

Colloidal Processing of BaCeO₃-Based Electrolyte Films

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ABSTRACT

Preparation of high-quality electrolyte films on porous substrates is critical to the fabrication of high-performance solid-state ionic devices such as solid oxide fuel cells and chemical sensors. In this study, a colloidal process has been investigated for the preparation of BaCeO₃-based electrolyte films on both dense and porous substrates for electrochemical applications. The important processing variables affecting the microstructures of green films are identified and optimized to obtain uniform, crack-free green films of BaCe_{0.8}Gd_{0.2}O₃ with high packing density of the electrolyte particles. Further, dense ceramic films of BaCe_{0.8}Gd_{0.2}O₃-based electrolyte have been successfully fabricated on different substrates by careful process control. In addition, observations indicate that small amounts of additives can dramatically influence the densification behavior of barium cerate-based electrolyte films.

Introduction

Solid oxide fuel cells (SOFCs) and chemical sensors are among the solid-state ionic devices whose performance can be considerably enhanced by the use of thin-film electrolytes. In order to achieve high ionic conductivity, the electrolyte must be dense and uniform, with a well controlled crystal structure and stoichiometry. SOFCs based on yttria-stabilized zirconia (YSZ) are operated at high temperatures (800–1000°C) to insure sufficient ionic conductivity. High operating temperatures result in high operating costs as well as system degradation due to reactions at the interfaces between cell components.¹ These problems can be overcome by reducing the operating temperatures to intermediate temperatures (600–800°C). Solid electrolytes based on barium cerate have been reported to have conductivities similar to those of YSZ at temperatures about 200°C lower.^{2–4} Barium cerates doped with 10–20% of lanthanide series elements have been widely reported to have high ionic conductivity at intermediate temperatures and thus hold good promise as electrolyte materials for SOFCs to be operated at these temperatures. In particular, BaCe_{0.8}Gd_{0.2}O₃ (BCG) exhibits the highest ionic conductivity at temperatures below 800°C among the electrolytes stable in a fuel cell environment.² Thus, BaCe_{0.8}Gd_{0.2}O₃ is an attractive choice as an electrolyte material for high-performance, intermediate-temperature SOFCs, provided that it can be fabricated in a thin-film form.

For efficient operation of a solid-state ionic device at intermediate temperatures, the electrolyte must be used in a thin-film form to reduce resistive losses in the electrolyte.⁵ Further, the electrolyte membranes must be continuous and crack-free in order to prevent gas leakage, and must rest between two porous electrodes through which gases can transport freely to or away from the electrode-electrolyte interfaces, where electrochemical reactions occur. Thus, the fabrication of high-quality electrolyte films on porous electrodes assumes significant importance.

There are a number of techniques for depositing thin films of ceramics on porous and dense substrates, includ-

ing chemical vapor deposition, electrochemical vapor deposition, sol-gel processes, and various sputtering processes using ion beam, magnetron, electron beam, and so forth.¹ The drawbacks of physical vapor deposition and sputtering techniques include difficulties in obtaining good compositional homogeneity^{6,7} and high costs due to the requirement of high vacuum conditions.⁷ Sol-gel techniques, on the other hand, overcome these problems and offer many additional advantages. Films derived from a sol-gel process have good compositional homogeneity and high purity, and can be prepared over a wide range of compositions in a relatively simple manner.^{8–10} Another advantage of a sol-gel process is that it requires much less equipment and is thus relatively inexpensive as compared to the other techniques.¹¹ According to the precursors used or the gelation mechanism involved, sol-gel process is usually classified to alkoxide method and colloidal method. In a colloidal method, a sol is prepared from fine powders and a substrate is then coated with the sol solution to form a green film, which is subsequently heated to elevated temperatures to remove the organics and to densify the film.

The objective of this study is to fabricate films of BaCeO₃-based electrolytes on dense and porous substrates using a colloidal process. The processing parameters affecting the film microstructure are systematically investigated. The key processing variables are subsequently identified and their effect on the quality of films is studied in detail. The effect of a "sintering aid" on the densification behavior of the electrolyte films is also observed. The electrochemical properties of the films and the performance of the solid-state ionic devices based on these films will be discussed in a subsequent communication.²⁵

Experimental

Preparation of BaCe_{0.8}Gd_{0.2}O₃ electrolyte powders.—Powders of BaCe_{0.8}Gd_{0.2}O₃ (BCG) were prepared using the Pechini process^{12–14} and conventional ceramic processing. In the conventional ceramic processing, barium carbonate, cerium oxide, and gadolinium oxide (all from Alfa AESAR) were mixed in stoichiometric amounts and ball-milled (in alcohol with zirconia balls as the grinding media) for 24 h to ensure good mixing of the powders and to reduce the particle size to 1–4 μm. The mixtures were then calcined at 1350°C for 8 h¹⁵ and x-ray diffraction

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(XRD) was used to confirm the formation of $\text{BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ phase in the powder. In case of incomplete reaction, calcination at 1350°C and ballmilling were repeated until pure perovskite phase (as verified by XRD) was obtained. The reacted powders were then ballmilled for another 24 h to break up the particle aggregates and to reduce the particle size to 1–4 μm , as determined using scanning electron microscopy (SEM) and Microtrac particle size analyzer. In the Pechini process,^{12–14} stoichiometric amounts of barium carbonate, cerium nitrate, and gadolinium nitrate (all from Alfa AESAR) were dissolved in citric acid (Aldrich) and ethylene glycol (Fisher) to form a sol having a stable metal cation dispersion. The sol was then dried to form a gel, which was further heated to elevated temperatures to remove the organic constituents and to produce fine particles of desired barium cerates. It was found that heat-treatment at 1000°C for 4 h was necessary to ensure pure perovskite phase (as determined using XRD); powders from heat-treatment at temperatures below 1000°C often contained BaCO_3 phase. The mean size of aggregates (as measured using microtrac particle size analyzer and SEM) in the powders derived from the Pechini process ranged from 0.1 to 0.3 μm .

Preparation of colloidal sols.—Colloidal sols were prepared from each of the two types of powders by dispersing the powders in deionized water. The range of solids loading was varied from 0.1 to 1.0 gram powder per ml water in increments of 0.1 gram powder per ml of water. Dispersing agents were subsequently added and the sol was ultrasonicated for 10–15 min to assist the dispersion of electrolyte particles. Three types of dispersing agents were used in this study: 1-1-1-trichloroethane (type A), a mixture of 50 volume percent (v/o) 1-1-1-trichloroethane and 50 v/o isopropanol (type B), and sodium polyacrylate (type C).^a Type A and B dispersants were obtained from Fisher Scientific Company and type C dispersant was obtained from R.T. Vanderbilt Company, Inc. At each solids loading, the amounts of the dispersants were varied until a visually homogeneous and stable sol was obtained. A sol was considered unstable if the particle sedimentation took place in less than 2 h. For the type A and B dispersants, the volume fractions of dispersant in water were varied from 0.05 to 0.1, while for the type C dispersant the volume fractions of the dispersant in water ranged from 0.02 to 0.07. The pH of the sol was monitored using an Oaktron electronic pH meter.

Preparation of substrates.—The choice of a substrate is determined primarily by the application of the electrolyte film. However, the wettability of the colloidal sol with the substrate and the thermal expansion mismatch between the film and the substrate must be acceptable. In this study, pellets of $\text{BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ were used as dense substrates in order to minimize problems due to chemical reactions and thermal mismatch between the film and substrates. Disks of $\alpha\text{-Al}_2\text{O}_3$ (from Vesuvius McDanel Company) were also used as dense substrates since the thermal expansion coefficient of $\alpha\text{-Al}_2\text{O}_3$ is similar to that of the electrolyte. Composite $\text{NiO-BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ pellets were used as conductive, porous substrates in order to facilitate the electrical characterization of the electrolyte films. The powders of $\text{BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ and $\text{NiO-BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$, prepared by the conventional ceramic processing as described earlier, were uniaxially pressed to pellets in a die at a mechanical pressure of 35,000 psi. The $\text{BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ pellets were sintered at 1500°C for 8 h and the $\text{NiO-BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ pellets were sintered at 1350°C for 3 h to achieve densities greater than 95% of the theoretical values. The use of a dense substrate eliminates the problem of sol infiltration into the substrate that is usually encountered with porous substrates. Since the surface microstructure and the cleanliness of a sub-

strate are important factors affecting the wettability of a sol, all sintered substrates were ground to 1200 grit, cleaned ultrasonically in methanol, and then dried in an oven at 65°C for 2 h before coating.

Dip coating, drying, and firing.—Films of colloidal sols were deposited on the substrates by a dip-coating method. Substrates were typically submerged in a sol for about 5 s before they were pulled out at a rate of approximately 20 cm/min. The coated substrates were dried overnight to evaporate the solvent. Typically, the thickness of a deposited layer varied from 3 to 7 μm , depending on the solids loading. Two types of drying conditions were studied: one in an oven at 65°C and the other in the ambient atmosphere at about 27°C. After drying, the resulting green films were fired at temperatures ranging from 1100 to 1500°C in 50°C intervals, and the firing time at each temperature was varied from 1 to 4 h in 1 h intervals. Firing rates were varied from 3 to 10°C/min. Unless stated otherwise, all heat-treatments were carried out in a tube furnace in air and sintered samples were cooled down at a rate of 10°C/min.

Characterization of films.—Thermogravimetric analysis (TGA) was used to characterize the thermal evolution of the films. X-ray diffraction was used to analyze the phases present in the films. The microstructures of green and fired films were revealed using an SEM equipped with EDS. The detailed characteristics of the microstructures were further quantified using stereological techniques.¹⁷ The mean intercept method was used to estimate the particle or grain sizes and pore sizes in green and fired films and the point count method was used to estimate the porosity of films and substrates. Typically, about 10 measurements were taken for each data point to improve the confidence of the data.

Results and Discussion

Characteristics of the colloidal sols.—The stabilities of the sols were studied as a function of particle size and the type of dispersant. When the powders derived from the Pechini process were dispersed using dispersant A or B, the most stable sols had pH of about 6.5 and were stable for about 24 h. When the same powders (from the Pechini process) were dispersed using dispersant C, the most stable sols had pH of about 12.5 and were stable for 7–10 days. The dispersion mechanism of dispersant A and B may be electrostatic whereas the dispersion mechanism of dispersant C is perhaps a combination of steric and electrostatic (*i.e.*, electrosteric)¹⁸ because of the polymeric nature of dispersant C and the polar nature of the solvent (water). Also observed were the difficulties encountered in preparing stable sols using powders with larger particles (1–4 μm) obtained from conventional ceramic processing. Sedimentation usually took place in 5–6 h when type A or B dispersant was used and in 24 h when dispersant C was used.

To form a stable dispersion, the minimum volume fractions of dispersant A or B in water were 0.07 for conventional powders and 0.06 for the Pechini powders. With dispersant C, the minimum volume fractions were 0.05 for conventional powders and 0.04 for powders from the Pechini process. The sol viscosities ranged from 5 to 7 centipoise at 27°C. The optimum solids loading for an acceptable wettability was found to be 0.4 to 0.6 gram powder per ml water for the three types of dispersants on both $\text{BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ and $\text{NiO-BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ substrates. A solids loading of 0.5 gram powder per ml water was thus chosen for further studies.

Microstructures of green films.—In order to achieve high density of sintered films, it is necessary to start with uniform, crack-free green films with high packing density of the electrolyte particles. Accordingly, the effect of various processing parameters on the microstructures of green films was studied, including particle size and distribution, nature and amount of dispersant, types of substrate, and the drying conditions.

^a The use of sodium polyacrylate will introduce some Na to the electrolyte. XRD studies suggested that all Na doped into the electrolyte and electrical measurements indicated that the doped Na has no observable detrimental effect on the electrical properties of the electrolyte.¹⁶

Shown in Fig. 1a is the microstructure of a green film obtained from a sol using type A or B dispersant and powders from conventional processing. The sol was deposited on a $\text{BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ substrate and dried in the ambient atmosphere at about 27°C. The microstructure shows extremely loose packing (nearly 50% porosity) of particles with large amount of needle-like organics in the green state. These needle shaped organics may have formed due to the recrystallization of the dispersants A and B during drying. The use of the same dispersant (type A or B) with sol-gel derived powders led to particle agglomerates in the green film as shown in Fig. 1b. The use of dispersant C with conventional powders along with agglomerate formation in the green film. The use of dispersant C for the sol-gel derived powders, however, gave stable dispersion and excellent packing (about 6% porosity) in the green state, as shown in Fig. 1c. The particles are densely packed and the microstructure of the film is free of cracks and agglomerates. Although the

green film microstructure was very sensitive to the particle size and the nature of dispersant, it was relatively insensitive to the different types of substrates and the drying conditions studied. The optimum volume fraction of dispersant C in water was 0.04 for crack-free green films. An increase in volume fraction of dispersant C to 0.05 or greater resulted in cracks in the green state, as shown in Fig. 1d, due to overdeflocculation of particles in the colloidal sol. Overdeflocculation may occur when excessive amount of polymer dispersant absorbs together a larger number of individually dispersed particles to form clusters.¹⁹ These clusters of particles can lead to cracks in the green films as seen in Fig. 1d. Thus, the amount of dispersant C should be no more than just enough to form a thin polymer coating on each individual particle.

Thermogravimetric analysis.—Green films with optimized microstructure were then fired at elevated temperatures to remove the organics and to form dense ceramic

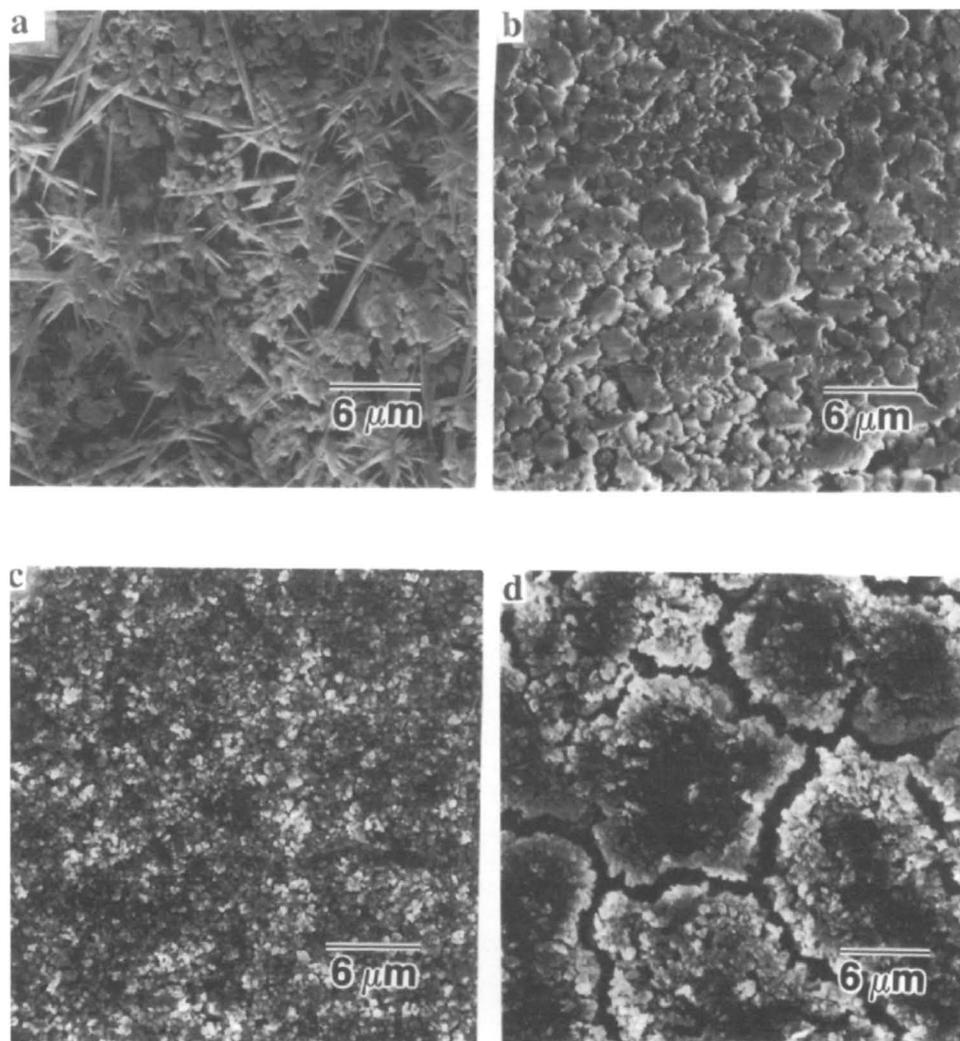


Fig. 1. Surface views of green films of electrolyte on dense $\text{BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ substrates: (a) type A dispersant and powders prepared using conventional ceramic processing, (b) type B dispersant and powders prepared using the Pechini process, (c) type C dispersant and powders from the Pechini process, and (d) excessive type C dispersant and powders from the Pechini process.

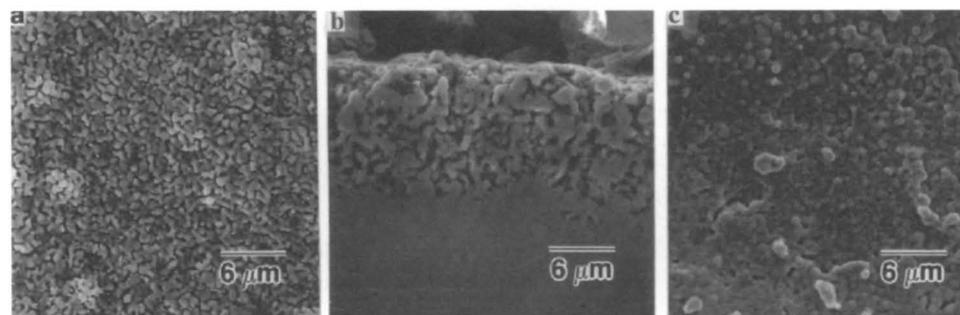


Fig. 2. (a) Surface view and (b) cross-sectional view of an electrolyte film (on a dense $\text{BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ substrate) after sintering at 1450°C for 2 h. (c) Surface view of an electrolyte film after sintering at 1450°C for 2 h when a mechanical pressure of 30,000 psi was applied in the green state.

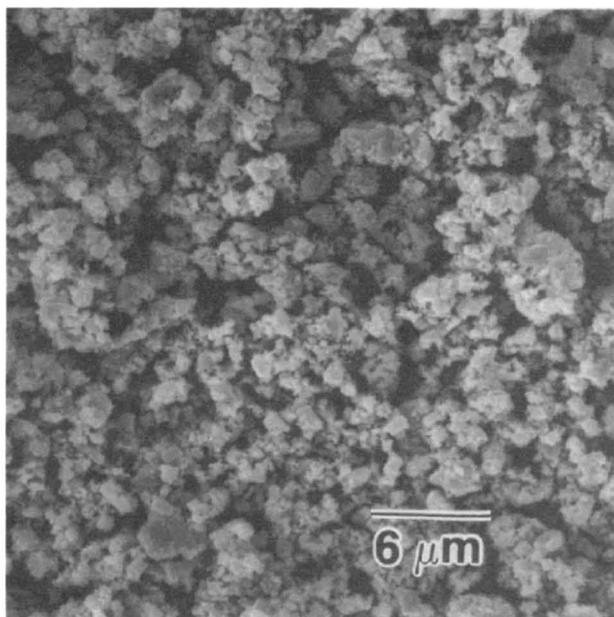


Fig. 3. Surface view of an electrolyte film (on a $\text{BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ substrate) fired at 1100°C for 2 h.

films. TGA studies of a green film containing 24 v/o (9.3 weight percent (w/o)) dispersant C indicated that there is a continuous weight loss from about 125 to about 450°C . The weight loss above 450°C was negligible. The estimated shrinkage due to the organics burn-off is about 24%. TGA studies of other green films indicated that most of the organics were removed by 450°C . Accordingly, the use of slow firing rates below 450°C is necessary to eliminate delamination or spalling of films from substrates. Typically, the heating rate was kept at $5^\circ\text{C}/\text{min}$ from room temperature to 450°C and at $7^\circ\text{C}/\text{min}$ above 450°C .

Microstructures of sintered films on dense $\text{BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ pellets.—Shown in Fig. 2a is the microstructure of a film (with 24 v/o dispersant C) derived from an optimized colloidal sol with powders from the Pechini process. The film was fired at 1450°C for 2 h and it appears to be uniform with an average grain size of $0.75\ \mu\text{m}$. It is, however, quite porous as seen from the cross-sectional view in Fig. 2b. The film seemed to adhere well to the substrate. The porosity of the film, as determined using the point counting method, was about 30% and the pore size ranged from 0.1 to $0.5\ \mu\text{m}$. An increase in the firing temperature from 1450 to 1500°C and in the firing time from 2 to 5 h resulted in an increase in grain size, but there was no significant improvement in densification. The porosity is due primarily to the inefficient packing of particles and large volume fraction of organics (dispersant) in the green

films. The reduction in the amount of organics, however, was limited by the stability of the sols; further lowering the amount of dispersant made the sols unstable.

In order to improve particle packing in the green state, a green film on a dense BCG substrate was subjected to a uniaxial pressure of about 30,000 psi (using a die and a hydraulic press). After firing at 1450°C for 2 h, the pressed films (Fig. 2c) showed some improvement in density (from 70 to 85%) over the unpressed films (Fig. 2a). In practice, however, subjecting a green film to a mechanical pressure of 30,000 psi may be difficult because the electrolyte films are usually supported by a porous electrode structure. A porous electrode may not have sufficient mechanical strength to withstand such pressure. It is possible, however, to use osmotic pressure to improve the packing density²⁰ of particles in a green film deposited on a porous substrate.

Electrolyte films derived from multiple coatings.—Another approach taken to improve film density was to apply a second layer of sol film on the top of a fired film which was deliberately made porous. The rationale behind this approach was that the second layer would fill in the pores left in the first layer. Also, the first porous layer may act as a good substrate by effectively absorbing the solvent through the pores which improves the particle-particle contact and hence the adhesion and packing of particles. Clearly, it is necessary to ensure that the particles are smaller than the pores. Since the pore sizes in the films fired at 1450°C were between 0.1 and $0.5\ \mu\text{m}$ (Fig. 2a), the application of a second colloidal layer (with particles sizes of 0.1 to $0.3\ \mu\text{m}$) on the top of this fired film would not be effective in filling in the pores in the first layer. In order to increase the porosity and the sizes of the pores in the first layer, the firing temperature of the layer was lowered. Shown in Fig. 3 is the surface view of a film (on a dense BCG substrate) after firing at 1100°C for 2 h. Stereological analysis of this micrograph indicated that most pores were between 1 and $3\ \mu\text{m}$ in size, with some pores up to $6\ \mu\text{m}$. The particles (0.1 to $0.3\ \mu\text{m}$) in a sol film deposited on the top of this porous layer will have high probability of falling into the pores in the first layer. Once inside the pores, the particles may also promote densification by acting as a "bridge" between the particles during sintering. Shown in Fig. 4a and b are the surface and cross-sectional views of a dense (greater than 97% of theoretical density), uniform, and crack-free film obtained by firing a first layer at 1100°C and then a second layer at 1450°C . The thickness of the resulting film is about 7 to $8\ \mu\text{m}$ and the grain size is about 1 - $1.5\ \mu\text{m}$. The film seems to adhere well to the substrate. As shown in Fig. 5, the x-ray diffraction pattern of the electrolyte film (on an $\alpha\text{-Al}_2\text{O}_3$ substrate) matched well with that obtained from a $\text{BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ pellet,¹⁵ confirming the formation of perovskite phase in the film. The result indicates that the pores in the first layer fired at 1100°C were filled by depositing a second layer of colloidal coating on the top of it. Thus, multiple coatings can improve the density of derived films.

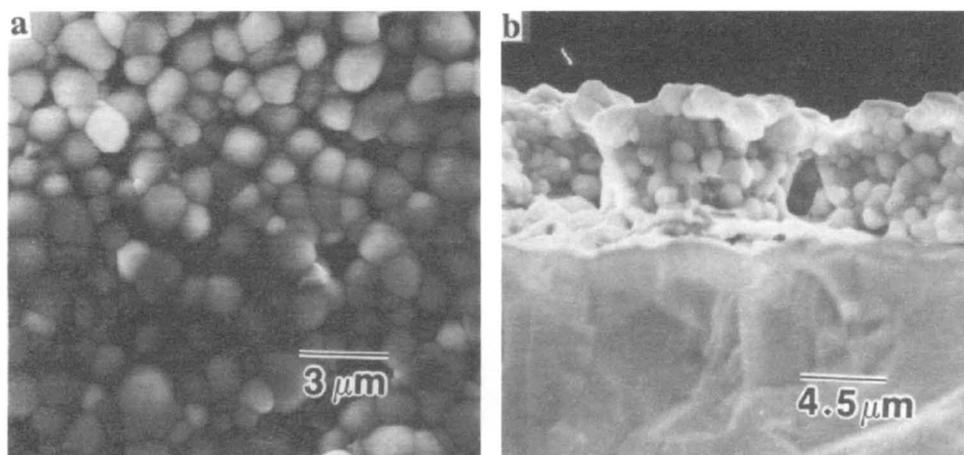


Fig. 4. (a) Surface view and (b) cross-sectional view of an electrolyte film (on the top of a dense $\text{BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ substrate) prepared by the multiple coating technique. The film was sintered at 1450°C for 3 h.

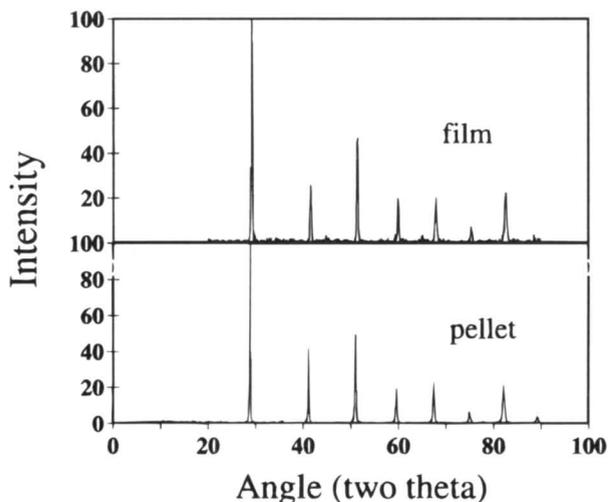


Fig. 5. X-ray diffraction patterns of a $\text{BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ pellet and an electrolyte film (derived from colloidal processing) on the top of an alumina substrate.

Electrolyte films on $\text{NiO-BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ composite electrodes.—In order to construct a solid-state ionic device based on an electrolyte film, such as an SOFC, the substrate must be electrically conductive and porous. Thus, we then focused on fabricating dense electrolyte films on electronically conductive and porous substrates. It is well

known that NiO-YSZ cermets have been widely used as anodes in YSZ-based fuel cells.^{1,21,22} Accordingly, NiO- $\text{BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ composite would be a logical choice as anodes for BCG based fuel cells. In this case, an electrolyte film could first be deposited on a dense NiO- $\text{BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ substrate, and the NiO could then be reduced to Ni metal to create porosity in the substrate. Figure 6a shows a surface view of a dense (greater than 97% of theoretical density) NiO- $\text{BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ pellet. According to EDS analysis, the darker phase in the microstructure is NiO and the lighter one is $\text{BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$. The powder composition was chosen such that after reduction of NiO to Ni metal, the volume fraction of Ni would be 40% of the composite. This volume fraction of Ni in the substrate would ensure that the substrate is electronically conductive.²³ Dense films were then deposited on the top of this substrate using the multiple coating approach. Figure 6b and c show the cross-sectional and surface view of a film (sintered at 1500°C for 3 h) on the top of a NiO- $\text{BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ substrate. The micrographs indicate that the film is dense (greater than 97% of the theoretical density) and the grain size in the film is large (10–30 μm). An EDS analysis of the film indicated the presence of Ni in the film. It is believed that Ni diffused from the substrate into the film and influenced the densification of the film, as evidenced from the observed high density and large grain size.

In our next processing step, the samples were exposed to hydrogen (at 750°C) to reduce NiO to Ni in order to create some porosity in the substrate. Figure 6d shows a cross-sectional view of the sample after reduction in hydrogen. The microstructure of the film was unaffected while pores were formed in the substrate (about 15% porosity) due to

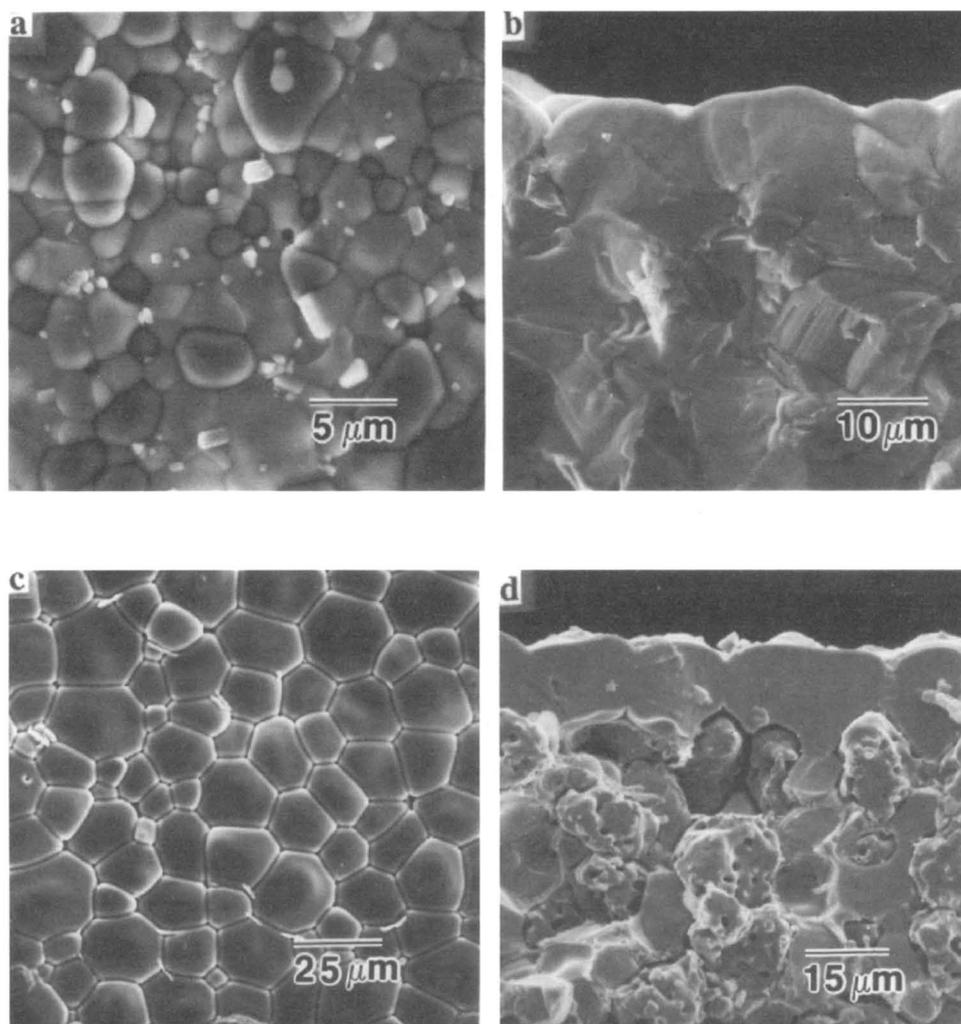


Fig. 6. (a) A surface view of an NiO- $\text{BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ substrate fired at 1350°C for 3 h, (b) cross-sectional view and (c) surface view of an electrolyte film (sintered at 1500°C for 3 h) on the top of an NiO- $\text{BaCe}_{0.8}\text{Gd}_{0.2}\text{O}_3$ substrate, and (d) cross-sectional view of the same sample after reduction in hydrogen at 750°C for 2 h.

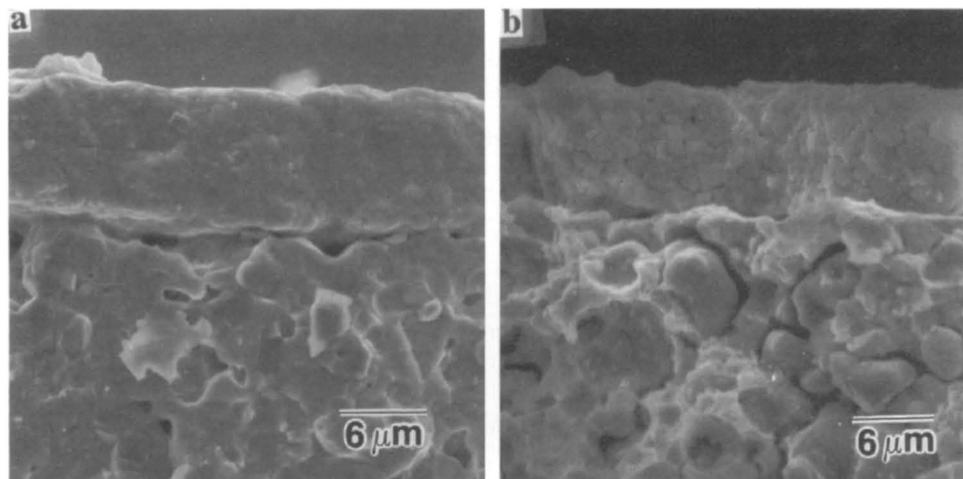


Fig. 7. A cross-sectional view of an electrolyte film (sintered at 1200°C for 2 h) on the top of an NiO-BaCe_{0.8}Gd_{0.2}O₃ substrate: (a) before and (b) after reduction in hydrogen at 750°C for 2 h. The substrate was pre-fired at 1000°C for 3 h.

the reduction of NiO. The pre-firing temperature of the NiO-BaCe_{0.8}Gd_{0.2}O₃ substrates was then lowered from 1350 to 1000°C to introduce additional porosity (about 10%) in the substrates.

To verify the role of Ni as a "sintering aid," the sintering temperatures and time of the film were systematically lowered from 1500°C to determine the lowest temperature at which dense films could be formed. It was found that a minimum temperature of 1200°C and a sintering time of 2 h was necessary to form an electrolyte film with density greater than 97%. Figure 7a and b show the cross sections of a film (sintered at 1200°C for 2 h) on the top of a NiO-BCG substrate before and after reduction in hydrogen. The film is dense (greater than 97% of the theoretical density) and the grain size is much smaller (1-3 μm) in comparison to the films fired at higher temperatures. It is thus clear that Ni has a dramatic effect on the densification behavior of the electrolyte films. Films on a BaCe_{0.8}Gd_{0.2}O₃ substrate could not be densified at temperatures below 1450°C while films on a NiO-BaCe_{0.8}Gd_{0.2}O₃ substrate were readily densified at temperatures as low as 1200°C. The only difference between the two substrates is the presence of Ni in the latter substrate. Apparently, the diffusion of Ni into the film somehow accelerated the densification of the films and lowered the sintering temperatures from 1450 to 1200°C. This observation has important implication to tailoring the sintering temperature of the electrolyte films.

Although the dramatic influence of Ni on the sintering behavior of barium cerate-based films was observed, the detailed mechanism of "sintering aid" effect is still unknown. Other transition metal oxides,²⁴ such as the oxides of Cu and Co, were also found to have similar effect on sintering behavior of BaCeO₃-based electrolytes. The mechanism of the observed sintering aid effect and the effect of these additives on the electrical properties will be discussed in a subsequent communication.²⁵

Conclusions

Particle size of BaCe_{0.8}Gd_{0.2}O₃ powders as well as the nature and the amount of dispersant in the colloidal sol are identified as the main processing variables affecting the green film microstructure in the colloidal processing. Crack-free, dense, and uniform green films have been obtained by optimizing the processing variables. Dense ceramic films of BaCe_{0.8}Gd_{0.2}O₃-based electrolyte have been successfully fabricated on both dense and porous substrates using a "multiple coating" approach through careful control of processing parameters. Results indicate that NiO acts as a "sintering aid" and dramatically improves the densification behavior of the BaCe_{0.8}Gd_{0.2}O₃ films. Accordingly, dense films of BaCe_{0.8}Gd_{0.2}O₃-based electrolyte have been successfully prepared on porous Ni-BaCe_{0.8}Gd_{0.2}O₃ substrates at temperatures as low as 1200°C.

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