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Effect of temperature difference on presintering behavior of gelcast thick alumina bodies

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Abstract

Alumina spherical green bodies (∅ ≈ 75 mm) prepared by gelcasting were presintered at 1000 °C. Green bodies' temperature difference between the center and surface as a function of solids loading, heating rate, and gel system were extensively investigated. Temperature in the center was lower than that on surface of the alumina spheres cast using PIBM gel system during presintering, and temperature difference (ΔT: $T_{\mathrm{center}} - T_{\mathrm{surface}}$) slightly decreased with increased solids loading but significantly increased with heating rate. Temperature difference ($\Delta T < 0$) generated compressive stress near the surface and tensile stress in center, which led to a porosity difference between the center and the surface. Moreover, the porosity exponentially decreased with increased compressive stress. Tensile stress in the center of the green body cast using PIBM gel system was estimated as 0.3 MPa, which was less than the minimal strength. Therefore, the body was successfully presintered without cracking. However, cracking was found on the surface of the body cast by epoxy-amine gel system, which exhibited a more severe temperature difference fluctuation.

Keywords: Gelcasting; Presinter; Temperature difference; Thermal stress

1. Introduction

Gelcasting is the most promising way to prepare large and/or complex ceramic components with a homogeneous microstructure using simple equipment and processing techniques [1–4]. However, cracks are easily generated during presintering because of the relatively low strength of the green body and subsequent heat treated body.

During the presintering stage, the stress and strength of the green body evolve with the increase of temperature and cracks are generated if the tensile stress is larger than its strength. There are many factors that generate stress in the green body [5]. Internal factors include complex chemical and physical processes caused by organic removal [6–8], and microstructure inhomogeneity. External factors include a temperature difference caused by heat transfer [9]. In the past few decades, the majority of efforts have been made on the decomposition kinetics and models during the burnout process in ceramic green bodies by injection molding [10,11]. However, the organic removal problem is less severe for the gelcast green bodies, because much less organic additives were used. For example, only 0.3 wt% organics was employed to gelcast alumina by a new gel system [12]. On the other hand, the temperature difference problem between the surface and inside of the green body with a large dimension should be given more attention, because the thermal conductivity is unfavorably low in ceramic green body [13]. Yet, few studies involved the temperature difference within green bodies during presintering, which would cause thermal stress. Furthermore, the relationship between thermal stress and density (porosity), stress and strength of the green bodies during presintering are not clarified.

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This study focused on the temperature difference evolution and the thermal stress and strength of the gelcast alumina green bodies during presintering. Alumina spheres with a diameter of ~75 mm were prepared from slurries using a nontoxic and water-soluble co-polymer of isobutylene and maleic anhydride (PIBM, hereinafter), acting as both a dispersant and gelling agent [12,14], and using epoxy resin and amine gel system [15], respectively. The temperature difference within the alumina green bodies is tested as a function of solids loading, heating rate, and gel system. The relationship between compressive stress and porosity was investigated by presintering green bodies under different loads. Based on this result, the tensile stress was estimated and compared with the strength of green bodies. Finally, feasibility of presintering a thick ceramic green body was demonstrated.

2. Materials and methods

2.1. Sample preparation

A commercial alumina powder (AES-11, Sumitomo, Osaka, Japan) with an average particle size of 0.45 μm was used as the raw material. PIBM with an average molecular weight of 5500–6500 (Isobam 600AF, Kuraray, Osaka, Japan), was used as both dispersant and gelling agent. Isobam 104 from the same company with a molecular weight of 55,000–65,000 was added to increase the gelling ability. The process for preparation of alumina green bodies using PIBM was similar to a previous report [12]. The slurries containing 50–58 vol% solids were prepared by planetary ball milling the deionized water, alumina powder, and 0.3 wt% PIBM (0.2 wt% Isobam 600AF and 0.1 wt% Isobam 104, relative to the weight of alumina powder) for 1 h at a speed of 250 r/min. The alumina green body was also prepared from 50 vol% solids by an epoxy-amine gel system containing about 4.5 wt% organics [15]. Before casting, the slurries were centrifuged and degassed in a planetary vacuum mixer (ARV-310, Thinky, Tokyo, Japan) to reduce trapped bubbles. Then, the slurries were cast into a spherical mold made by polyethylene (plastic ball) with the diameter of 80 mm. After gelling and demolding, the wet alumina spherical bodies using PIBM, and epoxy resin and amine (noted as Al–PIBM sphere and Al–EA sphere, respectively) were dried in a commercial dryer (HW-150, Sumsung, Shanghai, China) under a controlled temperature and relative humidity (Temperature = 40 °C; $RH = 80\%$) for two days and put into an oven at 60 °C until a constant weight was attained. Presintering of the spheres was carried out by heating to 1000 °C with different heating rates in the muffle furnace (CWF12/13, Carbolite, United Kingdom).

2.2. Temperature measurements

To evaluate the temperature difference between the center and surface of the spheres, holes with a diameter of 5 mm were drilled to a depth of 4 and 37 mm separately in a sphere. Then K-type thermocouples were put into the holes. To avoid direct heat transfer, the residual powders after drilling were filled in the gap between thermocouple and the wall of the hole. Thermal signals were recorded by a computer. The temperature difference (ΔT) was defined as T_{center}–$T_{surface}$. Here T_{center} and $T_{surface}$ represent the temperature of center and surface of alumina sphere, respectively. The setting of alumina sphere and thermocouples in a furnace was shown in Fig. 1.

Fig. 1. The setting of alumina sphere (∅ ≈ 75 mm) and thermocouples in a furnace.

2.3. Compressive stress–porosity measurements

A special experiment was designed to evaluate the relationship between compressive stress and porosity of the green bodies during presintering. Cylindrical specimens with a dimension of ∅10 × 5 mm, prepared from 50 vol% solids by the PIBM gel system, were presintered at a heating rate of 0.5 °C/min under different loads. The specimens were cut from the top surface to about 3 mm thick (~0.5 g) for porosity measurement using mercury porosimetry with a PoreMaster (PoreMaster-33, Quantachrome Corporation, Boynton Beach, FL). Weight loss and organic oxidation were evaluated with a TGA (STA449C, Netzsch, Germany) at a heating rate of 2 °C/min from room temperature to 1000 °C.

2.4. Strength measurements

To evaluate the strength evolution of the green bodies with increased temperature, bars with a dimension of 8 × 8 × 50 mm^3 were heated to the designated temperature for 10 min at a constant rate (2 °C/min) and the strength test was performed at room temperature [5]. The bending strength was averaged from 5 bars which were measured with a three-point test at a crosshead speed of 0.5 mm/min (Instron5566, Norwood, MA).

3. Results and discussion

3.1. Influence of solid loading on temperature difference

Temperature difference as a function of solids loading for Al–PIBM sphere was shown in Fig. 2(a), where the heating rate was 0.5 °C/min. The temperature near the surface was higher than that in the center because the heat was transferred from the