



# Densification behavior of Spark Plasma Sintered $\text{La}_2\text{O}_3$ –YSZ ceramic composites

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## Abstract

The present paper describes the fabrication and densification behavior of 8 mol% Ytria-stabilized Zirconia (YSZ)-based composites with the addition of 11.6, 21.6 and 30.5 wt%  $\text{La}_2\text{O}_3$  fabricated by the Spark Plasma Sintering (SPS) process. Addition of  $\text{La}_2\text{O}_3$  to YSZ had a significant grain refining effect on the YSZ ceramic. In all of the studied composites, the desirable cubic structure of the YSZ matrix was retained and 94–96% densification was achieved. The matrix of the sintered composites evolved a solid solution due to interdiffusion of La, Zr and Y, as well as formation of pyrochlore lanthanate  $\text{La}_{1.6}\text{Y}_{0.4}\text{Zr}_2\text{O}_7$  phase formed as the reaction product between YSZ and  $\text{La}_2\text{O}_3$  during sintering.

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## 1. Introduction

Zirconia ( $\text{ZrO}_2$ ) is an attractive ceramic for engineering applications due to its very good high temperature stability, chemical inertness, low thermal conductivity and high fracture toughness [1–5]. At room temperature,  $\text{ZrO}_2$  has a monoclinic structure, which undergoes a tetragonal phase transformation at 1170 °C followed by a cubic phase transformation at 2370 °C [6]. The densities of the monoclinic, tetragonal and cubic phases are 5.65 g/cm<sup>3</sup>, 6.10 g/cm<sup>3</sup> and 6.27 g/cm<sup>3</sup> [7], respectively. Hence, the cubic to tetragonal and subsequently tetragonal to monoclinic phase transformations upon cooling from high temperature are often associated with volume expansion leading to large compressive residual stresses. This contributes to cracking of the as-sintered zirconia ceramic [6]. Suitable doping of  $\text{ZrO}_2$  with divalent, trivalent or tetravalent ions, such as  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Al}^{3+}$ ,  $\text{Y}^{3+}$ ,  $\text{Ce}^{4+}$ , enables stabilization of the cubic phase [8–13] and prevents undesirable cracking of the consolidated product. Due to the relative ease

of fabrication, yttria-stabilized zirconia (YSZ) ceramics are now gaining technological importance.

Literatures [7,13–18] suggest that the addition of lanthanum dioxide ( $\text{La}_2\text{O}_3$ ) to YSZ (up to 56 wt%) improves the thermal insulating properties of the composite, as well as promotes a strong grain refinement of the microstructure leading to enhanced mechanical properties [14]. Most available literatures [13–15,19] on the fabrication of  $\text{La}_2\text{O}_3$ –YSZ composites are limited to conventional sintering (CS) or preparation of coatings [7,16–18,20,21] using air spray pyrolysis and transfer arc plasma techniques. Some studies [7,15] have reported undesirable volume changes due to phase transformations upon cooling of the  $\text{La}_2\text{O}_3$ –YSZ composite. Challenges related to cracking, low density, as well as reaction of the composites with moisture in ambient air have been observed. Also, full stabilization of the  $\text{La}_2\text{O}_3$ –YSZ composites at low temperatures is highly challenging due to the large ionic radius of trivalent  $\text{La}^{3+}$  (103 pm) in comparison to that of  $\text{Zr}^{4+}$  (72 pm) [8]. Discrepancies in the literature about the microstructure evolution in the ceramic further hinder prediction of conclusive effects of lanthana addition to zirconia [7,13].

Recent studies indicate that zirconia-based composites prepared via Spark Plasma Sintering (SPS) can reach theoretical

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densities > 99% and ultra-fine grain size can be retained after SPS due to much lower sintering temperatures than typically used in CS [14,15,19,22–24]. Further, SPS offers rapid heating and densification, which leads to shorter sintering times in the range of 3–10 min [25–29], as well as the ability to sinter dissimilar materials [25,27,29] and possibly retain nano-structures [23,25]. However, there is a paucity of literature on SPS of lanthana-containing yttria-stabilized zirconia composites.

In the present work,  $\text{La}_2\text{O}_3$ –YSZ composites with the addition of 11.6, 21.6 and 30.5 wt%  $\text{La}_2\text{O}_3$  were prepared by SPS process and their densification behavior was studied in detail. Microstructure and phase evolution during sintering were examined and related to the  $\text{La}_2\text{O}_3$  doping level.

## 2. Experimental procedure

Precursor powders of 8 mol% (~13 wt%) yttria-stabilized zirconia (particle size 0.5  $\mu\text{m}$ ) and lanthanum dioxide (particle size 50–80 nm) with purity of 99.9% were obtained from Inframat Advanced Materials (CT, USA). The chemical composition of the precursor powders is provided in Table 1. Slurry blends of YSZ powder with the addition of 11.6, 21.6 and 30.5 wt%  $\text{La}_2\text{O}_3$ , respectively, were prepared in a beaker filled with alcohol. The slurries were dried for 24 h at room temperature in a desiccator. After drying, the powders were ground for 20 min using a pestle.

Blended powders were sintered using a Thermal Technologies Spark Plasma Sintering machine (Model 10-3) at Quad City Manufacturing Labs, USA. The powders were placed in a 19 mm diameter graphite die with a graphite foil lining. Sintering of all composite powders was carried out at a temperature of 1500 °C with uniaxial pressure of 50 MPa. The pressure was applied at the beginning of the heating cycle. Powders were heated to the sintering temperature at a rate of 100 K/min. After holding the sample for 10 min at the sintering temperature, the ram pressure was released and the composites were allowed to cool in the furnace. Upon ejection from the sintering die, the densities of the sintered composites were measured by the Archimedes' principle.

A Tescan MIRA3 XMU scanning electron microscope with Oxford X-max EDS detector were used for microstructural observation and chemical analysis. To reveal the grain width, subgrains and various phases within the composites, the samples were polished and thermally etched. Manual polishing of the samples was done using SiC and emery papers (grits 3/0 and 4/0) and a diamond paste (9  $\mu\text{m}$ , 6  $\mu\text{m}$  and 1  $\mu\text{m}$ ). Thermal etching (1 h) was carried out in air at 1450 °C. The grain size was subsequently measured via image analysis software

(Buehler OmniMet) using the linear intercept method. X-ray diffraction (XRD) of the powders and as-sintered composites was carried out with the X'Pert Pro (PANalytical) X-Ray diffractometer with Bragg–Brentano set-up, equipped with X'Celeratrim in a  $2\theta$  range of 20–90° using 0.02° step size and 20 s time per step. Cu- $K_\alpha$  radiation with  $\lambda = 1.541 \text{ \AA}$  was used. Precision lattice parameter of the cubic phase was calculated using the Nelson-Riley technique [30].

## 3. Results and discussion

Fig. 1 shows the XRD patterns of the YSZ and  $\text{La}_2\text{O}_3$  precursor powders, SPSed monolithic YSZ and SPSed  $\text{La}_2\text{O}_3$ –YSZ composites. The patterns in Fig. 1 confirmed that the original YSZ precursor powder had a cubic structure. Upon sintering of the monolithic YSZ and  $\text{La}_2\text{O}_3$ –YSZ powders, the matrix structure remained cubic. Interestingly, the addition of  $\text{La}_2\text{O}_3$  did not destabilize the matrix and the SPS technique was successful in fabricating cubic  $\text{La}_2\text{O}_3$ –YSZ composites. However, there was a considerable shift towards a lower  $2\theta$  angle for the peaks of cubic YSZ, suggesting an increase in the lattice parameter of the cubic YSZ when  $\text{La}_2\text{O}_3$  was added (Table 2). The peak shift in cubic YSZ also indicates that a solid solution formed between  $\text{La}_2\text{O}_3$  and YSZ with accompanying lattice size expansion, since  $\text{La}^{3+}$  has a larger ionic radius than  $\text{Zr}^{4+}$ . Although the peaks corresponding to the monolithic  $\text{La}_2\text{O}_3$  precursor powder could not be detected in the  $\text{La}_2\text{O}_3$ –YSZ composites, new peaks were detected in all of the sintered  $\text{La}_2\text{O}_3$ –YSZ composites. Based on detailed XRD

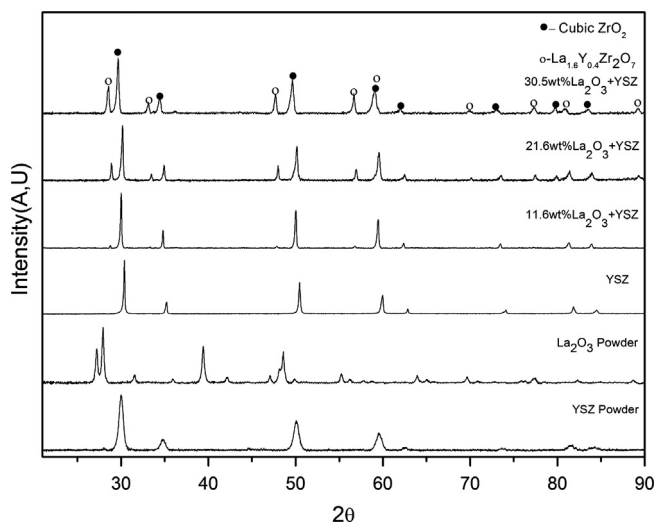


Fig. 1. XRD patterns of the SPS-processed composites.

Table 1  
Chemical composition of precursor powders.

Precursor powder	Concentration (wt%)						Particle Size (nm)
	Zr	O	Y	La	Cl	Hf	
YSZ	60.4	28.7	8.8	–	0.7	1.3	500
$\text{La}_2\text{O}_3$	–	24.1	–	75.9	–	–	~80

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