Journal of the European Ceramic Society 18 (1998) 1171–1175

PII: S0955-2219(98)00039-9
0955-2219/98/$—see front matter

Effect of Driving Force on Pressure Slip Casting of Silicon Carbide Bodies

J. M. F. Ferreira* and H. M. M. Diz

Department of Ceramics and Glass Engineering, University of Aveiro, 3800 Aveiro, Portugal

Abstract

Binary silicon carbide powder mixtures with the particle size distribution designed to maximise the particles packing density were dispersed in aqueous medium in the absence and in the presence of different amounts of a deflocculant. The solid loading was fixed at 70 wt% and correspond to the conditions that most facilitate homogeneously and high packed structures to be obtained. The influence of the amount of deflocculant and applied pressure on the kinetics of the pressure slip casting process and on the microstructure of the green bodies was investigated. It was found that the pressure promotes a slightly increasing trend of green density of bodies obtained from suspensions with no or low amounts of deflocculant. The structures obtained from well-stabilised slurries are pressure independent. However, the maximum packing densities were always lower than that obtained by slip casting and required higher concentrations of deflocculant. Further, for a given applied pressure, it was found that the green density was time dependent. This results could be interpreted in terms of different particle rearrangement mechanisms. © 1998 Elsevier Science Limited. All rights reserved

1 Introduction

Silicon carbide has been one of the most promising high performance ceramic materials for various structural, electrical and high temperature applications.[1–3] The structure and the properties of ceramic components depend directly on the density, the uniformity of microstructure, and the rheological properties of the green compacts from which they are formed. These properties can be improved by using colloidal consolidation techniques.[4,5] Our previous works[6,7] showed that homogeneous and very high green density (> 76%) silicon carbide bodies could be obtained by slip casting slurries in which the solid loading, the amount of deflocculant and the particle size distribution have been optimised. The use of bimodal particle size distributions and well-dispersed slurries lead to particle segregation for solid contents lower than the optimum level. These phenomena can be prevented by using high solid loading and/or less stable suspensions[6] or, alternatively, by using an external pressure to increase the consolidation rate.[8] This makes the driving force of the casting process an important factor for achieving a high degree of homogeneity. The optimal amount of deflocculant for the pressure casting process has not been already determined. However, our previous results suggest that the amount required to maximise the green density within pressure casting will be higher in comparison with slip casting.[8]

The aim of this work was to evaluate the influence of the amount of deflocculant, the casting time and the applied pressure upon the kinetics of the pressure casting process and on the characteristics of the green silicon carbide bodies obtained.

2 Experimental Procedure

2.1 Materials and reagents

The starting materials were two commercially available SiC powders, NF0 and 1200 P (Elektroschmelzw Kempten, GmbH, Germany), with mean diameters of 1·2 and 13 μm, respectively. The deflocculant used was an ammonium salt of a low molecular weight polycarbonate acid (Targon 1128, Benkiser–Knapsack GMBH, Germany).

2.2 Slip preparation and characterisation

The fine and coarse powders were combined in the proportion 45/55 that gave the highest packing density.[6–8] The suspensions were prepared by first adding the fine powder to distilled water or to solutions having different concentrations (from

---

*To whom correspondence should be addressed.

0·03 to 0·3 wt%) of Targon 1128, and hand stirring, followed by simultaneous stirring and ultrasonication for 10 min. The coarser component was then added and the simultaneous stirring and ultrasonication continued for a further 10 min. A solid content of 70 wt% was used since it corresponds to the solid loading that promoted the highest packing densities (75·5 %) by slip casting from the same powder's suspensions.[6]

The electrophoretic mobility (EPM) of the fine powder particles was measured at 20°C using a microscope-type electrophoresis equipment (Mark II, Rank Brothers, Cambridge, UK) with a planar silica cell and blacked platinum electrodes. The ionic strength of the liquid medium was adjusted to 0·001 M NaCl. The rheological properties of the finer powder suspensions were evaluated with a cone-plate viscometer (Ferranti–Shieley, Ferranty Ltd, Manchester, UK) coupled to an automatic data acquisition system previously described.[3] Measurements involving the coarse powder were not performed due to the possibility of the coarse particles damaging the instrument. The flow curves (yield stress versus shear rate) were built up by collecting 1000 points in both branches (up and down). Shear rate varied from 0 to 1176·8 s^{-1} in a sweep time of 10 s. The pseudoplastic behaviour observed was analysed under the Bingham approximation using the linear part (intermediate 900 points) of the down curve. The yield stress and the plastic viscosity were calculated, respectively, from the intercept and the slope of the straight line with the shear stress axis.

2.3 Preparation and characterisation of the green cakes

The pressure slip casting experiments were performed in an apparatus designed by the authors as previously described.[9] Green cakes were weighed and placed in a stove at 120°C for 24 h. After complete drying, samples were cooled in an desiccator to avoid water uptake from the atmosphere and then reweighed. Subsequently, the green density was measured on a mercury balance.

3 Results and discussion

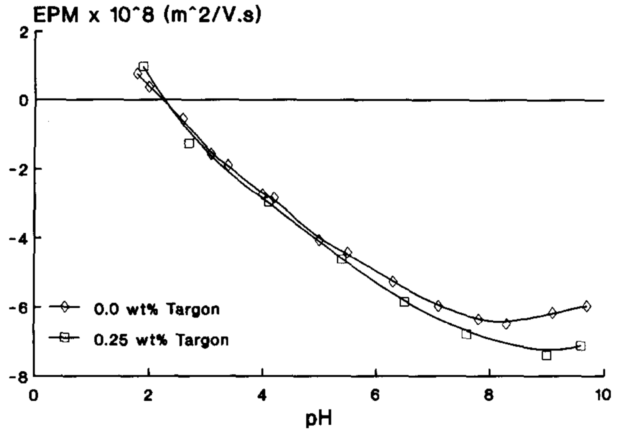

Fig. 1. Electrophoretic mobility of the finer powder particles in absence and in the presence of 0·3 wt% Targon 1128.

3.1 Slurry characterisation

The electrophoretic mobility of the fine SiC particles measured in the absence and in the presence of 0·3% Targon 1128 is plotted in Fig. 1 as a function of pH. In both cases, the isoelectric point (IEP) was located at about pH 2. This is in good agreement with values reported in the literature for the silicon carbide.[10,11] At the IEP, electrostatic repulsions between the suspended particles are absent, and the particle's interactions are governed by the attractive Van der Walls forces. This explains why the yield stress and the plastic viscosity present a maxima at the IEP (Fig. 2). With the pH increasing, electrical charge develops onto the surface of the particles giving rise to repulsive double layer forces between them. As a consequence, the Bingham yield stress and plastic viscosity decrease. The presence of Targon 1128, as a dispersing agent, had a negligible influence on the EPM values measured up to about pH 5, but caused gradual and significant decreases in Bingham yield stress and plastic viscosity. For a question of clarity, other curves correspondent to some intermediate dispersant concentrations (0·01, 0·03, 0·05 and 0·20%) were not represented in Fig. 2. For pH $\geq$ 5 the presence of the dispersant gradually improved the EPM but its effect on rheology was less remarkable. These results suggest that Targon 1128 can act through different stabilising mechanisms. In the acid region the polycarbonate groups adsorbed at particles surface should exist in the undissociated form, exerting a steric effect which is responsible for the observed decrease in yield stress and plastic

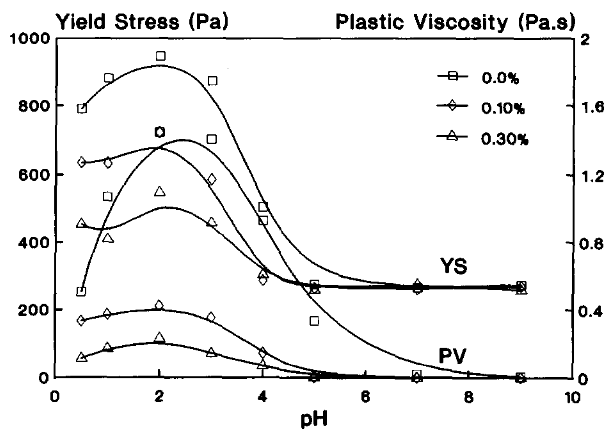

Fig. 2. Effects of the amount of dispersant and pH on the yield stress (YS) and plastic viscosity (PV) of the finer powder slurries.

viscosity. In the basic pH region the functional groups of the polyelectrolyte dissociate. The negative charge of these dissociated groups is added to the surface charge of the particles, enhancing its EPM. This improvement of the surface charge of the particles is not significantly reflected on rheology, because the electrostatic repulsions between the bare particles in the basic pH region are enough to stabilise the suspensions.

3.2 Pressure slip casting

The effects of applied pressure and the amount of deflocculant upon the green density and wall thickness for a casting time of 2 min are presented in Figs 3 and 4, respectively. It can be observed that the density is only slightly pressure dependent for the less stable slurries (0·0 and 0·03% dispersant), and practically independent of the applied pressure for the most stable slurries. The packing degree is mainly dependent upon the amount of dispersant added. These results are consistent with observations made by Fennely and Reed[12,13] and Lange and Miller[14] who found that dispersed slurries produce much higher packing densities relative to flocked slurries. In addition, Lange and Miller found that the packing density of the dispersed slurries was relatively pressure insensitive, whereas, the packing density of the flocked slurries was very pressure sensitive. This last point differs from the present observations.

The higher green densities are observed at 0·3 wt% Targon 1128. This amount of deflocculant is 2–3 times more than that required (0·1–0·15 wt%)[6] for maximising the green density in the conventional slip casting process from the same suspensions. This difference might be due to the higher shear rates that will result when an external pressure is added to the suction pressure of the plaster moulds. 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 greater amount of polyelectrolyte at particle's surface will be necessary to lubricate the particle contacts and promote particle rearrangements.

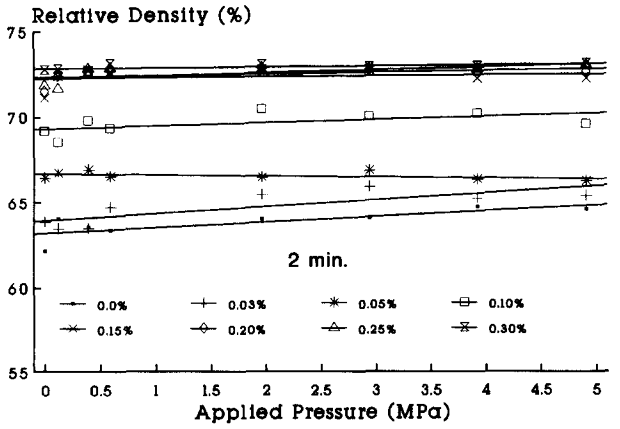

Fig. 3. Effect of applied pressure and amount of dispersant on green density.

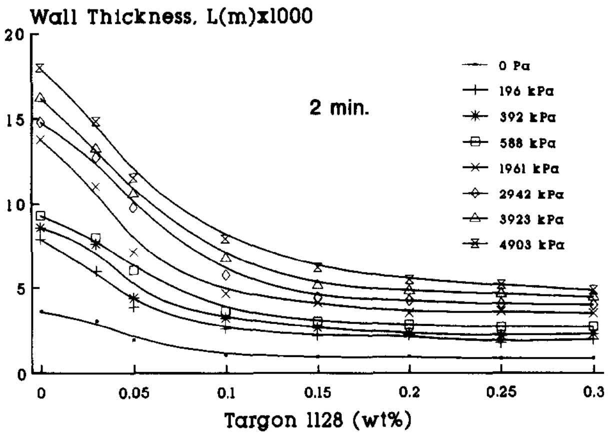

Fig. 4. Effect of applied pressure and amount of dispersant on wall thickness, *L*.

Concerning the thickness, *L*, Fig. 4 shows that it strongly depends on both experimental variables. For a given pressure, well-dispersed slurries form less permeable cakes that offer a higher resistance to the fluid flow and the rate of built up at the wall decreases. The dependance of the body formation rate (wall thickness squared and divided by the elapsed time, *t*) upon the driving force appears obvious from eqn (1):[9]

$$L_2/t = 2K\phi P/\eta(\phi_c - \phi) \qquad (1)$$

where P is the applied pressure, η is the liquid viscosity and ϕ and ϕ_c are the volume fractions of particles within the slurry and cake, respectively. The permeability, K, is inversely related to the resistance to fluid flow through the consolidated layer. This equation assumes unidirectional filtration, negligible mould resistance, constant rheological properties of the suspension, absence of particle settling, and constant permeability, that is, incompressible body.

In practice, cakes are more or less compressible, depending upon the interaction forces between particles,[12–14] pressure level,[15] and particle size distributions.[8] Cakes obtained from well-dispersed slurries and/or having particle size distributions with high packing ability are usually almost incompressible.[9] Some authors[9–15] have suggested that permeability is a function of the effective pressure expressed as

$$K = K_0 P^{-s} \qquad (2)$$

where s is the coefficient of compressibility. If $s = 0$, the cake is incompressible ($K = K_0$); if $s = 1$, the body is totally compressible. With the binary particle size distribution used in this work, designed to maximise the packing density, the coefficients of compressibility were very small ($s = 0{\cdot}12$ and $s = 0{\cdot}03$ for 0·0% and 0·05% Targon 1128, respectively). According to this, all pressure casting data were treated assuming incompressible cakes.

The effect of casting time on the green density was evaluated for suspensions dispersed with 0·15 and 0·30 wt% Targon 1128. The results plotted in Fig. 5 show an increase of relative density (ϕ_c) with increasing casting time. As a consequence, the consolidated layer will offer an incremental resistance to the fluid flow, relative to that expected from eqn (1) that assumes a constant permeability. As a consequence, the deposition rate will be significantly reduced.

However, even for the longer casting time used, the relative density achieved is still ≈ 2% lower compared to that obtained in normal slip casting from the same suspensions. This difference may be explained as follows. Particles within well-dispersed slurries can flow and pack as individuals. Since the particles still repel each other as they join the consolidation layer, they experience rearranging movements before being fixed. These rearranging movements require time to occur. They will be favoured in slip casting process in which the rate of deposition is relatively low. 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 rearranging movements of one particle might now be restricted by new incoming particles and a decrease in the packing density should be expected.[8]

The evolution of the cake thickness with casting time for the two chosen dispersant concentrations is typical of a controlled diffusion process through an increasing wall thickness as described by eqn (1). However, the curves (not shown) do not start at the origin of the axis. This disharmony might be related with the set-up time, t_0 (the time required to fill the pressure chamber and to adjust the pressure level, which was usually about 0·1 min), and also with the permeability variation with increasing casting time.[9] The plots log L^2 versus log P_t where P_t is the total pressure (suction pressure of the mould + applied pressure), for suspensions containing 0·15 and 0·30 wt% Targon 1128 are shown in Fig. 6. For a question of clarity, data collected at 0·3 wt% Targon 1128 and for the shortest casting times (0·5 and 1 min) were not presented. The correlation coefficients of the straight lines represented are very high (≈1). However, these lines do not pass through the origin of the axis due to the set-up time and permeability variations.[9]

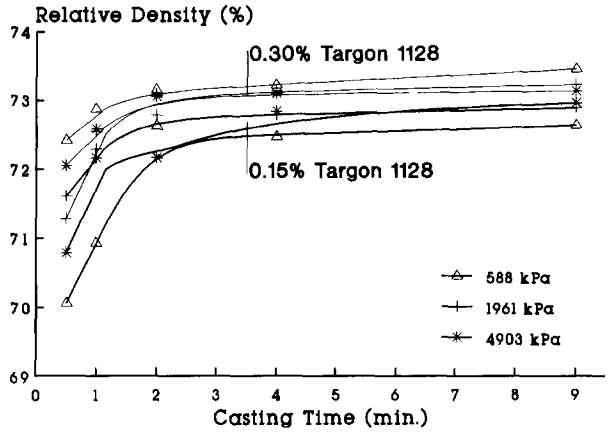

Fig. 5. Effect of casting time and applied pressure on packing density of green cakes prepared from suspensions containing 0·15 wt% and 0·30 wt% Targon 1128.

According to eqn (1), the slopes of the lines in Fig. 6 represent the ratios

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

which may be related to the average permeabilities of the cakes. These calculated average permeabilities are presented in Table 1. It can be seen that the permeabilities are lower at 0·30 than at 0·15 wt% Targon 1128. This is according to the results shown in Figs 3 and 4. A decreasing tendency of the permeability with increasing casting time can also be observed at both dispersant concentrations. The combined effects of the permeability variations and the set-up time will result in a decrease of the slopes of the straight lines shown in Fig. 6, as demonstrated in our previous work.[9] For the shorter casting times the effect of set-up time is more important. As a consequence, the calculated permeability values might result under-evaluated.

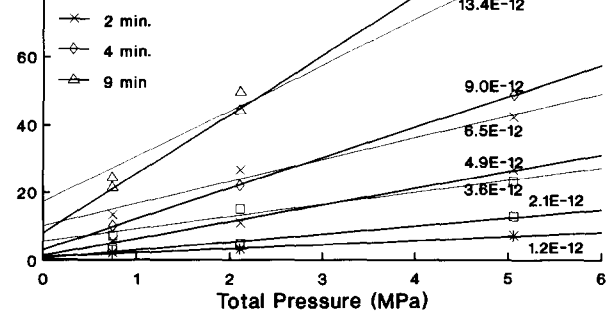

Fig. 6. Effect of total pressure and casting time on wall thickness for suspensions containing 0·15 wt% (thick lines) and 0·30 wt% (thin lines) Targon 1128.

Table 1. Effects of casting time and amount of dispersant on cake permeabilities

Targon 1128 (wt%)	*Casting time (min)*	*Permeability* $(m^2) \times 10^{16}$
0·15	0·5	8·4
	1	8·3
	2	8·2
	4	8·1
	9	7·1
0·30	0·5	7·5
	1	6·4
	2	6·5
	4	5·9
	9	5·4

4 Summary and Conclusions

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

1. Interparticle potentials play a dominant role in governing the body formation rate and the green packing density of the formed green bodies. To achieve high packing densities, strong repulsive interparticle potentials are required, which can be controlled by suspension pH and further improved by adding a suitable dispersant.
2. The amount of dispersant required to achieve maximum packing densities in pressure casting were 2–3 times those used in the conventional slip casting process. Nevertheless, the relative densities obtained by pressure casting were normally more than 2% lower in comparison with slip casting.
3. The driving force of the pressure slip casting process has only a minor influence on cake permeability, which seems to decrease slightly with the increments in this variable. Contrarily, permeability was found to be very sensitive regarding to the casting time. This is consistent with the concomitant increase observed in green density. This last effect as well as the set-up time effect contribute to a decrease in body formation rate.

References

1. Takeda, Y., Development of high-thermal-conductive SiC ceramics. *J. Am. Ceram. Soc. Bull.*, 1988, **67**, 1961–1963.
2. Kahlman, L., Rundgren, K., Lidén, E., Nyberg, B. and Carlstrom, E., Processing of liquid phase sintered silicon carbide—mechanical and wear properties. In *Third Euro-Ceramics*, P. Duran and J. F. Fernandez, (Eds.), Vol. 1. Faenza Editrice Ibéraci,Castellón de la Plana, Spain, 1993, pp.168–176.
3. Ferreira, J. M. F. and Diz, H. M. M., Effect of the slurry structure on the slip casting of silicon carbide powders. *Journal of the European Ceramic Society*, 1992, **10**, 59–64.
4. Roosen, A. and Bowen, H. K., Influence of various consolidation techniques on the green microstructures and sintering behaviour of alumina powders. *J. Am. Ceram. Soc.*, 1988, **71**, 970–977.
5. Lange, F. F., Powder processing science and technology for increased reliability. *J. Am. Ceram. Soc.*, 1989, **72**, 3–15.
6. Ferreira, J. M. F. and Diz, H. M. M., Effect of solid loading on slip casting performance of silicon carbide slurries. *J. Am. Ceram. Soc.*, submitted.
7. Ferreira, J. M. F. and Diz, H. M. M., Effect of the amount of deflocculant and powder size distribution on the green properties of silicon carbide bodies obtained by slip casting. *J. of Hard Materials*, 1992, **3**, 17–27.
8. Ferreira, J. M. F., A interface carboneto de silício–Solução aquosa e o enchimento por barbotina. Ph.D. thesis, University of Aveiro, 1992.
9. Ferreira, J. M. F. and Diz, H. M. M., Effect of pH on pressure slip casting of silicon carbide bodies. *Journal of the European Ceramic Society*, 1997, **17**, 259–266.
10. Rizzo, C., Angelis, B. A. D., Ferretti, E. and Howlett, S. P., Slip casting of α-silicon carbide. Materials Science Monographs, 66B. In *Proceedings of the International Meeting on Modern Ceramics Technologies*, 7th CIMTEC–World Ceramics Congress, ed. P. Vincenzini, Elsevier, Amsterdam, 1991, pp. 1095–1102.
11. Ferreira, J. M. F. and Diz, H. M. M., Effect of aging time on pressure slip casting of silicon carbide bodies. *Journal of the European Ceramic Society*, 1997, **17**, 333–337.
12. Fennelly, T. J. and Reed, J. S., Mechanics of pressure slip casting. *J. Am. Ceram. Soc.*, 1972, **55**, 264–268.
13. Fennelly, T. J. and Reed, J. S., Compression permeability of Al_2O_3. *J. Am. Ceram. Soc.*, 1972, **55**, 381–383.
14. Lange, F. F. and Miller, K. T., Pressure filtration: consolidation kinetics and mechanics. *Am. Ceram. Soc. Bull.*, 1987, **66**, 1498–1504.
15. Frassek, L. and Hennicke, H. W., Some aspects of pressure slip casting with nonclay suspensions. *Ceram. Forum Intern.*, 1990, **67**, 443–446.