

Freeze granulation: Powder processing for transparent alumina applications

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Available online 26 March 2012

Abstract

Use of freeze granulated powders is successfully used as an industrially viable alternative to loose powder sintering for transparent polycrystalline alumina (PCA). Freeze granulation with narrow granule size distribution was realized after suspension condition optimization, with very good flowability and regular spherical shapes. The key factors are low viscosity slurries linked to the complex interactions between the organic processing additives and their interaction with dopant ions in solution. Real in-line transmittances (RITs) of 52% were achieved by pulsed electric current sintering (PECS) of dry pressed green bodies from doped granulated powders. This is the first example of a high RIT (>50%) alumina produced from simple dry pressing of a granulated powder. The results indicate that higher granule solid loads and lower organic additive concentrations give better RITs after PECS. Aging of the powder slurry before freeze granulation proves a crucial step for the optical performance of the final transparent PCA.

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Keywords: A. Sintering; C. Optical properties; D. Al_2O_3 ; E. Armor; SPS

1. Introduction

The constant effort to improve the mechanical properties of ceramic materials by microstructure refinement and defect reduction has ultimately led to scientific research and development of transparent ceramic materials. More than any mechanical properties, the optical performance relies on defect-free processing to allow full densification and residual porosity elimination.^{1–3} In fact, pores are highly efficient scattering centers depending on their size due to the large refractive index difference between the pore and the ceramic bulk material.^{4,5} For anisotropic materials such as alumina, microstructural refinement is an additional requirement to reduce inter-granular scattering due to birefringence.³

In recent years, many advances have been made in production of transparent ceramics by various processing and sintering strategies.^{1,6–14} Mainstream ceramic processing techniques rely either on dry^{11,14,15} or wet powder processing [1,6,9,13,16]—both allowing production of highly dense green bodies (GBs), one key factor for transparent ceramics—as

well as on classic sintering followed by hot isostatic pressing (post-HIP) or novel fast sintering techniques. Among the latter, the most popular for transparent ceramics is pulsed electric current sintering (PECS), also known as spark plasma sintering (SPS).^{7,8,11,12,14,17–21} In general, powders for these fast sintering techniques have been loose or freeze dried powders^{11,14,15,19,20,22} not necessarily well suited to industrial scale production. Whatever the sintering technique used, increasing focus must be put on industrialization aspects, and especially on the green body fabrication method. Wet processing is rather slow and loose powder handling too difficult and possibly hazardous for processing and health aspects, respectively.

To improve the powder flowability and prevent individual particles from becoming airborne, granulation is the classical approach. When well controlled, self-flowing granulated powders allow automated production of highly dense and defect-free green bodies by relatively quick and simple uniaxial or isostatic pressing methods. The main granulation method used in ceramic processing is spray drying,^{23–25} with freeze granulation attracting increasing interest.^{26,27}

During spray drying, a powder suspension is sprayed into a hot air cyclone.²⁸ During drying, the droplets shrink to form powder granules. These granules may be more or less spherical and/or hollow depending on the slurry formulation (i.e. powder load and additives) and drying temperature.^{25,29} Binder

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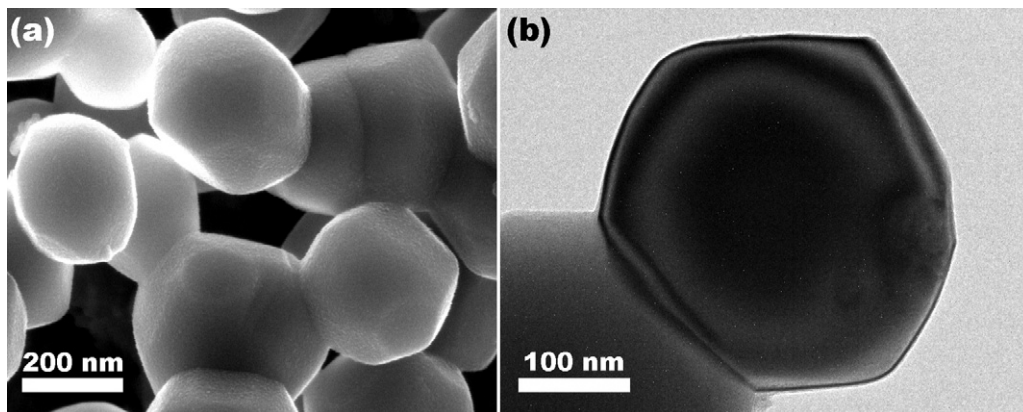


Fig. 1. (a) SEM image and (b) TEM image of the α - Al_2O_3 powder used.

migration during the drying can additionally reduce the homogeneity and thereby quality of the granules, leading to inhomogeneities in the green bodies.³⁰

Freeze granulation is an alternative to spray drying, solving many of its limitations. During freeze granulation, droplets are rapidly frozen in liquid nitrogen before being recovered and subsequently freeze-dried. Freezing being a fast process, diffusion dynamics are insufficient to induce any binder migration, thereby better preserving the granule homogeneity.^{26,27} Additionally, the granules do not shrink during the whole process and therefore maintain their shape and powder density—consequently allowing higher granule deformability compared to spray drying.

In the present study, freeze granulation is explored as an alternative to simple freeze drying for the preparation of transparent polycrystalline alumina (PCA) by PECS in a first step toward a more industrially friendly forming for PCA. The strong dependence of the real in-line transmittance (RIT) on the residual porosity and grain size inhomogeneity makes the RIT an ideal parameter to control the quality of the powder compact. Doped granules with varying solid loads and additives are produced by an experimental setup facilitating control over the granule size and allowing for narrow granule size distributions. As a reference of the granulation performance, the results are compared with those obtained by PECS on the same loose, identically doped and freeze-dried powder. Indeed, use of loose freeze dried powder for PECS has shown promising results with RITs around 56% in an earlier study.¹¹ That 56% value is only five percent below the expected maximum RIT performance taking into account the particle size of the starting powder, defining the minimal theoretic final grain size. The goal of the study is to show the feasibility and performance of PCA production by simple dry pressing of granulated powders, not yet seen in the literature, compared to reported loose dry or wet green body powder processing techniques.

2. Materials and experimental methods

The powder used was a polyhedral near-spherical high-purity α - Al_2O_3 (Sumitomo, Japan) with a median particle size D_{v50} of 510 nm (Laser diffraction, Mastersizer, Malvern, UK), a total

impurity concentration of less than 0.01 mass% (≤ 5 ppm for Si, Na, Mg, Cu and Fe) and a specific surface S_{BET} of $4.2 \text{ m}^2/\text{g}$ (Fig. 1).

The viscosity of the powder suspension was measured by rheological measurements (RheoStress RS100, Haake, Germany) using a double-gap DIN 53544 concentric cylinder setup. During the rheology measurements, the setup and the slurries were kept at $25 \pm 0.1^\circ\text{C}$ by a thermostatic bath. The data acquisition cycle was: (1) ramp from 0 to 200 s^{-1} in 90 s, (2) hold at 200 s^{-1} for 60 s, and (3) decrease from 200 to 0 s^{-1} in 90 s.

ζ -Potential measurements (AcoustoSizer II, Colloidal Dynamics, USA) were performed during titration by aqueous dopant-nitrate solutions. The powder was dispersed in 0.005 M HNO_3 and the pH adjusted by an ammonia solution (NH_4OH). The total powder solid content was 2.5 wt.% with or without 1 wt.% poly-acrylic acid (PAA) added with respect on the total powder content mass. To avoid interactions between PAA and baseline electrolyte ions, no other salt was added.

To dope the powder and prepare the suspensions for freeze granulation, 50 g of powder was dispersed in ultra-pure water before addition of the desired amounts of a 10 wt.% PAA solution (M_w 2000, pH 9.5) as a dispersant. After an ultrasonic bath (UB) treatment of 15 min, Mg^{2+} , Y^{3+} and La^{3+} aqueous nitrate solutions (purity >99%, Fluka for La- and Aldrich for Mg- and Y-hexa-hydrated nitrates) were added. The final total dopant level was 450 ppm total cationic ratio (150 ppm for each). Finally, aqueous solutions of polyvinyl alcohol (PVA, Mowiol 4-88, Aldrich) as a binder and polyethylene glycol (PEG, M_w 3000 or 300, Fluka) as a plasticizer were added together with octanol as anti-foaming agent. The final slurries with a pH around 9 were stirred and UB treated for another 15 min before the freeze granulation. The various slurry compositions are summarized in Table 1.

For freeze granulation, the slurry was drawn into a syringe through a $70 \mu\text{m}$ filter to safely avoid any agglomerates that may obstruct the granulation nozzle. The syringe was then inserted into a syringe pump, shaken periodically throughout the granulation process to avoid PVA segregation, and the freeze granulation process started. The freeze granulation setup (Encapsulator, Inotech Encapsulation AG, Dottikon, Switzerland) is schematically represented in Fig. 2. The slurry droplets are formed by

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