Effect of a Weak DC Electric Field on the Grain Size Distribution in Isothermally-Annealed Yttria-Stabilized Zirconia (3Y-TZP)

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Abstract

The grain size distributions (GSDs) in 3 mole% yttria-stabilized zirconia (3 Y-TZP) which had been isothermally annealed for various times at 1300°C and 1400°C without and with a weak DC electric field $E_0^* = 18$ V/cm were determined. The GSDs for all test conditions fit a single curve whose form is in accord with the Bitti-Di Nunzio model, and moreover is in accord with the GSDs for sintering and plastic deformation. The results for all three processes suggest the GSD to be a unique function of the mean linear intercept grain size.

Keywords: zirconia, grain size, electric field, annealing

1. Introduction

It is well-known that the processing and properties of polycrystalline ceramics vary with the mean grain size (GS) and, moreover, with the grain anisotropy and size distribution (GSD) [1]. Further, it is now well-established that a weak electric field ($E \leq \sim 25$ V/cm) applied during plastic deformation [2,3], isothermal annealing [4] and sintering [5-9] of 3 mole% yttria-stabilized zirconia (3Y-TZP) retards the corresponding grain growth. In the case of sintering and plastic deformation it was found that the field had no effect on the grain geometry nor the size distribution [10-13]. Moreover, it was found that the form of the GSDs for sintering and plastic deformation

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was in accord with the Bitti-Di Nunzio (B-N) model [14].

The above-mentioned GSD measurements on sintered and on plastic deformed specimens [10-12] suggest that the GSD is a unique function of the mean linear intercept grain size ($\bar{d}$). However, in both of these processes, other phenomena are involved along with grain growth, namely porosity in sintering, and stress, strain and cavitation in plastic deformation. These corresponding phenomena could conceivably influence the development of the GSD. They are however either appreciably reduced or eliminated in isothermal annealing of fully-sintered (relative density $\rho_r \approx 1.0$) specimens. Although the annealing tests by Ghosh et al. [4] confirmed that a small electric field retards grain growth, they did not determine its effect on the GSD. Hence, the objective of the present investigation is to determine the effect of a small electric field during isothermal annealing of 3 Y-TZP on the mean grain size and on the stereology (geometry and size distribution) of the corresponding grains.

2. Experimental

The starting material in the present tests was the same as that in our prior work [6-9], namely 3 mole% yttria-stabilized zirconia powder ($d_0 \approx 26$ nm) purchased from Tosoh Corp.. Details regarding specimen geometry, and the processing and test procedures are given in [7]. Briefly, following cold compaction (98 MPa) of the as-received powder the resulting compacted specimens were heated in air to 800°C and held at this temperature for 2 h to remove the binder. They were furnace cooled back to room temperature (RT) and the “green” specimens were then sintered in air at a heating rate of $\sim 10^\circ\text{C}/\text{min}$ without and with an initial applied DC electric field $E_0 = 14$ V/cm to 1300°C and 1400°C, where they were directly isothermally annealed for 0.5 to 24 h. As a result of the shrinkage during the sintering, the applied electric field at the start of the

annealing process (E_0^*) had become 18 V/cm. Relative density (ρ_r) of the specimen was determined at the end of the sintering treatment and throughout the annealing from the longitudinal shrinkage.

Following each annealing treatment the specimens were furnace-cooled to RT, sectioned, mechanically polished (diamond paste) and thermally etched (1 h at 1200°C in air) and examined by scanning electron microscopy (SEM). The mean linear intercept (MLI) grain size $\bar{d}$ and the corresponding GSD were determined from SEM micrographs which were taken at 3 locations for each annealing treatment, namely: (a) ~ 5 mm below the upper positive (+) electrode, (b) ~ 5 mm above the lower negative (−) electrode and (c) midway between. Approximately 200 intercepts were measured at each location giving a total of ~ 600 measurements for each annealing time. The mean value for each of the three locations varied within $\pm 5\%$ with no clear trend with respect to locations. Also, it was determined, by comparing the mean values from mutually perpendicular lines drawn on the SEM micrographs, that the grain geometry was essentially isotropic (within $\pm 5\%$) at all three locations and for all annealing treatments.

3. Results and Analysis

Examples of the SEM micrographs, the corresponding mean linear intercept (MLI) grain size $\bar{d}$ and the corresponding GSD for the as-sintered specimens in the middle location are shown in Fig.1. To be noted are: (a) the grains are essentially isotropic, (b) the GSDs approximate a log-normal form, (c) the mean grain size increases with temperature and (d) the mean grain size is smaller with application of electric field compared to without. Similar features were observed at the (+) and (−) locations, and for all annealing conditions.

Plots of $\bar{d}$ vs T are presented in Fig.2. It is here seen that: (a) the gain size (GS) increases with annealing time t_a at each temperature, and includes the range from ~ 100 nm to ~ 500 nm,

(b) the GS increases with annealing temperature for each t_a and (c) the GS for each annealing condition decreases with electric field.

Included in Fig.2 is the magnitude of the relative density ρ_r determined from the corresponding specimen longitudinal shrinkage measurements. To be noted are: (a) ρ_r increases from ~ 0.85 to ~ 0.95 for annealing with field at 1300°C and from ~ 0.9 to ~ 1.0 with field at 1400°C and (b) ρ_r decreases with application of the electric field, being especially pronounced at 1300°C .

The normalized (with respect to the mean $\bar{d}$) grain size distributions for all the annealing treatments are presented in Fig.3. It is here seen that the data points for all annealing treatments fit a single curve, which is in accord with the Bitti-Di Nunzio model [14] as expressed by Eq. 9 in their paper, and with the constant A typically having values in the range of 10 to 20. Moreover, the form of the GSDs for annealing is similar to those for sintering [11, 12] and plastic deformation [10], the derived values of A being 12.1 for annealing, 12.3 for sintering and ~ 15.0 for plastic deformation.

4. Conclusions

The results on isothermal annealing along with those obtained previously on sintering and plastic deformation give that: (a) the grain size distribution curves (GSDs) for all three conditions are similar in form, (b) they are a unique function of the mean grain size, and thereby independent of an applied weak electric field, (c) they are in accord with the Bitti-Di Nunzio model, which states that “the coarsening processes are essentially governed by an interface related energy term”.

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Illustrations

Fig.1. SEM micrographs and the corresponding grain size distribution at the middle location of the as-sintered specimens ($t_a=0$).

Fig.2 Relative density ρ_r and mean linear intercept grain size $\bar{d}$ vs the annealing time t_a for the isothermal annealing of 3 Y-TZP without and with all applied DC electric field $E_0^* = 18$ V/cm: (a) 1300°C and (b) 1400°C. From J. Wang, H. Conrad, J. Mater. Sci., 49(17) (2014)6074-6080.

Fig.3. Fit of the present results corresponding to the isothermal annealing of 3 Y-TZP to the Bitti-Di Nunzio grain coarsening model: $\rho = d/\bar{d}$.

Fig.1

Fig.2

Fig.3