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THE INFLUENCE OF THE GRANULE STRUCTURE ON THE STRENGTH OF PRESSED AND SINTERED Si_3N_4

L. Carlinger and E. Carlström, Swedish Ceramic Institute, Sweden

ABSTRACT

Several studies have shown that hard agglomerates limit the uniform consolidation of green compacts (1), leaving large pores in the sintered bodies. This paper deals with the effects of the powder content and of the stability of the slip on the microstructure of the granules. By using freeze granulation (spray freezing followed by freeze drying) it has been possible to focus on the properties of the slip vs. the properties of the granules, without the added effects of migration and shrinkage during drying (as in spray drying).

INTRODUCTION

Freeze-granulation is a method of spray freezing with subsequent freeze drying, see Figure 1.

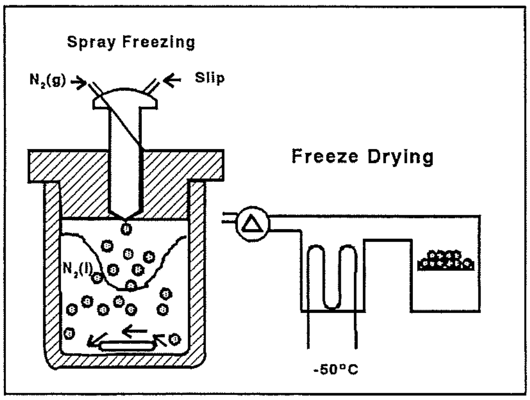

Figure 1: The freeze granulation technique.

Compared with spray-drying (2) this granulation technique gives free-flowing, spherical granules with an homogeneous additive distribution, giving green bodies without large pores after pressing. The granule density and the granule strength can be varied by changing the powder content of the slip and there is no shrinkage involved as the water removal occurs through a sublimation (no liquid transport as in spray drying). The variation of the granule density can be done without affecting the slip stability which makes the method suitable in this study.

EXPERIMENTAL PROCEDURE

A commercial Si_3N_4 powder (P95H, Permascand AB) was mixed and dispersed in water with 3 wt% of Y_2O_3 (HC Starck, fine) and varying amounts (0.02- 0.2 wt%) of dispersant (D-3021, Rohm & Haas) by a planetary mill (200 rpm, 50 min.). Binders (4 wt%; 5 Latex:1 PEG, DM 765s, Hoechst : PEG 400, Kebo) were added and the slurries were stirred for 2 h by a propeller. The solid loading (S_L) was varied between 35 and 45 vol. % and the pH for each slurry was carefully measured. The flocculation behaviour was obtained by varying the amount of dispersant. Rheological properties (constant shear rate, yield stress and oscillation stress sweep) were used to control and characterise the different slurries, before and after the addition of binder, using a rotational rheometer, (StressTech, ReoLogica Instr. AB). The slurries have been freeze granulated under nearly identical operating conditions using a two-fluid nozzle. The granules were screened (45-250 μm), in different sieves to maintain a constant size distribution, and the compaction behaviour and the green strength (diametrical compression of green compacts, same density) were tested in a universal testing machine. Finally the granule surface, the bulk structure of the granules and the fracture surface of the green compacts were analysed in a SEM.

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RESULTS AND DISCUSIONS

Earlier studies based on spray drying (1,3) have indicated that a flocculated slurry contributes to the elimination of large pores in the green compacts. By varying the slip stability, see Table I, and by using freeze granulation it has been possible to both study the effect of a looser granular structure and the effect of flocculation on the granule itself (forming hard agglomerates in the granules).

Table I: Summary of rheological studies

Slurry	Dispersed [1]high S_L	Flocculated [2]high S_L	Flocculated [3]low S_L
Viscosity (at 1 s^{-1})	230 s^{-1}	650 s^{-1}	33 s^{-1}
Thixotropy (loop test)	Low or none	Medium	High

[1] 45 vol%, C_D=0.02 wt% [2] 45 vol%, C_D=0.2 wt% [3] 35 vol%, C_D=0.2 wt%

The granules prepared from the flocculated slurries, regardless of solid loading, have a more inhomogeneous pore structure (open porosity) than the granules prepared from the dispersed slurries. There is no difference in fracture, the fracture surfaces of the green bodies have the same appearance in both the flocculated and the dispersed case (same number of unfractured granules). All measurements, see Table II, indicate that the best results, the highest green strength, a transgranular fracture and the best compaction behaviour are obtained with the slurry with the lowest solid loading (lower granule density) .

Table II: Summary of microstructure and compaction studies

Slurry parameters	Compaction density (150 MPa)	Green strength fracture load[4] (N)	Structure granulate (pore distribution)	Fracture unfractured granules
Dispersed high S_L	1.86 g/cm^3	0.058	homogeneous	few granules
Flocced high S_L	1.91 g/cm^3	0.052	inhomogeneous	few granules
Flocced low S_L	1.83 g/cm^3	0.076	inhomogeneous	no granules

[4] Same green density (1.8 g/cm^3)

As the freeze granulation was performed under nearly identical operating conditions, but the slurries had different viscosity, a variation in size distribution was seen. To keep the size distribution constant the granulates were screened in different sieves (45-250 μm). Analysis of compaction and green strength, Table II, were performed on samples with the same mixture of fractions (same weight percent of each fraction 45-250 was added).

CONCLUSIONS

Dispersion studies and controlled flocculation of slurries with different solid loading show that the granule and green properties are effected by the looser granule structure obtained by the lower solid loading and not as much by the flocculation itself. Further studies of sintered properties and new slurries stabilised or flocculated are in progress.

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