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Ceramics International 25 (1999) 491–495

CERAMICS
INTERNATIONAL

Pressure slip casting of bimodal silicon carbide powder suspensions

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Received 28 April 1997; accepted 22 September 1997

Abstract

Two silicon carbide powders with different particle size distributions were blended in various proportions and then dispersed in aqueous media with a deflocculant. Bodies were consolidated from these suspensions via pressure slip casting. The relative density and the water content of the green bodies were measured and the optimum body in terms of packing ability was then chosen to evaluate the effects of: (i) the amount of deflocculant, (ii) the casting time on deposition rate, (iii) the microstructure of the consolidated layers. The results showed that the proportion between coarse and fine powders that produced a maximum packing density is independent of applied casting pressure but that the packing degree is normally lower than that obtained by normal slip casting. The amount of deflocculant required to maximise the green density within pressure casting was found to be higher in comparison with slip casting. Further, as the casting time increased the packing density continuously improved. These results may be interpreted in terms of deposition rate and time allowed for particle rearrangements. © 1999 Elsevier Science Ltd and Techna S.r.l. All rights reserved

1. Introduction

Ceramic materials based upon silicon carbide exhibit an interesting set of physico-chemical properties that make them potential candidates for various structural, electrical and high temperature applications [1–4]. It is commonly accepted that the sintering step and the achievement of fully dense microstructures are the most difficult aspects in the processing of this material. This difficulty has been attributed to the covalent nature of the chemical bond, particularly, the low surface energy and the low self diffusion coefficient [5]. The attainment of fully dense sintered bodies depends directly on the density and the uniformity of the of green microstructures. These features can be improved by using colloidal shaping techniques which enable manipulation and control of the interaction forces between the particles [6–8]. Interparticle potentials also play a dominant role in governing the packing behaviour and the rheological properties of these slurries and green bodies. Other relevant factors that can determine the manner in which individual particles pack together in the powder compact are the particle (aggregate) size distribution. The use of bimodal mixtures enables to prepare green bodies with high packing density and narrow pore size distributions [9,10]. However, particle segregation phenomena may occur at low deposition rates, such as those used in slip casting [11]. Segregation phenomena can be prevented by increasing the consolidation rate. This makes the driving force of the casting process an important factor for achieving a high degree of homogeneity. Milne et al. [12] suggest that an important consideration in achieving uniform sintering is to maximise the number of interparticle contacts throughout the compact and that bimodal, rather than monosized, systems should be adopted. Liu [13] emphasised the importance of a narrow pore size distribution within the green compacts on densification of SiC–Al_2O_3–Y_2O_3. The aim of this work was to evaluate the influence of particle size distribution, the amount of deflocculant and the casting time upon the packing degree and the porous microstructure of silicon carbide bodies obtained by pressure slip casting.

2. Experimental procedure

2.1. Materials and reagents

The starting materials were two commercially available SiC powders, NF0 and 1200P (Elektroschmelzw Kempten, GmbH, Germany) with mean diameters of

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PII: S0272-8842(97)00083-7

1.2 and 13 m, respectively. The deflocculant used was an ammonium salt of a low molecular weight polycarbonate acid (Targon 1128, Benkiser-Knapsack GmbH, Germany).

2.2. Techniques

Slurries with a total solid content of 70 wt%, in which the fine fraction varied between 0.4 and 0.7, were prepared. The fine powder was first added to an aqueous solution containing a fixed amount (1 wt%) of dispersant, hand stirring, followed by simultaneous stirring and ultrasonic dispersion for 10 min. The coarser component was then added and the simultaneous stirring and ultrasonic dispersion continued for a further 10 min. Rheological properties of the slurries (flow curves) of the finer powder was already evaluated using a cone-plate type viscometer [14]. Due to the possibility of the coarse particles damaging the instrument, only the apparent viscosity was measured for the bimodal suspensions, using a Brokfield RVT viscometer at a constant rate of 50 rpm. The rheological measurements were correlated with the slip casting performance of the slurries. Considering these results, the optimum coarse/fine powder's combination was then chosen to study the effects of the dispersant concentration and the casting time upon the characteristics of the green bodies. The pressure slip casting experiments were performed at a constant applied pressure of 588 kPa in an apparatus designed by the authors as previously described [15].

Subsequently, the green density was measured on a mercury balance, and the pore size and pore size distributions of the green bodies were determined with a Hg porosimetry (PoreSizer 9320, Micromeritics,USA). The high pressure part of each experiment was carried out in the automatic mode with an equilibration time of 10 s at each point.

3. Results and discussion

3.1. Effect of fine (coarse) fractions in the mixture

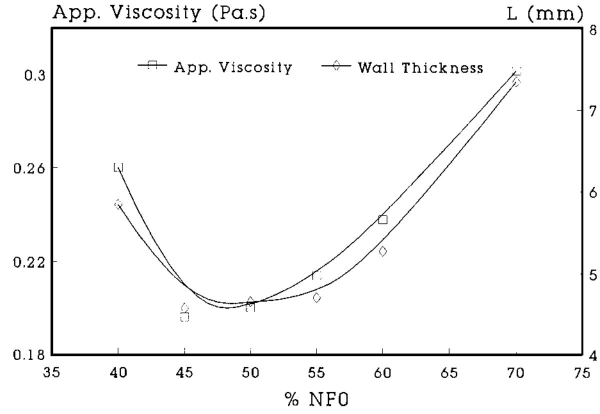

Fig. 1. Effects of the fine (coarse) fraction in the mixture on apparent viscosity of the slurries and on wall thickness. Casting time = 2 min.

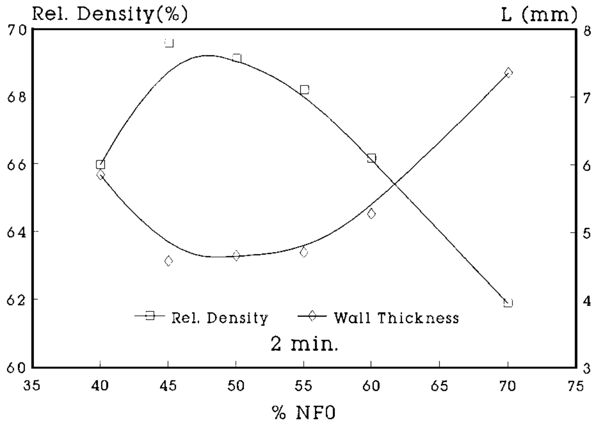

Fig. 2. Correlation between packing density and wall thickness of the consolidated layer.

The particle size distribution will influence the packing ability of the powders both within a suspension and within consolidated bodies. Further, it will determine the rheological properties of the slurries and the permeability of the green microstructures. Fig. 1 illustrates the evolution of apparent viscosity of the slurries and the thickness of the consolidated layer for a casting time of 2 min. These results suggest a close relationship between the rheological properties of the suspensions and the microstructures of the resulting green bodies. The plotted results in Fig. 1 indicate a minimum at a fine/coarse ratio of 45/55, which also corresponds to a maximum packing density, as can be observed in Fig. 2. These trends are in agreement with those observed in previous slip casting experiments [11,16] and suggest that the best fine/coarse combination, for optimum packing ability, was independent of the applied pressure within the range of the study. However, the maximum relative density achieved by pressure casting is about 3% lower than that obtained from the same suspensions by slip casting. This difference may be explained as follows. When well dispersed slurries are used, the repulsive forces between suspended particles allow them to flow and pack as individuals. In slip casting, the rate of deposition is relatively low. Since the particles are still repulsive as they join the consolidation layer, they are free, within certain translational limits, to move to positions of lowest free energy, promoting a closed packed arrangement. When an external pressure is added to the suction pressure of the plaster mould, the number of particles that reach the interface between cake and slurry, per unit time, increases. The lateral rearranging movements of one particle might now be restricted by new incoming particles and a decrease in the packing density should be expected [16].

The proportion fine/coarse = 45/55 corresponds to the situation were fine particles can occupy the interstices between the coarser ones, according to the Furnas model [9,16]. Lower fine fractions will result in greater apparent volumes of the solid phase and a greater amount of liquid retained within the interstices between coarse particles, and the apparent viscosity should increase, as observed. For higher fractions of the fine component, one can expect an increase in the apparent volume of solids, but also, the resulting higher specific surface area exposed to the liquid phase will contribute to an observed increase in apparent viscosity. Figs. 1 and 2 suggest that the relative density (permeability) of consolidated bodies might play a dominant role in the casting rate.

3.2. Effects of the amount of deflocculant and casting time

The evolution of the relative density of the samples (prepared from suspensions containing different amounts of dispersant) is shown in Fig. 3, as a function of casting time. It can be observed that the packing density increases with casting time, this is somewhat more pronounced for samples with lower dispersant contents. This trend is in agreement with observations made by Fennely and Reed [17,18] and Lange and Miller [19] who found that dispersed slurries produce much higher packing densities compared to flocked slurries. In addition, Lange and Miller found that the packing density of the dispersed slurries was relatively pressure independent, whereas, the packing density of the flocked slurries was very pressure dependent. In the present work, significant differences between dispersed and non dispersed slurries were not observed.

A greater packing efficiency is achieved at 0.3 wt% Targon 1128. This amount of deflocculant is 2–3 times more than that required for slip casting (0.1–0.15 wt%) [16]. Under applied external pressure, the higher deposition rates result in higher shear rates. Consequently, the optimum amount of dispersant adsorbed at particle surface, for the slip casting process, may not provide sufficient coverage to cope with the higher shear rates produced in the pressure casting process. The adsorption of a grater amount of polyelectrolyte at particle's surface will be necessary to lubricate the particle contacts and promote particle rearrangements.

The evolution of the wall thickness as a function of casting time is plotted in Fig. 4 for the different dispersant concentrations used. The results in Fig. 4 can be approximated by the form of the parabolic law, which may be described by Eq. (1) [15,16]:

$$L^2 = 2K\phi Pt/\eta(\phi_c - \phi) \quad (1)$$

where L is the thickness of the consolidated layer, K is the permeability, (inversely related with the resistance to fluid flow through the consolidated layer), η is the liquid viscosity and ϕ and ϕ_c are the volume fractions of particles within the slurry and cake, respectively, P is the total pressure (suction pressure of the plaster mould + applied pressure) and t is the casting time.

This equation assumes unidirectional filtration, negligible mould resistance, constant rheological properties of the suspension, absence of particle settling, and constant permeability, that is the body is incompressible. Except for constant permeability, these assumptions are not contradicted in the current work [16]. In fact, the increasing green density observed with the increasing casting time will result in an incremental resistance offered by the consolidated layer to the fluid flow, relative to that expected from Eq. (1). As a consequence, the deposition rate will be significantly reduced. This will allow more time for "spontaneous" particle rearrangements [16]. Alternatively, "forced" particle rearrangements caused by the applied pressure, due to static

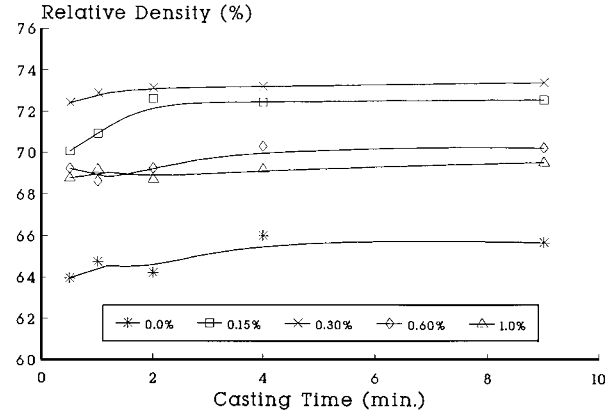

Fig. 3. Influence of the amount of dispersant and casting time on green packing density.

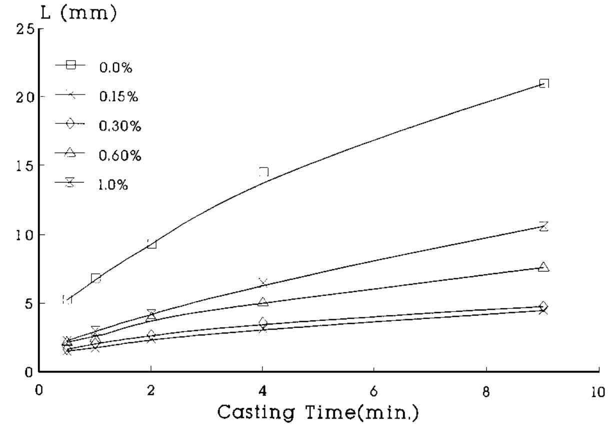

Fig. 4. Influence of the amount of dispersant and casting time on wall thickness.

instabilities, as discussed by Kuhn et al. [20] for a touching and coagulated particle network, seems less probable with a particle size distribution which has a higher packing potential. It is proposed that the differences between the dispersed and the non dispersed systems can be attributed to this particle rearrangement mechanism.

The plots of L^2 vs casting time are shown in Fig. 5 together with the slopes of the fitted lines. According to Eq. (1), these slopes represent the ratios

$$2K\phi P/\eta(\phi_c - \phi)$$

The cake permeabilities, K, calculated from these slopes are presented in Table 1. These results explain the data reported in Figs. 3 and 4 and show a good consistency. It is evident that the probability of particles being individually deposited and forming a close packed structure, increases with deflocculation degree [8]. These structures are less permeable and hence offer a higher resistance to fluid flow and the rate of built up at the wall reduces. Less stable slurries enable particle association and the formation of open and voluminous flocs which, upon deposition, result in more permeable structures. Fig. 6 shows the pore size distributions of the different dispersant contents. For the samples with 0.15 and 0.3 wt% Targon 1128 the results were very similar and, for clarity, only the last one was displayed. As expected, the sample with 0.3 wt% Targon 1128 exhibits the narrower pore size distribution, whilst the opposite can be seen for the sample without deflocculant. Excessive deflocculation produces intermediate cases in which both pore size and pore size distribution tend to increase gradually. These results confirm that the structure of the particles in a suspension are transmitted to the solid wall being formed, as was already suggested [4,11,15,16]. This has a significant influence upon the properties of the material in its final form [12,13].

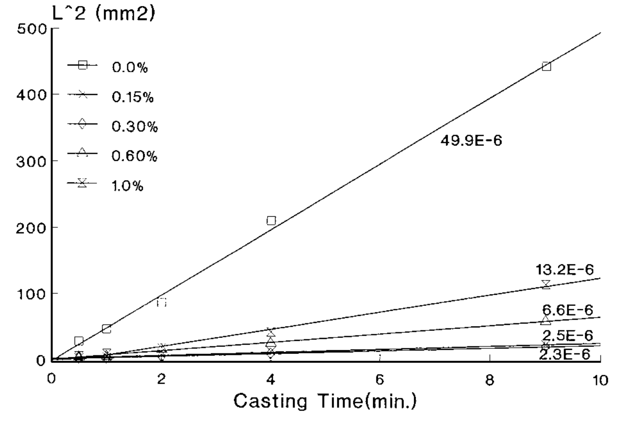

Fig. 5. Influence of the amount of dispersant on the kinetics of the pressure casting process.

Table 1
The cake permeabilities calculated from the slopes indicated in Fig. 5

Targon 1128 (%)	0.0	0.15	0.3	0.6	1.0
Permeability (m2)×10^{15}	18	2.5	2.3	2.8	5.6

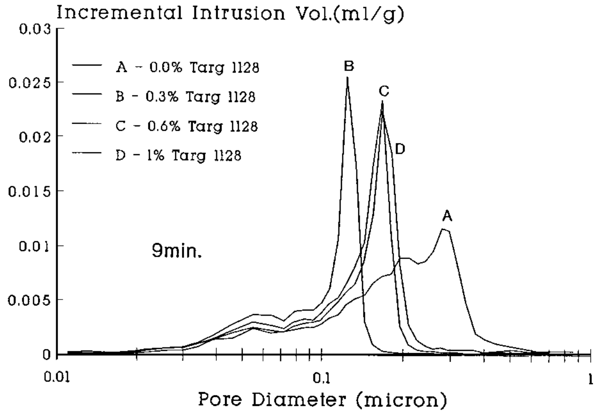

Fig. 6. Influence of the amount of dispersant on pore size distributions of samples cast for 9 min.

4. Summary and conclusions

The data discussed in this work allows the following conclusions to be drawn:

1. Both the particle size distribution and the interparticle potentials play dominant roles in governing the body formation rate, the green packing density, and the microstructure of the pressure cast bodies.
2. To achieve the highest packing densities, well-dispersed slurries and a bimodal particle size distribution in which finer particles can fill the interstitial pore spaces between the coarser ones, according to the Furnas model, are required. However, the packing efficiency obtained by pressure casting these slurries is normally lower when compared with slip casting. This is due to a faster deposition rate and an insufficient time allowed for "spontaneous" particle rearrangements when an external pressure is applied.
3. The amount of dispersant necessary to achieve the maximum green density in pressure casting was nominally 2–3 times greater than that required for a density via the slip casting process.
4. The cake permeability and the green density are nominally dependent upon casting time due to a decrease in the body formation rate and due to the longer time allowed for particle rearrangements.

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