

# MICROSTRUCTURAL DEVELOPMENT AND MECHANICAL PROPERTIES OF PRESSURELESS SINTERED $\text{Al}_2\text{O}_3/\text{SiC}$ COMPOSITES

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## ABSTRACT

$\text{Al}_2\text{O}_3$  green bodies containing 5 vol.% SiC were prepared by slip casting. In order to obtain stable slips, the zeta potential of the SiC was adapted by chemical modification of the surface using functional silanes (e.g. amino ethyl amino propyl trimethoxy silane). Another route for the preparation of homogeneous slips was developed by heterocoagulation of  $\text{Al}_2\text{O}_3$  and SiC under well controlled pH conditions and by stabilizing of the heterocoagulated system with a non-ionic surfactant. Both routes led to homogeneous aqueous slips which could be slip-casted to green compacts with densities around 60 % of theory showing an excellent distribution of the SiC within the  $\text{Al}_2\text{O}_3$  matrix. Green compacts were pressureless sintered at 1800 °C to densities above 98.5 % showing a homogeneous microstructure with average grain sizes between 2 and 2.5  $\mu\text{m}$ , a fracture strength of about 650 MPa and fracture toughness around 3.0  $\text{MPa} \cdot \text{m}^{0.5}$ .

## INTRODUCTION

The improvement of the mechanical properties of alumina ceramics by inert second phases like SiC particles or platelets is a promising alternative for the more common whisker reinforcement. It is reported that strength and fracture toughness of alumina can be increased by the inclusion of fine SiC particles [1,2,3]. The improvement of the fracture strength in  $\text{Al}_2\text{O}_3/\text{SiC}$  composites is traced back to flaw healing, machining induced surface compressive stresses and the fine grained microstructure of alumina matrix. The grain refinement has been attributed to a decreased grain boundary mobility and retarded grain growth of alumina caused by SiC particles [4]. The enhancement of the fracture toughness is explained by the interaction between propagating cracks and stress fields around the SiC inclusions. The formation of stress fields around the SiC particles is based on the differences in thermal expansion of SiC and  $\text{Al}_2\text{O}_3$  ( $\alpha_{\text{SiC}} = 4.6 \times 10^{-6} \text{K}^{-1}$ ,  $\alpha_{\text{Al}_2\text{O}_3} = 8.5 \times 10^{-6} \text{K}^{-1}$ ) which causes radial compressive stresses around SiC particles upon cooling.

Beside the amount and size of the SiC particles, their distribution within the  $\text{Al}_2\text{O}_3$  matrix is an important factor for the development of microstructures and mechanical properties. Therefore, homogenization is carried out by wet milling the powders in a liquid using an appropriate

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dispersing agent. The milling technique commonly used [2] can not provide the desired homogeneity, especially if the size of SiC in the range to < 100 nm. Furthermore, it is rather difficult to obtain an ideal particle distribution in  $\text{Al}_2\text{O}_3/\text{SiC}$  slurries because the surface properties of both compounds differ strongly from each other. From this point of view, it is of interest to develop processing techniques which allow a proper control of homogeneity in this two-component system. One approach is the utilization of heterocoagulation of the two materials in a suspension at a certain pH [5] and preparing a well-dispersed slurry in the presence of a non-ionic surfactant. Another possibility is to adapt the surface chemistry of both compounds to each other in order to obtain an ideal distribution of separated particles in the suspension under the same dispersing conditions [6]. Both concepts have been used for the preparation of alumina/SiC composites and have been tested with respect to the achievable microstructures as well as mechanical properties.

#### EXPERIMENTAL PROCEDURES

The starting powders were commercial aluminum oxide (CS 400M, Martinswerk,  $d_{50} = 400$  nm) and  $\alpha$ -silicon carbide (UF45, H. C. Starck,  $d_{50} = 120$  nm). The SiC powder was used as received and after surface modification with aminoethyl trimethoxysilane, respectively. The details for the surface modification of SiC are given elsewhere [6]. Polydispersed aqueous slurries were prepared from surface-modified SiC (5 vol.%) and alumina (95 vol.%) by attrition milling in the presence of a polymeric acid. Heterocoagulated powders were obtained by dispersing alumina and SiC separately in water and then combining both suspensions at pH = 4.0. The precipitate (30 vol.%) was redispersed by adding 2 wt.% of a non-ionic polymer and ultrasonic agitation for 15 minutes. Green compacts were formed by slip casting and were then pressureless sintered at 1800 °C for 60 minutes. Slurries were characterized with respect to viscosity, pH dependence of zeta potential and particle size distribution using standard techniques. Microstructural characterization of sintered samples was carried out on polished and etched cross sections as well as on fracture surfaces by SEM. For the determination of the 3-point bending strength, samples with an average size of  $3 \times 3 \times 25$  mm<sup>3</sup> were used. Preliminary results of the fracture toughness was obtained after Indentation-Crack-Length-Method (ICL) by using the equation of Anstis [7].

#### RESULTS AND DISCUSSION

One important prerequisite for the utilization of the above described concepts is the knowledge of the chemical surface properties of the starting powders. These properties were determined by measuring the pH dependence of the zeta potential. Figure 1 displays the zeta potential curves of the as received powders as well as the curve of SiC modified with aminoethyl aminopropyl trimethoxysilane. The as received SiC powder shows the already described zeta potential curve with an isoelectrical point around pH = 3 and negatively charged particles for pH > 3 with high zeta potentials only above pH = 8. In contrast, a completely different behavior was found for the modified SiC powder. The isoelectrical point is shifted to pH = 9 and maximum positive zeta potentials are obtained at pH < 9. This indicates a similar surface chemistry as for the alumina from zeta potential measurement, and one can assume that a well-dispersed slurry of both compounds can be prepared for pH values below 5. On the other hand, for heterocoagulation of the as received alumina and SiC the pH range between 4 and 5 seems to be suitable because the powders are oppositely charged in this range. Following these assumptions, aqueous  $\text{Al}_2\text{O}_3/\text{SiC}$  slurries with a solid content of 30 vol.% were prepared and characterized with respect to rheological properties and particle size distribution. In Figure 2, the

shear stress/shear rate curves of the two different slips are shown. Both suspension possess a nearly ideal Newtonian flow behavior for shear rates larger than  $40 \text{ s}^{-1}$ , indicating a good state of dispersion. This could be further confirmed by particle size measurements which gave particle

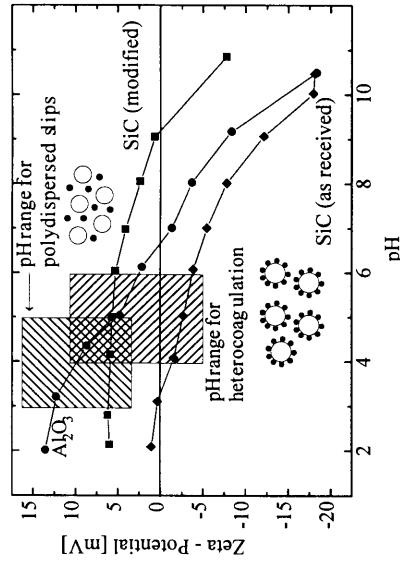


Fig. 1: Zeta-potential of the different powders as a function of pH (●: SiC, ○:  $\text{Al}_2\text{O}_3$ ).

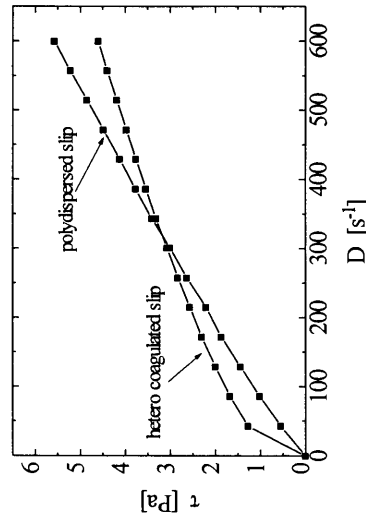


Fig. 2: Shear stress vs. shear rate for aqueous  $\text{Al}_2\text{O}_3/\text{SiC}$  (5 vol.%) slurries. a) slurry prepared from heterocoagulated powder (30 vol.%); b) polydispersed slurry (35 vol.%).

size distributions in range of 200 to 500 nm for the heterocoagulated system. In contrast, for the polydispersed slurry a bimodal distribution ranging from 70 - 200 and 200 - 500 nm was found.

From these slurries green compacts were slip casted in porous gypsum moulds. Compacts prepared from heterocoagulated slurries exhibit a density of 56 to 58 % of theory whereas specimens obtained from the polydispersed systems reached a slightly higher density (58 to 60 % of theory). Determination of the pore size distribution reveals for both preparation techniques a homogeneous pore structure ranging from 40 to 70 nm with an average pore diameter of about 59 nm. Sintering of the green specimens was performed in flowing nitrogen at 1800 °C for one hour, and in both cases a final density above 98.5 % was achieved.

In Figure 3 the microstructure of the  $\text{Al}_2\text{O}_3/\text{SiC}$  composites prepared from the heterocoagulated slurry is compared with the microstructures of an alumina sample. The sample was prepared by slip casting from the starting powder and sintered under the same conditions as the composites in order to ensure the data for the composites.

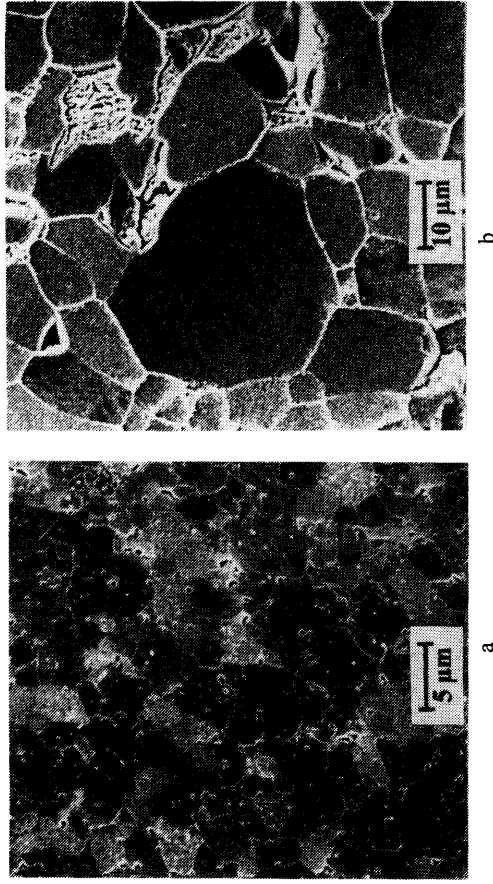


Fig. 3: Microstructure of alumina with 5 vol-% SiC (3a) and monolithic alumina (3b) after sintering at 1800 °C for 60 min.

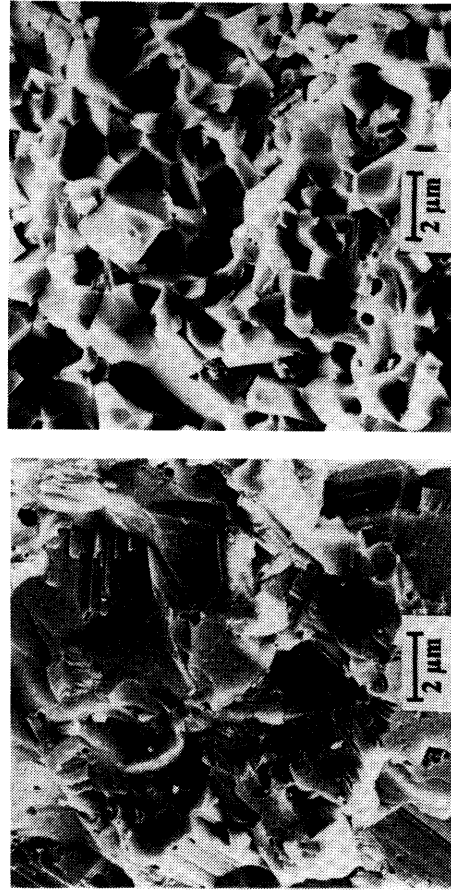


Fig. 4: Fracture surface of alumina with 5 vol-% SiC sintered 1800 °C for 60 min and monolithic alumina of same grain size and density sintered at 1400 °C for 60 min (4b)

Samples containing 5 vol.% SiC are characterized by a homogeneous fine grained microstructure with an average grain size of 2.4 µm, as determined by the linear intercept method. The SiC particles with an average size of ≈100 nm are homogeneously distributed in the alumina matrix.

It is evident that SiC particles are mainly located within the alumina grains and to a less extend at the grain boundaries. Similar microstructures were obtained for composites prepared from the polydispersed slurry. In contrast, the monolithic alumina sintered under same conditions exhibits a much coarser and heterogeneous microstructure with an average grain size of about 17 µm (Fig. 3b). The average fracture strength of the composite was 650 MPa. The comparison of the fracture strength of monolithic alumina and composite samples with similar microstructure showed that the fracture strength of composite was much higher than that of the alumina (300 MPa). In addition, detailed investigations of the fracture surface by SEM showed that the fracture of the composite mainly occurs via an intragranular fracture mode whereas the fracture mode of the alumina is predominantly intergranular as shown in Fig. 4.

Thermal expansion anisotropy of alumina causes in polycrystalline monolithic alumina stresses between grains of different orientation leading to an intergranular fracture mode. The observed intragranular fracture mode for composite samples indicates that these stresses can be compensated by present SiC inclusions. As a result of this, the grain boundary strength of the alumina is significantly increased resulting in an increased fracture strength of the composite. In opposite to the fracture strength, toughness of the composite does not differ very much from monolithic alumina. Values between 2.9 and 3.2 MPa.m<sup>0.5</sup> were measured for both materials.

#### CONCLUSION

The results show that homogeneous two component slips can be prepared from starting powders with different surface chemical properties like alumina and silicon carbide by following the concept of heterocoagulated powder and polydispersed systems in combination with chemical surface modification. Green compacts with an excellent homogeneity can be prepared by common slip casting techniques, enabling the fabrication of complex shaped products. The homogeneity of the green bodies allows, in turn, the production of sintered parts with enhanced material properties. The presented concept for slip preparation is not limited to the  $\text{Al}_2\text{O}_3/\text{SiC}$  system and can be applied to every multicomponent system.

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