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Laminated alumina/zirconia ceramic composites prepared by electrophoretic deposition

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Abstract

The electrophoretic deposition of alumina and zirconia powders from isopropanolic suspension in the presence of monochloroacetic acid was studied in the constant-current regime. The different levels of electric current during deposition from 250 μA to 48 mA were used. The green density of the deposit depends on the current density and then on the particle velocity during deposition, reaching values from 58% to 61% according to the electric current used. It was found that the lower the green density of the green deposit, the larger the pores. The low green density led to low final fired density and subsequently to the low Vickers hardness HV5 ranging from 2000 to 1650 depending on electric current used. Based on these findings microlaminates having various thickness ratios to achieve different residual stress levels were prepared consisting of alternating layers of alumina and zirconia.

Keywords: Electrophoretic deposition; Composites; Hardness; Al_2O_3; ZrO_2

1. Introduction

Electrophoretic deposition is an experimentally simple and cheap technique enabling the formation of deposits from particles of nanometric dimensions to particles of several micrometers in size.[1–3] The technique of electrophoretic deposition appears to be a promising technique in the preparation of one-component[4–6] and two- or more-component[7–9] bulk ceramics. The technique enabling effective preparation of thin surface layers[10–12] and layered composites consisting of layers of given thickness and density.[13–16] The laminates prepared with strongly bonded interfaces contained residual internal stresses originated from thermal expansion mismatch between layer. These materials embodied unique crack behaviour: crack propagated through layer interface is deflected from or towards the layer interface depending on residual stress orientation in the layers.[17–22]

The aim of this work was to describe the kinetics of electrophoretic deposition of alumina and zirconia powders and the influence of deposition parameters on the final microstructural and mechanical properties of deposits. Based on these findings layered alumina/zirconia composites were prepared.

2. Materials and methods

The alumina (type HP-DBM, Malakoff Ind., USA) and tetragonal-zirconia stabilized with 3 mol% of Y_2O_3 (type TZ-3YSE, Tosoh, Japan) powders were used for deposition having mean particle size 470 nm and 140 nm respectively. The dispersion medium used for suspensions preparation was isopropanol (p.a., Onex, Czech Republic). Monochloroacetic acid (99%, Aldrich, Germany) was added to the suspensions to aid stabilization and dispersion. The amount of water in the ceramic powders was reduced to the minimum via drying at 100 °C for 40 min. For the same reason, the isopropanol was distilled prior using. The suspensions used for electrophoretic deposition were prepared by mixing of 15 wt.% of alumina or zirconia powder with 12.75 wt.% of monochloroacetic acid and 72.25 wt.% of isopropanol.

Electrophoretic deposition was performed in a horizontal electrophoretic cell with two stainless steel electrodes. The electrode distance d was 26 mm and the effective electrode surface S (the surface actually immersed in the suspension) was 18.7 cm^2.

Electrophoretic deposition proceeded in the constant current mode at 250 μA, 500 μA, 5 mA, 20 mA, 25 mA and 48 mA. In order to prevent particles from settling the suspension was stirred repeatedly each 5 min during electrophoretic deposition. The moist deposit was first dried on the electrode for a

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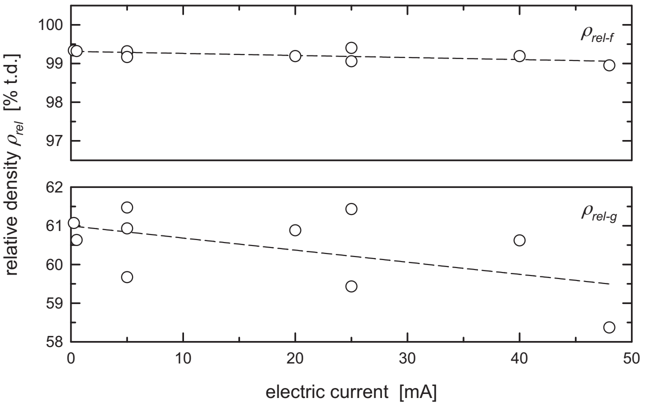

Fig. 1. Dependance of relative density of the alumina deposits after annealing (down) and after sintering (up) on electric current during electrophoretic deposition.

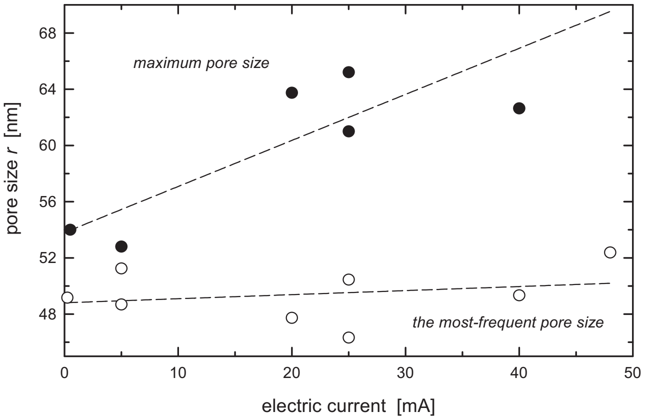

Fig. 2. Dependance of maximum and most-frequent pore size of alumina deposits on electric current during electrophoretic deposition.

period of 3 h at room temperature. Partially dried deposits were removed from the electrode and dried for another 12 h under the same conditions. Subsequently, the deposits were annealed (800 °C/1 h; heating rate + 2 °C/min) and sintered (1500 °C/2 h; heating rate + 2 °C/min) in air. After every step of the deposition the deposit weight was measured. The density of the deposit was determined using Archimedes' method (EN 623-2).

3. Results and discussion

As is evident from Fig. 1, the green densities and subsequent fired densities of the deposits depended on the electric current used for the electrophoretic deposition. The velocity of particles in the electric field during electrophoretic deposition is proportional to the electrophoretic mobility μ and electric field intensity E:

$$v = \mu \cdot E. \tag{1}$$

According to the Ohm's law, the voltage is proportional to the electric current and to the resistance of suspension R_{susp} and deposit R_{dep}:

$$v = \frac{\mu \cdot (R_{susp} + R_{dep}) \cdot I}{d}. \tag{2}$$

During electrophoretic deposition the electric current was changed in the range between 250 μA and 48 mA and then the velocity of particles deposited changed from 0.05 mm s^{-1} to 10 mm s^{-1}. The velocity of particles reaching electrodes affected the arrangement of the particles forming deposit. The deposits obtained by electrophoretic deposition under low currents (250 μA) contained small pores comparable to interparticle spaces in size (see Fig. 2). The layers deposited under high electric currents (48 mA) obtained large pores originated from non-ideal arrangement of particles in the deposit. As can be inferred from Fig. 1, the current density during deposition affected strongly the green density of the deposit, but just small effect of electric current was found on the final density.

The kinetics of the electrophoretic deposition have been described by Zhang et al.[23] who derived equation for deposited mass m as a function of initial mass of particles in the suspension m_0 and time t during electrophoretic deposition in the constant-current regime:

$$m = m_0 \cdot (1 - e^{-((\mu \cdot E)/d)\cdot t}), \tag{3}$$

where m_0 (kg) is the initial mass of particles in the suspension, d (m) is the distance between electrodes, E (V m^{-1}) is the intensity of electric field, μ (m^2 V^{-1}) is the electrophoretic mobility of particles, and t (s) is the time of deposition. The kinetics curves obtained during electrophoretic deposition of alumina in constant current regime ($I = 250$ μA, 500 μA and 5 mA) are given in Fig. 3. The dashed line on top representing initial mass of ceramic particles in the suspension $m_0 = 11.6$ g and thus limiting value of deposited mass. The full lines representing theoretical predictions for given electrical conditions, while white symbols representing experimental data. As is evident from Fig. 3 experimental kinetics data differs from theoretical prediction. No each particles reaching during deposition process electrode composes the final deposit. Zhang suggested to describe this by adding to the kinetics Eq. (3) efficiency factor f takes the value from 0 to 1. The dashed lines representing prediction of deposition

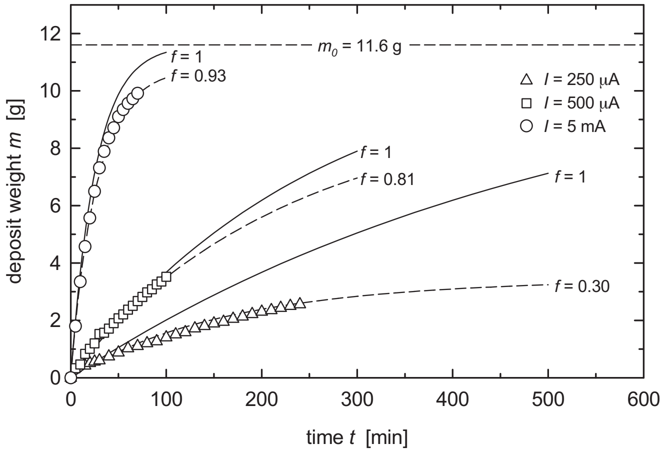

Fig. 3. Electrophoretic deposition kinetics of alumina suspensions under different electrical conditions.

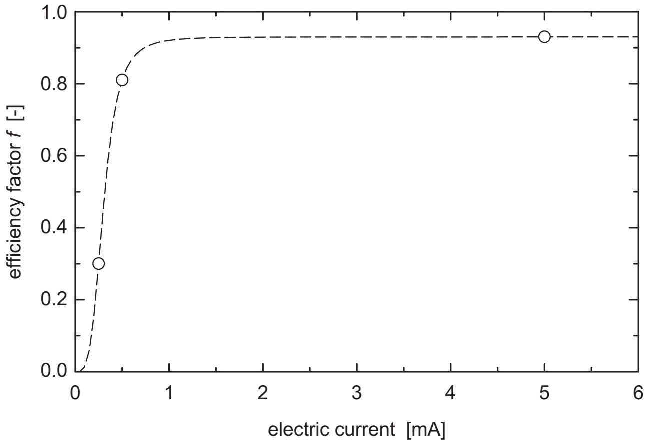

Fig. 4. Dependance of efficiency factor *f* of electrophoretic deposition of alumina deposits on electric current.

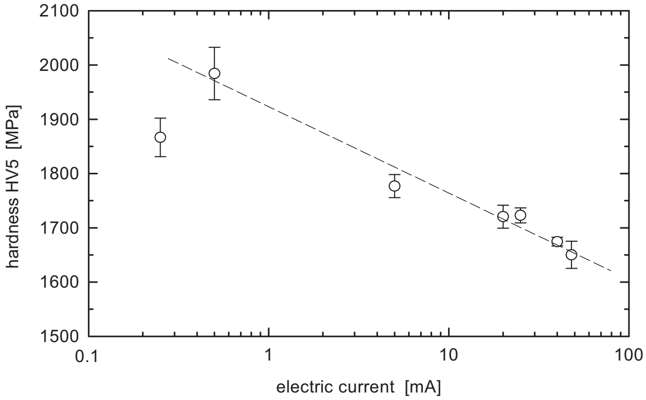

Fig. 5. Influence of electric field during electrophoretic deposition on final hardness of alumina deposits.

kinetics assuming experimental data and appropriate efficiency factor *f*. From the three points found in this work the estimation of the efficiency factor *f* dependance on electric current used could be done (see Fig. 4). The lower the electric current is the lower is the efficiency factor *f* and thus the part of particles forming deposit. As was shown in Fig. 2 the deposits obtained by electrophoretic deposition under low currents (250 μA) was probably formed by small particles and thus contained small pores and after sintering probably also smaller grains. It is assumed that during deposition under low currents just the smaller particles and/or agglomerates take part in the deposition process and the deposit containing low amount of small pores is obtained.[24]

Surprisingly the significant influence of electric current density on final mechanical properties was found by measuring of Vickers hardness of the deposits (see Fig. 5). The Vickers hardness of prepared ceramic layers embodied dependance on electric current during deposition. The hardness of the deposits grew with decreasing electric current. Hardness of the deposit obtained by deposition on lowest electric current 250 μA was affected by the small thickness of the deposit and subsequent cracking during indentation. The increasing of green density (see Fig. 1) and increasing of homogeneity (see Fig. 2) of deposits probably caused the differences in the grain size of the deposits which was not measured in this work. But it was described in the literature that two deposits of close final densities and different green densities can achieve during sintering different grain sizes.[25,26]

By using technique described above layered microlaminates consisting of alternating alumina and tetragonal zirconia layers of different thickness could be prepared (see Fig. 6). The interface of alumina/zirconia layers in the composite prepared did not contain any defects and the layers were properly sintered. The laminates prepared with strongly bonded interfaces contained residual internal stresses originated from thermal expansion mismatch between layers.[27] These materials embodied unique crack behaviour: crack propagated through layer interface is deflected from or towards the layer interface[7,14,28] depending on residual stress orientation in the layer.

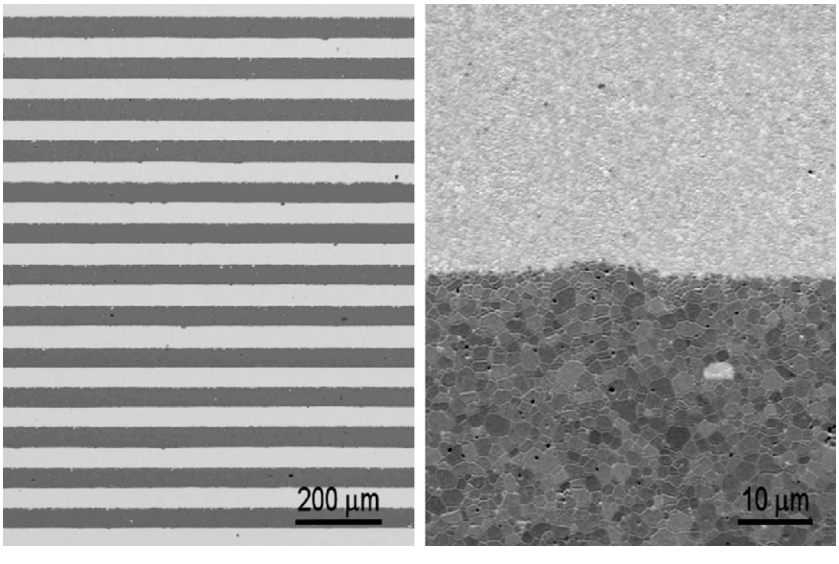

Fig. 6. Microphotographs of the layered alumina/zirconia microlaminate (dark-alumina; light-zirconia).

4. Conclusions

Electrophoretic deposition under different current densities of alumina and zirconia powders from isopropanolic suspension in the presence of monochloroacetic acid was studied. It was found that with decreasing electric field intensity increased the homogeneity of particle packing which led to the higher sintered density as well as hardness of the deposit. The kinetics of electrophoretic deposition was precisely described taking into account deposit drying. Using this technique, an alumina/zirconia laminate consisting of strongly bonded layers was prepared.

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