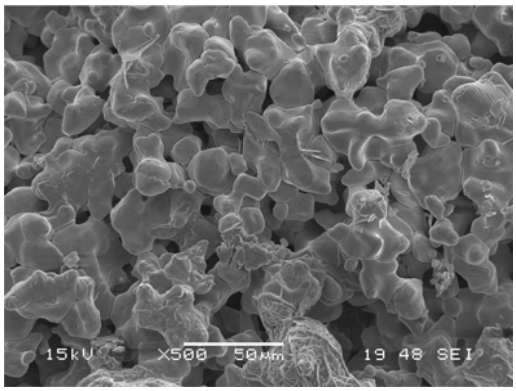

Figure 2 The microstructure of $Ba(Ti_{0.95}Zr_{0.05})O_3$ taped products at 500 x.

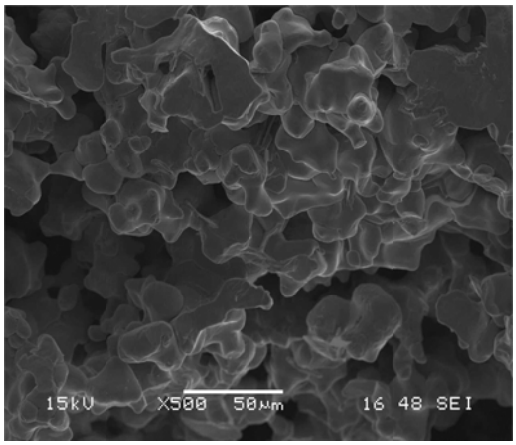

Figure 3 The microstructure of $Ba(Ti_{0.95}Zr_{0.05})O_3$ pellet ceramics at 500 x.

Table 2 The comparison of physical and electrical properties between BZT taped products and pellet ceramics.

Properties	Taped products	pellet ceramics
Average density (g/cm^3)	3.9329	2.9570
Porosity (%)	1.17 - 6.85	37.0 - 42.0
Pore size (μm)	9 - 15	15 - 19
Linear shrinkage (%)	10.50	12.39
Average Resistance (MΩ)	2.647	0.354

$AgNO_3$ filtration efficiency

Figure 4 and Figure 5 show the SEM micrographs of the surfaces of the porous ceramics, which prepared from tape casting and uniaxial pressing, respectively. The agglomeration of $AgNO_3$ particles strongly appeared on the taped products, whereas dispersion occurred on the pellet ceramics. This result indicated that the pore size of the ceramics was affected by the particle arrangement. The $AgNO_3$ particles preferred agglomerate on the taped products, whereas dispersion occurred in the pellet ceramics. This result indicated that the taped product, which was much smaller in pore size (Table 2) than that of the pellet ceramic that affected to the particle arrangement. The average electrical resistance was determined to be 2.647 MΩ for the taped product and 0.354 MΩ (Table 2) for the pellet ceramic. The result with a low electrical resistance in the pellet ceramic was correlated with the high amount of porosity, which can filter the high value of $AgNO_3$ particles. The porosity was relatively high at 42.0%, which can be advantageous when pellet ceramics are used in the filtering area because they consist of a high number in both porosity and surface area (Jiang *et al*., 2009; German, 1996).

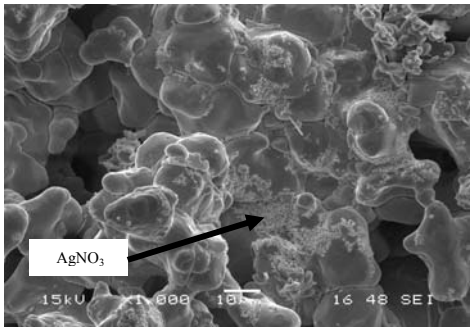

Figure 4 The microstructure of $Ba(Ti_{0.95}Zr_{0.05})O_3$ taped products with $AgNO_3$ adsorption at 1000 x.

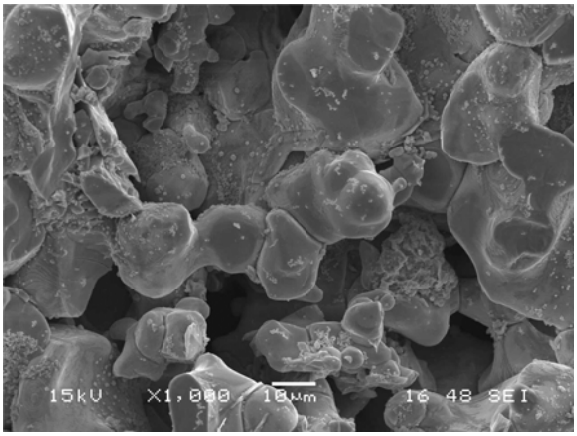

Figure 5 The microstructure of $Ba(Ti_{0.95}Zr_{0.05})O_3$ pellet ceramics with $AgNO_3$ adsorption at 1000 x .

CONCLUSIONS

The percentage of porosity rather than other properties of the porous ceramics affected the efficiency of $AgNO_3$ filtration. Optimum pore size (15-19 µm) and percentage of porosity (37.0-42.0%) were obtained in the porous ceramics, prepared with the uniaxial pressing method. The tape casting process could not produce a large amount of porosity because the powder had a regular orientation compared with another process that well agreed with its density (3.9329 g/cm^3). The result of this experiment showed that the efficiency of $AgNO_3$ filtration concerned high levels of $AgNO_3$ particles inside the porous ceramics which were determined by the reduction of electric resistance value. Therefore, the uniaxial pressing method is a suitable technique to produce porous ceramics for $AgNO_3$ filtration.

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