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Sintering behavior, microstructural evolution, and mechanical properties of ultra-fine grained alumina synthesized via in-situ spark plasma sintering

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Abstract

Ultra-fine grained Al_2O_3 was fabricated by in-situ spark plasma sintering (SPS) process directly from amorphous powders. During in-situ sintering, phase transformation from amorphous to stable α-phase was completed by 1100 °C. High relative density over 99% of in-situ sintered Al_2O_3 was obtained in the sintering condition of 1400 °C under 65 MPa pressure without holding time. The grain size of in-situ sintered Al_2O_3 body was much finer (~400 nm) than that of Al_2O_3 sintered from the crystalline α-Al_2O_3 powders. For in-situ sintered Al_2O_3 from amorphous powders, we observed a characteristic microstructural feature of highly elongated grains in the ultra-fine grained matrix due to abnormal grain growth. Moreover, the properties of abnormally grown grains were controllable. Fracture toughness of in-situ sintered Al_2O_3 with the elongated grains was significantly enhanced due to the self-reinforcing effect via the crack deflection and bridging phenomena.

Keywords: Alumina; Amorphous; Ultra-fine grain size; Spark plasma sintering; Abnormal grain growth

1. Introduction

Alumina (Al_2O_3) is one of the most widely used structural ceramic materials in the various fields of industry due to its good mechanical properties, excellent oxidation resistance and corrosion resistance [1]. The mechanical properties of Al_2O_3 can be controlled by modification of its grain size as other materials [2]. Especially, it is well known that refinement of the grain size in Al_2O_3 enhances its strength and hardness [3]. Moreover, it is reported that the nanocrystalline ceramic may have low temperature superplasticity [4].

There are two approaches to obtain ultra-fine or nanocrystalline Al_2O_3. One is the sintering of nanocrystalline α-Al_2O_3 powders with restricted grain growth by controlling the sintering parameters such as the heating rate, annealing schedule and applied pressure. The other approach is refining Al_2O_3 grain size by the phase transformation of Al_2O_3 during the sintering process. Al_2O_3 has several low temperature metastable allotropic phases such as γ and θ phase, which are transformed into stable α-Al_2O_3 at the temperature of 1200–1300 °C [5,6]. The refinement of grain size can be obtained by controlling the nucleation and growth of α-Al_2O_3 during phase transformation from metastable phase [7]. However, the sintering of metastable Al_2O_3 phases such as γ-phase is difficult by conventional sintering process. Therefore, new sintering techniques, such as high pressure low temperature (HPLT) sintering, spark plasma sintering (SPS), and sintering forge, have been introduced to obtain ultra-fine or

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nanocrystalline α-Al_2O_3 with full densification from metastable Al_2O_3 powders [8–10].

Recently, a few studies have reported possibility of direct sintering of amorphous Al_2O_3 powder for ultra-fine α-Al_2O_3 [11–13]. However, there are few results reporting the successful densification and refinement of the Al_2O_3 grain size.

In this study, we successfully fabricated fully densified α-Al_2O_3 bodies with the ultra-fine grain size directly from the amorphous Al_2O_3 powders via SPS process. The in-situ crystallization and densification behavior of the studied samples during the sintering process were systematically investigated. Furthermore, this paper describes the microstructural evolution of the in-situ Al_2O_3 with various sintering parameters as well as the relationship between the characteristic abnormal grain growth of the in-situ sintered Al_2O_3 and mechanical properties to verify its self-reinforcement effect.

2. Experimental procedures

2.1. Amorphous Al_2O_3 powder preparation

The precursor used for the amorphous Al_2O_3 powder was Al $(NO_3)_3 \cdot 9H_2O$ (Aluminum Nitrate, Nonahydrate), produced by Junsei Chemical Co, Japan. The amorphous Al_2O_3 was fabricated by heating the precursor at 350 °C for 72 h for dehydration and elimination of the nitrogen compounds resulting in rapid collapse of the hydrate structure and amorphization. The amorphous Al_2O_3 powder was ball-milled with Al_2O_3 ball of 5 mm diameter in the 500 ml polystyrene bottle with 64 rpm. After ball-milling, the amorphous powder was sieved with 35 μm level sieve. The powder size distribution was analyzed by particle counter system (PAMAS-2120). For the sake of comparison, the particle size and size distribution of crystalline α-Al_2O_3 powders (AKP-50, Sumitomo Chemicals, Japan) with particle size of 100 nm were analyzed. The initial powder size and size distribution of amorphous powder and α-Al_2O_3 powder were similar as shown in Fig. 1. The estimated mean diameter sizes of amorphous and crystalline powders were 0.102 and 0.089 μm, respectively.

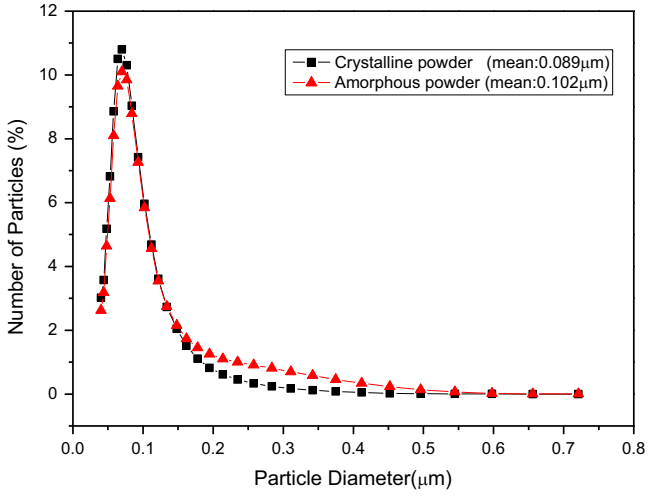

Fig. 1. Particle size distribution of amorphous Al_2O_3 powders and crystalline α-Al_2O_3 powders before sintering process.

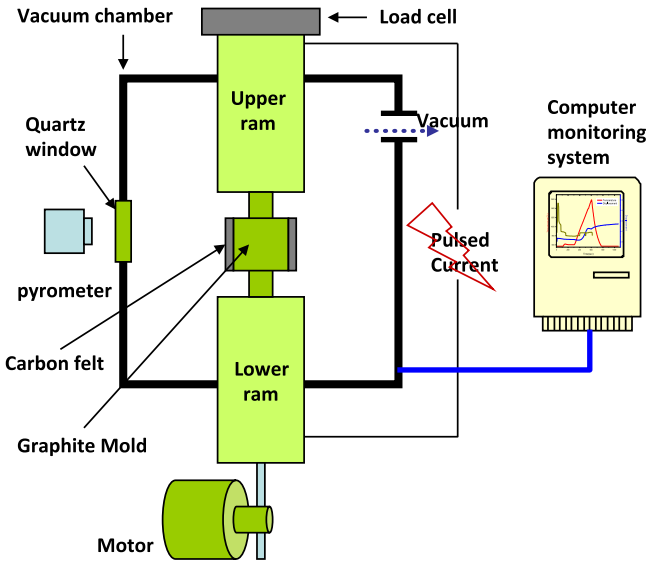

Fig. 2. Schematic illustration of SPS system.

2.2. In-situ spark plasma sintering process

The amorphous and crystalline α-Al_2O_3 powders were sintered by SPS (Dr. Sinter SPS-515S, Japan) within a graphite mold of 15 mm diameter. The schematic diagram of the SPS system is shown in Fig. 2. In the SPS process, the loaded powders in the graphite mold were heated by a pulsed electric current over 1000 A. The exact sintering mechanism during the SPS is not yet understood clearly. However, some mechanisms have been proposed [14]. Tokita et al. suggest that the sparked plasma generated between the particles removes the oxide layer easily. In addition, owing to the pulsed current, the high voltage and current are applied between particles by its large impedance.

The sintering temperature was varied from 1300 °C to 160 °C and heating rate was maintained at 20 °C/min in a vacuum atmosphere under a pressure of 65 MPa. The shrinkage, temperature and vacuum pressure of specimen were monitored during the sintering process. The relative densities of the sintered specimens were determined by Archimedes' method with distilled water as the immersion medium at least 5 times.

2.3. Characterization

The microstructures of the sintered Al_2O_3 were observed by scanning electron microscope (SEM) after polishing the sample surface with diamond slurries down to 1 μm and thermal etching at 1300 °C for 15 min. Based on the microstructural analysis, the grain size and distribution of the specimen were estimated using an image analyzer (analySIS v3.1, GmbH).

The fracture toughness was evaluated by the indentation microfracture (IM) method by and calculated by using the following equation by Anstis et al. [15],

$$K_{IC} = 0.016\sqrt{\frac{E}{H}}\left(\frac{P}{L}\right)^{1.5} \quad (1)$$