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Growth of Al_2O_3–PTFE composite film at room temperature by aerosol deposition method

Sung Hwan Cho [a,c], Young Joon Yoon [a,*], Hyo Tae Kim [a], Jihoon Kim [a], Hyung-Jun Kim [b], Song Min Nam [b], Hong Koo Baik [c], Jong-Hee Kim [a]

[a] Nano-IT Convergence Center, Korea Institute of Ceramic Engineering & Technology, Seoul 153-801, Republic of Korea
[b] Department of Electronic Materials Engineering, Kwangwoon University, Seoul 139-701, Republic of Korea
[c] Department of Materials Science and Engineering, Yonsei University, Seoul 120-749, Republic of Korea

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Abstract

To fabricate a ceramic-based substrate for 3-dimensional integration modules with a thick film coating process at room temperature, aerosol deposition method was employed. Al_2O_3 was chosen as a main coating material for the requirements of low permittivity and dielectric loss. Especially to give a functionality of plasticity, composite film with polytetrafluoroethylene (PTFE) was also studied. The effects of PTFE, which was incorporated in the film, were investigated by the microstructural characterization. It was confirmed that Al_2O_3–PTFE film with the grain size of 100–200 nm were grown at room temperature using Al_2O_3–0.5 wt% PTFE mixture powders. Dielectric constant and dielectric loss of Al_2O_3–PTFE film were 4.5 and 0.005 at 1 MHz, respectively.

Keywords: B. Composites; B. Grain size; C. Dielectric properties; D. Al_2O_3

1. Introduction

Aluminum oxide, α-Al_2O_3 is a promising ceramic material for integral substrate and packaging due to its low dielectric constant, low dielectric loss, high thermal conductivity and good mechanical strength. It has been commonly used as a base material for high frequency application through low temperature co-fired ceramic (LTCC) process, which can realize the system on packaging (SOP) of embedding passive components via 3-dimensional integration [1]. However, some technical issues such as shrinkage of ceramic matrix and interdiffusion at the interface of heterojunction in 3-dimensional structure formed by LTCC process are still remained because it requires the sintering temperature around 900 °C until now.

Considering the above drawbacks of conventional LTCC process, aerosol deposition method (ADM) developed by Akedo could be a useful candidate to fabricate a ceramic-based substrate for 3-dimensional integration [2–5]. The film formation via aerosol deposition was achieved by continuous impaction of starting powders accelerated from the nozzle with the speed of 200–300 m/s [3]. The main advantage of ADM is to obtain a high density film with nano-crystalline microstructure in spite of the growth at room temperature. In addition, its high growth rate about a few μm/min was so attractive to apply it to mass production with low cost. However, the high residual stress in the film was unavoidable due to continuous powder impaction and subsequent consolidation during the growth.

In this study, the possibility of Al_2O_3 thick films grown by ADM at room temperature for the application of integrated substrates for high frequency devices was examined. In addition, to give an appreciable plasticity to Al_2O_3 matrix, a direct method to fabricate the composite material between Al_2O_3 and polytetrafluoroethylene (PTFE) was tried using the powder mixture. PTFE is a well-known thermoplastic polymer, which shows low dielectric constant $\varepsilon_r \sim 2.1$ and low loss tangent tan $\delta \sim 0.0003$ over a wide range of frequencies [6]. After the optimization of composite film growth through the ADM, the dielectric constant and dielectric loss of Al_2O_3 and composite thick films were evaluated. Especially, to investigate the effect of addition of PTFE into starting powders on the dielectric properties of thick film, the change of surface microstructure and internal microstructure was monitored in detail.

* Corresponding author. Tel.: +82 2 3282 7854; fax: +82 2 3282 7838.
E-mail address: yjyoon@kicet.re.kr (Y.J. Yoon).

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2. Experimental

Aerosol deposition (AD) system is mainly composed of two parts, aerosol generation system and film deposition chamber and it is well described elsewhere [2,7,8]. Aluminum oxide (α-Al_2O_3) powders with average mean diameter of 0.52 μm and purity of 99.5% from Showa Denko (AL-160SG-3) were used. In the specification of as-received powders, impurities such as Fe_2O_3, SiO_2, Na_2O and MgO were existed but the powders with low sodium content were selected to maintain the high performance of electrical properties. In addition, to reduce the content of moisture and organic components in raw powders, the annealing was performed at 900 °C for 1 h in air ambient. Polytetrafluoroethylene (PTFE) powders with the mean diameter of 0.2 μm was used without any treatment and the quantity of PTFE was controlled below 1.0 wt%. To make an aerosol from Al_2O_3 and PTFE efficiently, the sufficient mixing of two kinds of powders was progressed in milling machine.

In AD process, the state of aerosol of raw powders could be formed by the feeding of carrier gas (helium, oxygen, and nitrogen) with the flow rate from 5 to 20 l/min and additional mechanical vibration in the vertical direction. Powders in aerosol states could be accelerated onto substrates in deposition chamber through the linear-type nozzle system. Working pressure during the deposition was maintained less than 10 Torr. Polished Cu plate was used for substrate to characterize the dielectric properties of thick films. The surface morphology and crystallinity of raw powders and as-deposited films were characterized by scanning electron microscopy (SEM, SM300,

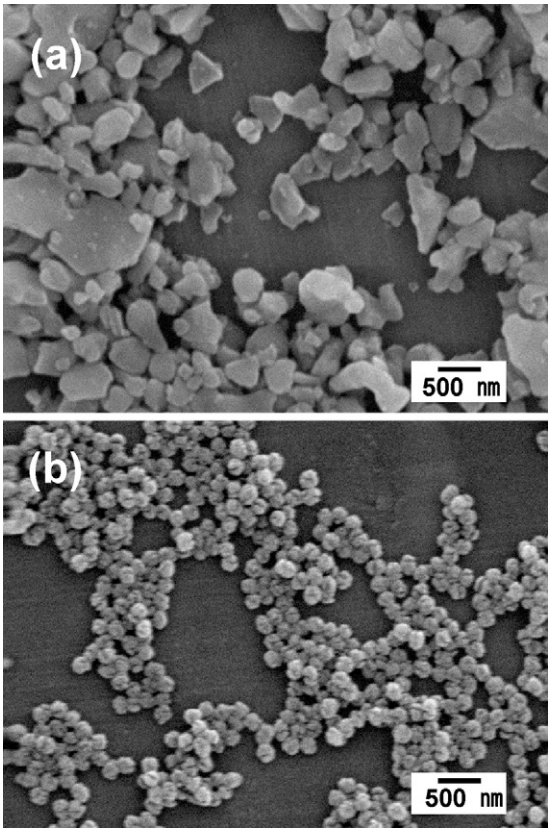

Fig. 1. Morphologies of starting powders: (a) Al_2O_3 and (b) PTFE.

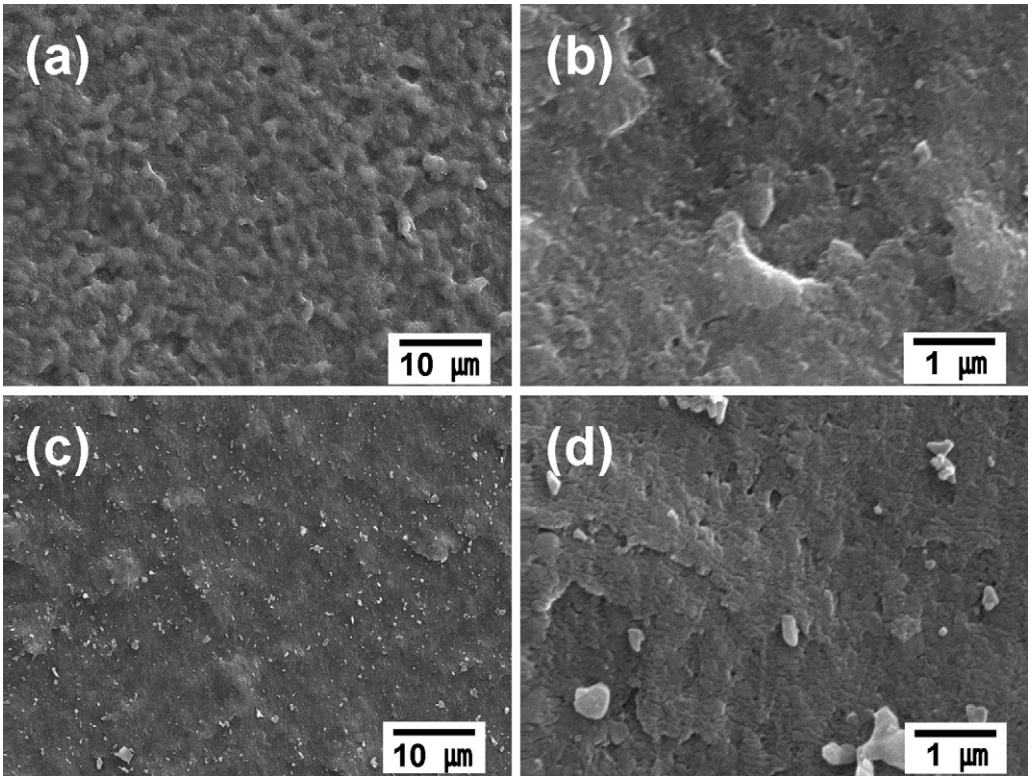

Fig. 2. Surface morphologies of as-deposited films by aerosol deposition at room temperature: (a) low magnification image of Al_2O_3 film, (b) high magnification image of Al_2O_3 film, (c) low magnification image of Al_2O_3–PTFE film and (d) high magnification image of Al_2O_3–PTFE film.

Topcon, Tokyo, Japan) and transmission electron microscopy (TEM, JEM-2000EX, Jeol Ltd., Tokyo, Japan), respectively. Cross-sectional TEM sample was prepared by focused ion beam (FIB, Helios 2000, FEI Company, Hillsboro, OR) after the coating of carbon and platinum protection layer. The dielectric properties were measured by impedance analyzer (HP4294A, Hewlett-Packard, Palo Alto, CA) in the range from 10 kHz to 10 MHz after the formation of Au upper electrode with the diameter of 1.5 mm by sputtering system.

3. Results and discussion

The morphologies of starting powders, such as Al_2O_3 and PTFE, are shown in Fig. 1(a) and (b), respectively. To grow composite film in ADM, the powder mixture of Al_2O_3 and PTFE was used varying the quantity of PTFE from 0.1 to 1.0 wt%. Physical properties of starting powders, such as size, density, morphology and composition, are critical to determine the optimum growth condition for high quality films. In our experiments, the quantity of PTFE in starting powders was optimized at 0.5 wt% due to its relatively low specific gravity compared to Al_2O_3.

Al_2O_3 and Al_2O_3–PTFE thick films were grown on polished Cu substrate at room temperature after the optimization the experimental condition. Their surface morphologies are shown in Fig. 2. As shown in Fig. 2(a) and (b), Al_2O_3 film showed the typical morphology grown by ADM containing a lot of craters on the whole surface. This phenomenon was explained by the growth mechanism of ADM. A ceramic film could be formed by the strong impaction and consolidation of starting powders with 200–300 m/s. Through the competition between growth and etching, a dense film could be formed in spite of room temperature [3]. On the other hand, the surface roughness of composite thick films grown by Al_2O_3–PTFE powders seems to be much smoother than that grown by Al_2O_3 powders. Although the quantity of PTFE in starting powders was small, it was enough to hinder the strong impaction of ceramic powders due to its low gravity.

The dielectric properties of Al_2O_3 and Al_2O_3–PTFE thick films grown on Cu plates were measured by impedance analyzer in the frequency from 1 kHz to 10 MHz. As summarized in Table 1, dielectric constant of Al_2O_3 film was similar to that of bulk but dielectric loss was increased greatly. On the other hand, dielectric constant of Al_2O_3–PTFE film was 4.5 at 1 MHz. It means that the incorporation of polymer into the Al_2O_3 matrix was successfully achieved by spraying the mixture powders through the nozzle in spite of room temperature condition. Considering the value of dielectric constant, the actual composition of PTFE in the film could be estimated about up to 50 vol.% by theoretical calculation as reported in our previous report [11]. However, the value of quality factor (Q) derived from dielectric loss of composite films was superior to Al_2O_3 film. It could be explained by the microstructural change in Al_2O_3–PTFE film.

Table 1
Dielectric and physical properties of Al_2O_3 and PTFE (at 1 MHz).

	Dielectric constant	Dielectric loss	Density (g/cm^3)
Al_2O_3 (bulk)	9.8	0.0002	4.1
PTFE (bulk)	2.1	0.0003	2.2
Al_2O_3 (AD film)	9.5	0.011	
Al_2O_3–0.5 wt% PTFE (AD film)	4.5	0.005	

Microstructural characterization of Al_2O_3 and Al_2O_3–PTFE thick films was performed and the cross-sectional TEM images are shown in Fig. 3. Fig. 3(a) showed the low magnification TEM image of as-deposited Al_2O_3 thick films on polished Cu plate. It was confirmed that a dense Al_2O_3 thick film could be well grown in spite of room temperature deposition condition. The Al_2O_3 films showed the typical images of ceramic films

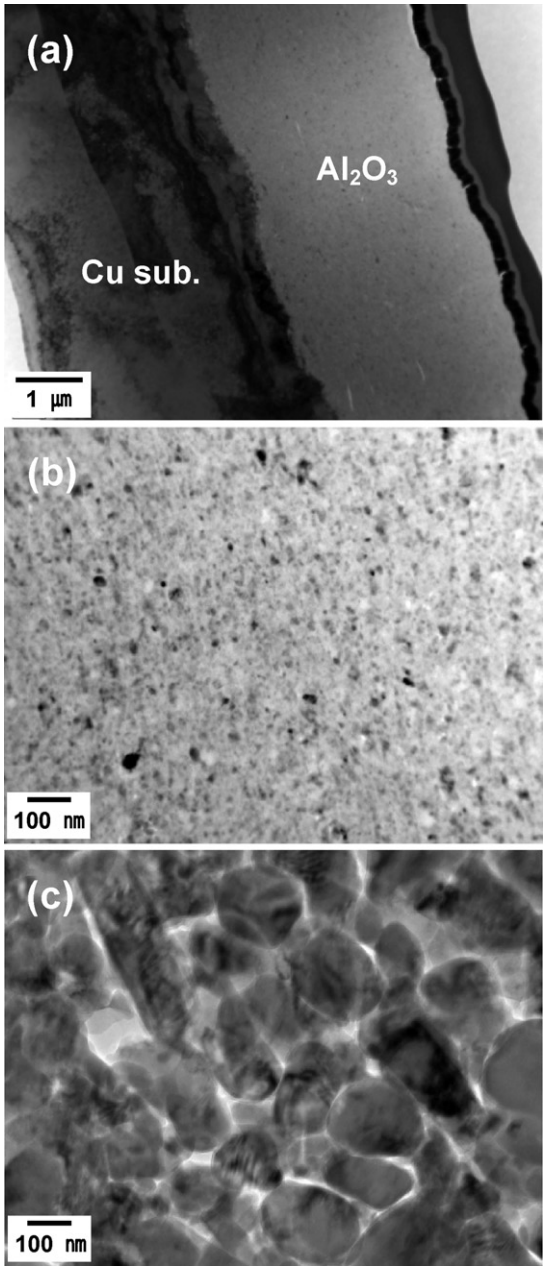

Fig. 3. Cross-sectional TEM image of as-deposited films by aerosol deposition: (a) low magnification image of Al_2O_3 film, (b) high magnification image of Al_2O_3 film, and (c) high magnification image of Al_2O_3–PTFE film.

grown by ADM, composing its microstructure of nanocrystals with the size of 5–20 nm grains as shown in Fig. 3(b). Nanocrystalline grain structure was originated from the strong impaction and consolidation of starting powders with the size of 0.52 μm. During the crushing of starting powders, it was inevitable that a large quantity of defect was formed in the crystal structure. On the other hands, the grain size of Al_2O_3–PTFE film was dramatically increased up to 10 times larger than that of Al_2O_3 films as shown in Fig. 3(c). It might be resulted from the hindrance of strong impaction of Al_2O_3 powders due to the existence of PTFE, which was a low hardness material.

Generally, dielectric loss is governed by intrinsic and extrinsic factors inside of materials [9,10]. The extrinsic factors, which were derived from crystal imperfections, such as defects, grain boundaries, porosity, and disordered orientations, almost determine the value of dielectric loss. Therefore, poor values of dielectric loss from Al_2O_3 film could be explained by the imperfections of crystals, which were generated by crushing of raw powders into nanocrystals during films growth. In the case of Al_2O_3–PTFE film, the fracture of Al_2O_3 starting powders was significantly suppressed due to the existence of PTFE powders. In addition, sufficiently low value of dielectric loss of PTFE was also helpful for composite film to show better dielectric properties compared to Al_2O_3 film.

4. Conclusions

Al_2O_3 and Al_2O_3–PTFE thick films were grown for the application of integrated substrates. Despite of room temperature condition, a dense film structure could be obtained by ADM. Dielectric property of Al_2O_3 film showed relatively poor compared to that of bulk-Al_2O_3, due to its crystal imperfections. In the case of Al_2O_3–PTFE film, dielectric constant was 4.5 and dielectric loss was 0.005 at 1 MHz. It was originated from the successful incorporation of PTFE component to Al_2O_3 matrix. It was also confirmed that the grain size of Al_2O_3–PTFE film was increased up to about 10 times compared to Al_2O_3 film.

Acknowledgements

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