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Consolidation of nanoparticle suspensions by centrifugation in non-porous moulds

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Abstract

Alumina and zirconia nanosuspensions with a mean particle size of 100 nm and 15 nm, respectively, were consolidated by centrifugal compaction in non-porous moulds. The nanosuspensions consolidated by high-speed centrifugation were deposited irregularly, resulting in a powder deposit with density profile. The homogeneity of the powder deposits was described and homogeneous and well packed deposit regions were identified. Plate-like bodies were prepared from the homogeneous regions of the deposit. The advantage of regular and dense nanoparticle packing by centrifugal compaction was demonstrated by fabricating transparent alumina and tetragonal zirconia ceramics. The transparent alumina had an in-line transmission of 55% in the visible light at a thickness of 0.8 mm. The transparent tetragonal zirconia reached a dense microstructure with an average grain size of 65 nm and an in-line transmission of 25% at a thickness of 0.5 mm.

Keywords: A. Suspensions; D. Al_2O_3; D. ZrO_2; Centrifugal compaction

1. Introduction

The utilisation of ceramic nanoparticles represents one of the most challenging tasks in the field of bulk ceramics processing. Due to an inherent tendency of nanoparticles to strong agglomeration, the compacted green bodies show an irregular and loosely packed structure. Colloidal processing of nanoparticles is a useful approach to dispersing and/or removing the agglomerates and obtaining stable and well-dispersed nanoparticle suspensions [1]. To fully exploit the advantages of colloidal processing, ceramic bodies must be formed directly from the slurry state [2]. However, the consolidation of ceramic nanosuspensions brings extraordinary difficulties. Direct-casting methods such as direct coagulation casting or gelcasting require high powder loading, which can be difficult to reach in nanoparticle suspensions [3]. Slip casting of nanosuspensions is limited by the low suction power of common porous moulds. Moreover, this suction decreases with increasing sediment thickness [4]. Similar problems with filter and sediment permeability can also be found in pressure filtration [5]. Alternative approaches to the processing of low-concentration nanoparticle suspensions must therefore be used. This contribution investigates a simple alternative method for consolidating nanoparticle suspensions into a bulk ceramic body, using centrifugal forces. During centrifugal consolidation in non-porous moulds the centrifugal force is acting on every ceramic particle in the suspension. The particles move in the direction of the acceleration and deposit on the mould wall, whereas the solvent moves in the opposite direction. The solvent is neither transported through the deposit (as in slip casting or pressure filtration) nor stays in the particle compact formed (as in direct coagulation casting or gelcasting). The drawback of the centrifugal consolidation method is the possible particle size segregation and microstructural gradient development in the deposit due to different settling of particles of different sizes and/or densities. This can be eliminated by

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using weakly flocculated suspensions (prepared from dispersed suspensions by adding a large amount of indifferent electrolyte) that are characterised by high viscosity with a pronounced yield stress [6]. Such an approach, however, results in a low-density deposit often with an inhomogeneous density profile [7–9]. A more suitable approach to overcoming the particle size separation problem utilises highly concentrated dispersed suspensions. The mass segregation can be avoided by hindered sedimentation in these high-solid-content suspensions [10,11]. It has been reported that even phase segregation in a composite system Al_2O_3/ZrO_2 could be successfully avoided using a concentrated suspension [12]. Moreover, a well-dispersed suspension produces a dense and regularly packed particle deposit that exhibits good sintering behaviour and a defect-free microstructure [10,13–15].

The sedimentation of dispersed colloidal suspensions with spherical and monosized particles in submicron range meets the condition for creeping flow that can be described by the equation [16]

$$\frac{dr}{dt} = K(\phi)\frac{d^2(\rho_s - \rho_l)g_0}{18\eta} \quad (1)$$

where dr/dt denotes the sedimentation rate, d is the particle diameter, ρ_s and ρ_l are the density of the particles and the liquid, respectively, η denotes the liquid viscosity, and g_0 is the particle acceleration. For centrifugal deposition $g_0 = r\omega^2$, where r is the centrifugal radius and ω is the angular speed. This equation describes the Stokes law for an isolated sphere modified by $K(\phi)$ so as to allow formally for an increase in fluid drag due to hydrodynamic interactions in the multiparticle system. This model assumes no other interaction between particles, except hydrodynamic effects. Brady and Durlofsky [17] have suggested a simple analytical expression for $K(\phi)$:

$$K(\phi) = 1 + \phi - 0.2\phi^2 - \frac{6\phi}{5}\left(\frac{5 - \phi + 0.5\phi^2}{1 + 2\phi}\right) \quad (2)$$

where ϕ denotes the particle volume fraction. Another correction was derived by Barnea and Mizrahi [18]:

$$K(\phi) = \frac{1 - \phi}{(1 + \phi^{1/3})\exp\{5\phi/3(1 - \phi)\}} \quad (3)$$

Dilute suspensions ($\phi < 0.1$) are sufficiently well described by Eqs. (2) and (3) but the behaviour of suspensions deviates from these models at a higher volume loading. Therefore Richardson and Zaki [19] have proposed an empirical correlation in the general form

$$K(\phi) = (1 - \phi)^{\alpha} \quad (4)$$

with $\alpha \approx 5$. Buscall et al. [20] have shown that the value $\alpha = 5.4$ gives a good fit for the suspension of silica particles with a diameter of 200 nm in a range of solid volume fraction from 0 to 0.5.

Centrifugal consolidation of nanosuspensions brings additional difficulties compared with submicron suspensions. It is not only the smaller particle size that makes the dispersed nanoparticles more stable against deposition. The surface forces become increasingly important and change the behaviour of dispersed nanoparticles. The adsorbed ions or polymer species increase the effective volume of dispersed nanoparticles and decrease their overall density [21]. Also the interparticle interactions play a more important role in nanosuspensions than in submicron suspensions because of the much closer contact between nanoparticles at the same suspension loading. Due to these reasons the commonly used centrifugal accelerations in the range of 10^3–$10^4 g$ (where g is the gravitational acceleration) are not sufficient to deposit the nanoparticles and the acceleration has to be increased above $10^4 g$ [22].

Although many investigations have proved the efficiency of centrifugal consolidation for the submicrometre-sized particle suspensions [10,12–15,23], reports on centrifugation of ceramic nanosuspensions are limited. Therefore the objective of the present work was an investigation of centrifugal consolidation of ceramic nanosuspensions and the evaluation of the resulting nanoparticle deposit from the viewpoint of obtaining dense, defect-free nanostructural ceramics.

2. Experimental procedure

Alumina powder (Taimicron TM-DAR, Taimei Chemicals Co., Japan) with a mean particle size of about 100 nm and a specific surface area of 14.5 $m^2\,g^{-1}$ was used for the preparation of alumina suspensions (see Fig. 1). A commercial dispersant, Dolapix CE 64 (Zschimmer & Schwarz, Germany), was used (1 wt% of ceramic powder) to stabilise alumina particles in the water suspension. Suspensions with two volume loadings were prepared, namely with 20 and 47.5 vol % ceramic powder. The ceramic suspensions were treated with ultrasound for 10 min and mixed for 24 h prior to centrifugation. The suspensions were poured into a two-part mould-container with rubber insert (Fig. 2) and then centrifuged (3K30, Sigma, Germany) with centrifugal accelerations of 12,500g, 25,200g, and 54,500g for 5, 10, and 20 min. The schematic diagram of the centrifuge with the mould is shown in Fig. 3. The mould design enabled removing the deposit from

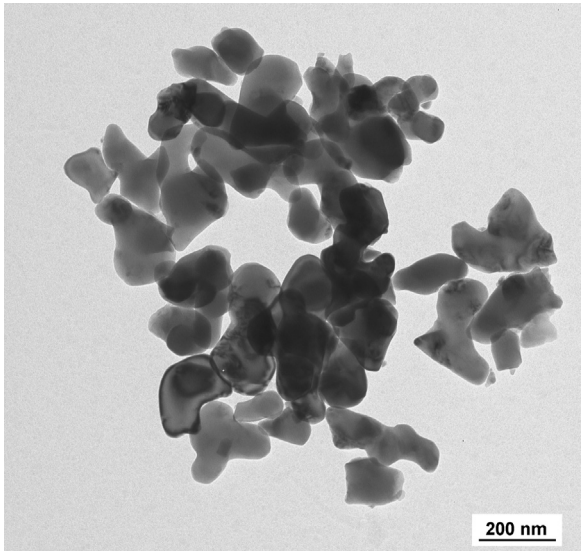

Fig. 1. TEM micrograph of alumina powder.