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Electrophoretic deposition of alumina suspension in a strong magnetic field

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Abstract

Textured monolithic alumina ceramics were synthesized by electrophoretic deposition (EPD) in strong magnetic field of 10 T. Single crystalline, granular α-alumina particles in aqueous suspensions were rotated due to their anisotropic diamagnetic susceptibility and then deposited on substrate. A multilayered alumina composite of oriented and randomly oriented layers was also synthesized by the alternate EPD of alumina suspensions which were placed in and out of a superconducting magnet. It was demonstrated that the EPD in a strong magnetic field is a promising ceramic processing technique for fabricating sophisticated ceramic composites.

Keywords: Al_2O_3; Composites; Electrophoretic deposition; Magnetic properties

1. Introduction

The controlled development of texture in ceramics is growing focus of interest in connection with processing. Physical properties of ceramics, such as mechanical, electrical, piezoelectric, and maybe optical, can be improved by controlling crystal orientation. Many materials which have non-cubic structure have anisotropic susceptibilities, $\Delta\chi = \chi_{//} - \chi_{\perp}$, associated with the crystal structures, where $\chi_{//}$ and $\chi_{\perp}$ are the susceptibilities parallel and perpendicular to the magnetic principal axis, respectively. The energy of anisotropy is given as

$$\Delta E = \Delta_{\chi} V B^2 / 2\mu_0, \qquad (1)$$

where V is the volume of the material, B is the applied magnetic field, and μ_0 is the permeability in vacuum. The development of superconducting magnet technologies has enabled one to introduce magnetic fields as high as 10 T in academic laboratories. Under such strong magnetic fields, magnetization force acting on non-magnetic materials is not negligible.[1] For example, α-alumina, corundum, has a rhombohedral structure and has a small anisotropic diamagnetic susceptibility,[2,3] and a $\Delta_{\chi} = 4.19 \times 10^{-9}$ emu/cm^3 value has been reported.[3] The energies of anisotropy estimated for a spherical, single-crystalline α-alumina particle is 8.78×10^{-23} J in 1 T of magnetic field and 8.78×10^{-21} J in 10 T, supposing the particle size is 0.2 μm. On the other hand, the energy of thermal motion kT at 300 K is 4.14×10^{-21} J, where k is Boltzmann's constant. These values suggest that the energy of anisotropy in a strong magnetic field can be higher than the energy of thermal motion at room temperature ($\sim k$T).[4] The energy of anisotropy is the driving force for magnetic alignment.[5]

We have demonstrated that the textured microstructure of an alumina ceramic can be manipulated by a strong magnetic field applied to a stable alumina suspension during slip casting, followed by heating.[4,6] This method is suitable for making monoliths but not suited for highly textured composites. Electrophoretic deposition (EPD) is a colloidal process wherein ceramic bodies are directly shaped from a stable colloid suspension by a dc electric field.[7] It is a facile and precise technique to synthesize not only monoliths but also composites with complex geometries.[8] This paper reports the synthesis of

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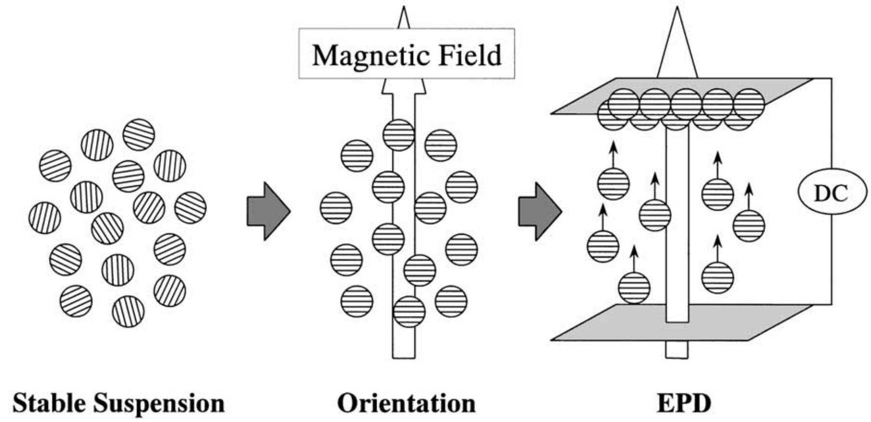

Fig. 1. Schematic diagram of the concept of the electrophoretic deposition in a strong magnetic field.

textured monoliths and laminate composites of α-alumina by applying both electric and magnetic fields to a stable alumina suspension.

2. Experimental

Single crystalline, granular α-alumina particles (Taimei Chem. Co., Ltd., TM-DAR, average particle size of 0.15 μm, high purity of $>99.99\%$) were dispersed at pH 4 in distilled water by ultrasound and stable suspension with a 10 vol.% solid content was formulated. Zeta-potential of the α-alumina particles in the suspension was $+40$ mV. The suspension was placed in a superconducting magnet (Japan Magnet Technology, JMTD-10T100) and then a strong magnetic field of 10 T was applied to the suspension to rotate the particles. A pair of electrodes, 20×20 mm^2 in area with 20 mm spacing, held on a phenol resin support were put in the suspension and then the electrical field was applied. The magnetic field was maintained on the suspension during the EPD at room temperature. The direction of the particle's flow was parallel to the magnetic field and reverse to the gravity so as not to be affected by the Lorentz force and sedimentation. A palladium sheet was used as the cathodic substrate to absorb hydrogen caused by the electrolysis of the solvent; therefore, bubble-free, dense monolithic deposits were produced.[9] A schematic illustration of the EPD in a strong magnetic field is shown in Fig. 1. A multi-layered composite of oriented and randomly oriented layers was synthesized by the alternate EPD of alumina suspensions which were placed in and out of a magnetic field. The EPD experimental setup is shown Fig. 2. The green compacts of monolithic and laminate deposits were dried at room temperature and densification was achieved by pressureless sintering in air at 1873 K for 2 h in air. X-ray diffraction (XRD) experiments and optical microscopic observation were carried out for the cross-sectional planes which are parallel and perpendicular to the substrate. Hereafter, those

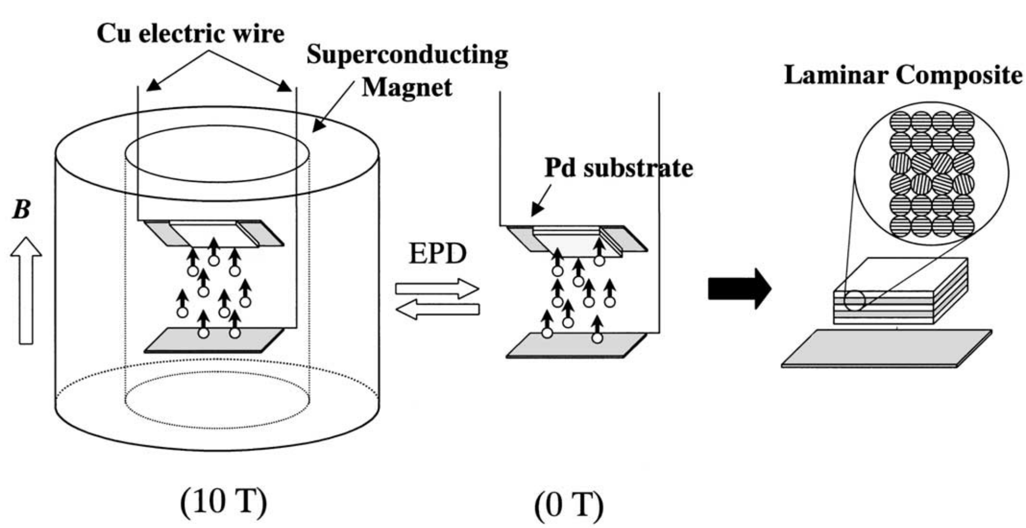

Fig. 2. Schematic diagram of the experimental setup of the alternate electrophoretic deposition in and out of the magnet.

planes are designated TOP and SIDE, respectively. Laminar structure of the multi-layered composite was observed for a thinly sliced sample by using a polarizing optical microscope.

3. Results and discussion

Fig. 3 shows the XRD patterns of the monolithic α-alumina deposited at 10 T using the stable suspension, followed by sintering at 1873 K for 2 h. Big difference is observed between the XRD patterns of the TOP and the SIDE. To characterize the XRD peaks, the interplanar angles ϕ_{hkl} between the planes (*hkl*) and the basal plane (*00l*) were calculated for the hexagonal unit cell of α-alumina ($a = 0.4758$ nm, $c = 1.2991$ nm).[10] The angle ϕ between the planes ($h_1k_1l_1$) and ($h_2k_2l_2$) for a hexagonal cell is calculated from the following equation.[11]

$$\cos\phi = \frac{h_1h_2 + k_1k_2 + \frac{1}{2}(h_1k_2 + h_2k_1) + \frac{3a^2}{4c^2}l_1l_2}{\sqrt{\left(h_1^2 + k_1^2 + h_1k_1 + \frac{3a^2}{4c^2}l_1^2\right)\left(h_2^2 + k_2^2 + h_2k_2 + \frac{3a^2}{4c^2}l_2^2\right)}} \quad (2)$$

The diffraction peak of the plane (*006*) ($\phi_{006} = 0°$) is observed only for the TOP, and the peaks of the planes (10$\underline{10}$) ($\phi_{10\underline{10}} = 17.5°$), (018) ($\phi_{018} = 21.51°$), (104) ($\phi_{104} = 38.25°$) and (116) ($\phi_{116} = 42.31°$) of the TOP are stronger than those of the SIDE. In contrast, the diffraction peaks of the planes (110) ($\phi_{110} = 90°$), (030) ($\phi_{030} = 90°$), (211) ($\phi_{211} = 83.16°$), (124) ($\phi_{124} = 64.38°$) and (113) ($\phi_{113} = 61.22°$) of the SIDE are stronger than those of the TOP. The XRD data clearly show the crystallite orientation of the α-alumina prepared in the strong magnetic field of 10 T. It is also confirmed that the *c*-axis is easy to align along the magnetic field.

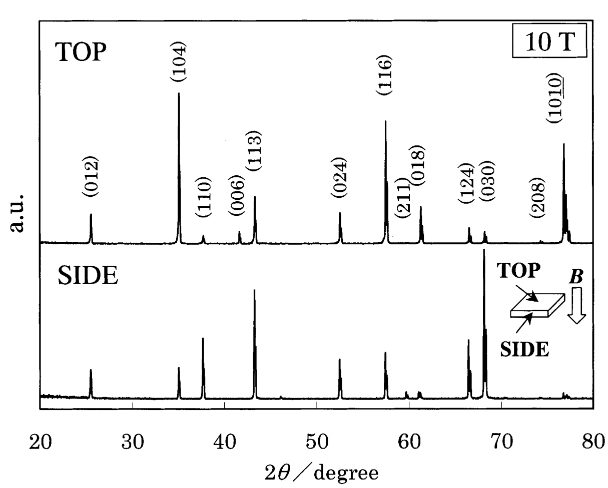

Fig. 3. X-ray diffraction patterns of the α-alumina prepared at 10 T.

Fig. 4 shows the XRD patterns of the monolithic α-alumina deposited at 0 T (external to the magnetic field) followed by sintering at 1873 K for 2 h. No difference is observed between the XRD patterns of the

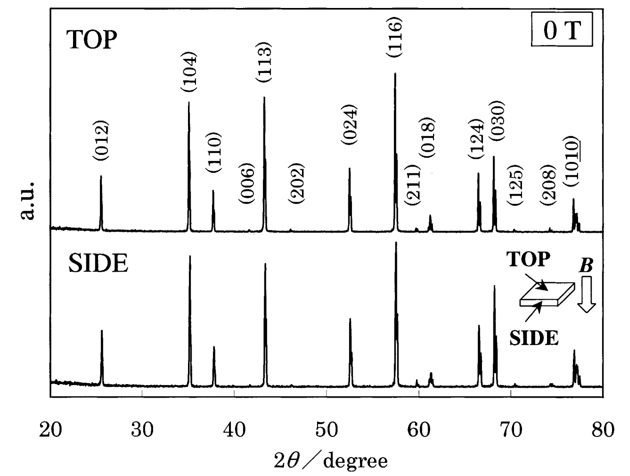

Fig. 4. X-ray diffraction patterns of the α-alumina prepared at 0 T (external to the magnetic field).

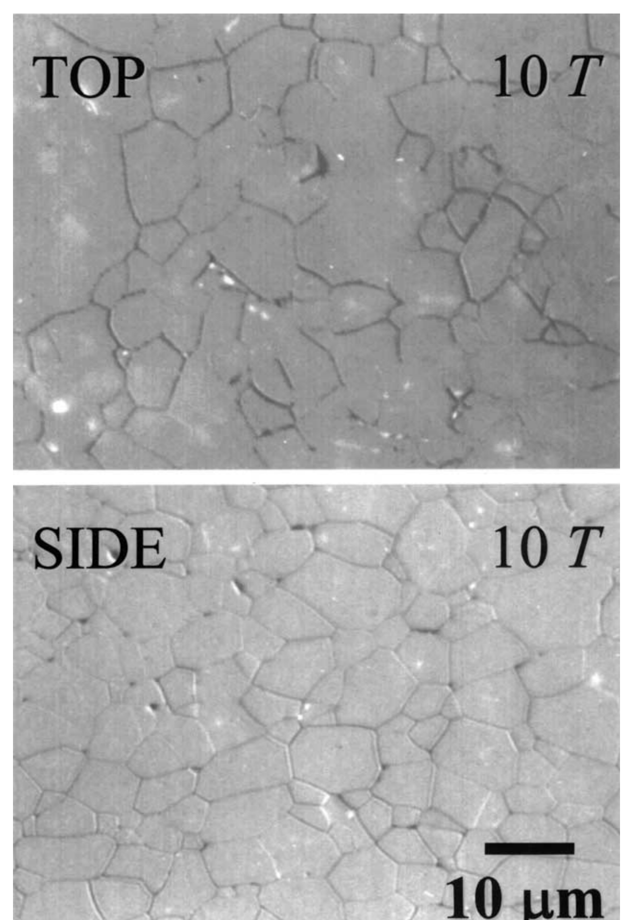

Fig. 5. Microstructure of the monolithic α-alumina prepared at 10 T.

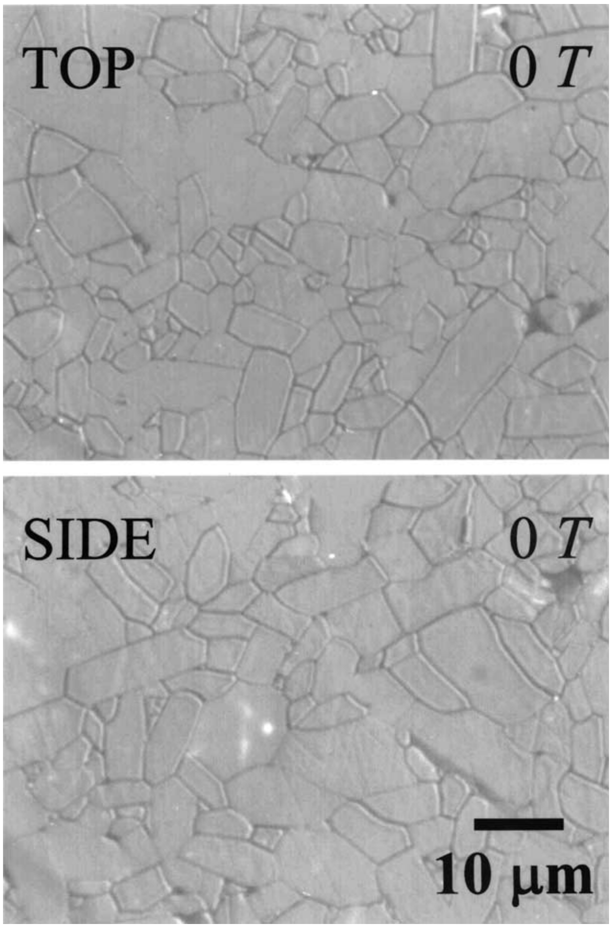

Fig. 6. Microstructure of the monolithic -alumina prepared at 0 T.

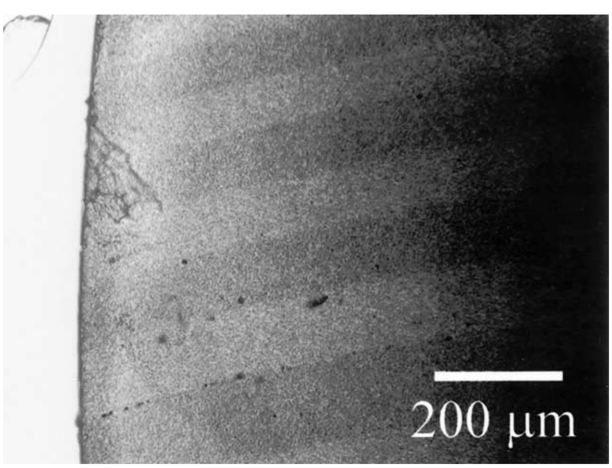

Fig. 7. Optical micrograph of the laminar alumina composite with oriented and randomly oriented layers.

TOP and the SIDE. These XRD patterns are consistent with the standard XRD data (ICDD card #10-173). It is obvious that the specimen prepared without a magnetic field has a randomly oriented poly-crystalline structure.

Fig. 5 shows the microstructure of the -alumina monoliths prepared at 10 T. Thermal etching of the polished surfaces was conducted at 1773 K for 2 h. Randomly developed granular grains are observed for the TOP and uneqiaxed grains developed in the perpendicular direction of the magnetic field are observed for the SIDE. It is noted that the difference in the sensitivity to thermal erosion between the *c*-plane and the *ab*-plane gives the blurred image of the grain boundaries of the TOP. However, without a magnetic field, randomly oriented grains are observed for both the TOP and the SIDE, and their microstructures are similar to each other as shown in Fig. 6.

Fig. 7 shows the optical micrograph of the laminar composite with oriented and randomly oriented layers. Dark and bright color layers are randomly oriented and oriented layers, respectively. Difference of the brightness is caused by the difference of the average crystallite orientation between the two layers. Alternate EPD in and out of a strong magnetic field enables to fabricate a laminar alumina composite composed from single element, oriented and randomly oriented layers.

4. Conclusions

Crystallite oriented α-alumina polycrystal was obtained by the EPD at 10 T using a stable α-alumina suspension. A multilayered composite of oriented and randomly oriented layers was synthesized by the alternate EPD of alumina suspensions which were placed in and out of a magnetic field. It was demonstrated that the EPD in a strong magnetic field is a promising ceramic processing technique for fabricating sophisticated ceramic composites.

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