

# RELATIONS BETWEEN MICROSTRUCTURE AND MECHANICAL PROPERTIES OF PRESSURELESS SINTERED SiC

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## ABSTRACT

SiC green bodies with B<sub>4</sub>C and nano-scale carbon black as sintering aids were prepared by slip casting. After presintering at 2160 °C for 10 min the samples were annealed at the same temperature for different times in order to obtain different microstructures. By this procedure the initial relative density of 0.981 was increased to 0.989 accompanied by grain growth and morphological changes. Samples annealed for 30 min possessed a fracture strength around 440 MPa, as determined by the double ring test. The maximum fracture toughness was 3.5 MPa.m<sup>0.5</sup>. Examination of the grain growth behaviour showed that the microstructure is up to relative densities of 0.985 stable against the accelerated grain growth.

## INTRODUCTION

As a mainly covalently bonded material SiC can be pressureless sintered by solid state diffusion only in the presence of sintering additives like combination of B/C, Al/C or B<sub>4</sub>C/C [1, 2]. An important parameter is the additive distribution, especially carbon black, because this strongly influences densification, microstructural development and at least material properties. According to [3], excellent carbon distributions can be obtained by precipitating of nano-scale carbon particles directly on the top of sub-micron SiC powders. It has also been reported that pressureless sintering leads to high density compacts with promising mechanical properties. However, in order to exploit the full potential of this novel processing technique, further investigations concerning e.g. relationships between densification behavior, microstructure development and mechanical properties are needed. First results will be reported in this paper.

## EXPERIMENTS

Powders used for experiments were  $\alpha$ -SiC (mean particle size 290 nm, BET-surface 14 m<sup>2</sup>/g, supplied by ESK, FRG), B<sub>4</sub>C (mean particle size 245 nm, BET-surface 18 m<sup>2</sup>/g, supplied by ESK, FRG) and carbon black (mean particle size 20 nm, BET-surface 300 m<sup>2</sup>/g, supplied by BASF, FRG). Aqueous SiC slips containing 2 wt.-% carbon black, 0.65 wt.-% B<sub>4</sub>C and an overall solid content of 35 vol.-% were prepared from SiC powder electrostatically coated with nano-scaled carbon black, as described in [3]. Disc-shaped green bodies (diameter 57 mm, thickness 5.5 mm) were prepared by slip casting. Sintering was carried out in a graphite lined high temperature furnace under flowing argon. After sintering the samples were ground and polished on one side and used for the determination of fracture strength by the ring-on-ring method

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(diameter upper support 40 mm, load ring 8 mm). For microstructural characterization sintered samples were embedded in epoxy resin. After grinding and polishing they were etched with boiling Murakami's solution. The average grain size of samples was determined by the linear intercept method. The elasticity modulus was indirectly determined by measuring of the ultrasonic velocity by the pulse-echo method. The fracture toughness was determined by the Intendation-Crack-Length method [4].

## RESULTS AND DISCUSSION

Prior to isothermal annealing experiments the sintering temperature was determined at which samples reach a relative density above 0.98. Pressureless sintering at 2160 °C for 10 min resulted in samples with a density of 3.14 g/cm<sup>3</sup> corresponding to a relative density of 0.981 (theoretical density of the present composition was 3.20 g/cm<sup>3</sup>). Based on these results, the samples were isothermally annealed at 2160 °C for different times. According to Fig. 1, the initial density of 0.981 % increases continuously up to densities of 0.989 % with decreasing densification rates for annealing times exceeding 30 min.

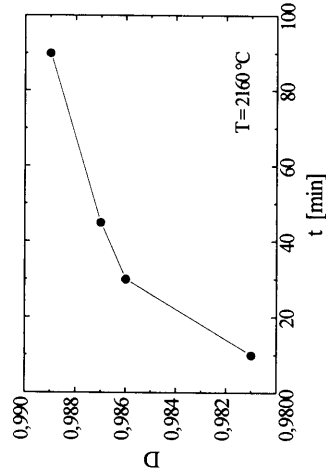
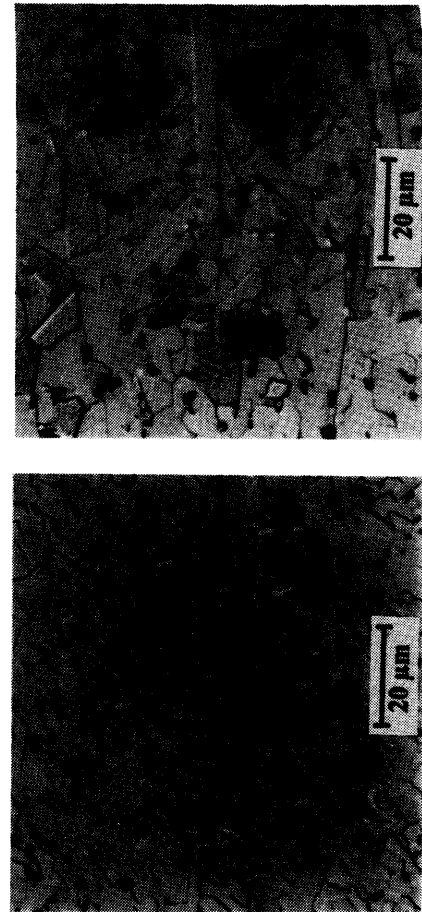


Fig. 1: Relative density of SiC samples after sintering at 2160 °C for different times.

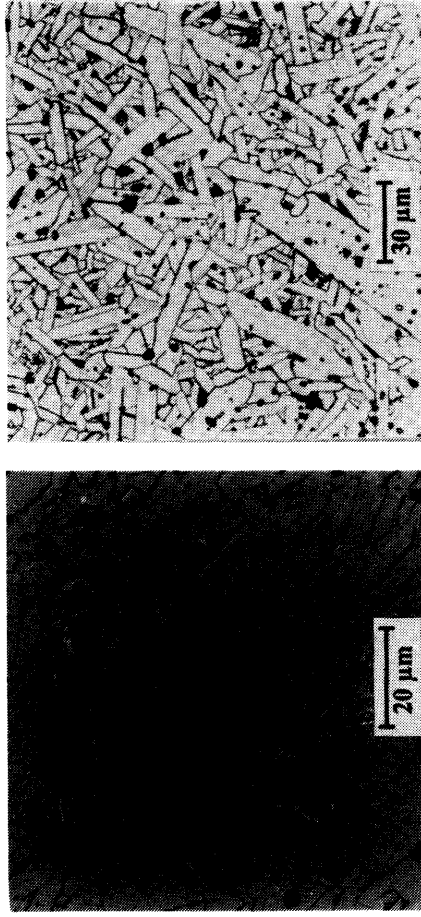
The microstructural evolution of these samples is shown in Fig. 2. A sample sintered for 10 min shows predominantly equiaxed grains with an average size of 4.6 μm (Fig. 2a). The density of this sample was 0.981 %. The residual pores, appearing as black dots, are at grain junctions. Further annealing (30 min) causes grain growth (average grain size 5.2 μm) and changes in the grain morphology.

Grains with an increased aspect ratio of 2-3 were found. After an annealing time of 45min the grains became more and more elongated (Fig. 2c).



2a: 2160 °C, 10 min

2b: 2160 °C, 30 min



2c: 2160 °C, 45 min

2d: 2160 °C, 90 min

The average grain size of this sample with a density of 0.987 % was 6 μm. Prolonged annealing times, e. g. 90 min, cause dramatic changes of the microstructure without further densification (Fig. 2d). The microstructure mainly consists of large SiC platelets. The length of these platelets ranges between 15 and 105 μm. A small percentage of equiaxed grains are located between the large grains. The linear intercept method gave an average grain size of 10 μm for this sample. But the use of this method was not sufficient for this microstructure. In this case the average aspect ratio represents a better measure for microstructure description. The aspect ratio was determined to be 9 which is typical for a platelet-like morphology.

For a given system the knowledge about the grain growth behaviour is essential and presents a base line for tailoring the microstructure by post heat treatments. For this reason the grain size/density plot in Fig. 3 was constructed. The curve indicates that for relative densities up to 0.985 a negligible grain growth takes places. An accelerated grain growth starts at relative densities above 0.985 with an onset corresponding to the onset of the formation of elongated grains as described before. The stability of the microstructure up to such high densities was surprising and can be explained in different ways. One explanation is the decreased grain boundary mobility by the precipitation of boron containing phases at the grain boundaries [5]. As reported in the literature, the solubility of boron in SiC at temperatures above 2000 °C is ≈ 0.5 wt.-% [6] corresponding to the boron content of sintered samples (0.65 wt.-% B<sub>4</sub>C). Therefore, the retarding of grain growth may be only possible by the presenting of boron containing phases which can be formed as a result of local concentration gradients and should disappear after certain time. On the other hand, it is well known that excess carbon in SiC can retard the grain growth [7, 8]. Since the sintered green bodies contain homogeneously distributed nano scaled carbon the retardation of grain growth can be traced back to the effect of presenting residual carbon particles. The microstructural stability up to such high relative densities can then be explained by the high degree of homogeneity introduced by slip preparation and green body formation.

For the determination of fracture toughness after Anstis [4] the exact knowledge about the E-modulus is necessary which depends for a given material strongly on the porosity. Therefore,

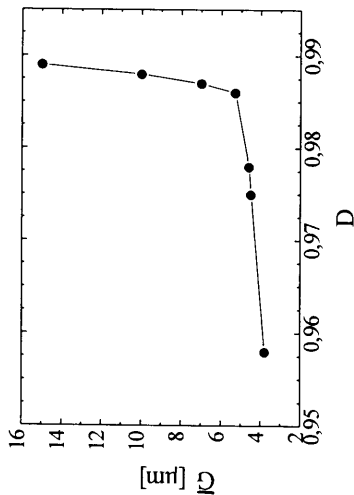


Fig.3: Grain size vs. relative density plot for pressureless sintered SiC.

Young's-moduli were measured for samples with porosities between 6 to 11%. It was found that the Young's-Modulus decreased linearly with the increasing porosity and can be described by a relation of the form

$$E = 445.6 - 11.28 P \quad [\text{GPa}]$$

with P = porosity in %

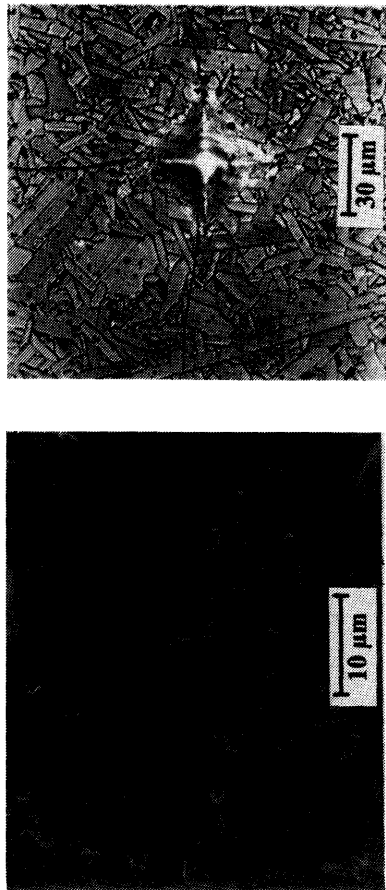
This equation was used for the determination of Young's modulus for compacts with other density values, and the data were then used for the calculation of fracture toughness following the ICL method. The material data as well as microstructural parameters of samples annealed at 2160 °C for different times are summarized in Table 1.

t [min]	D	G [μm]	$\sigma_b$ [MPa]	$H_{0.5}$ [GPa]	$K_{IC}$ [MPa·m <sup>0.5</sup> ]
10	0.981	4.6	367±44	25.1	3.1±0.10
30	0.986	5.2	443±10	26.8	3.5±0.15
45	0.987	5.8	405±10	26.9	3.0±0.20
90	0.985	9.0	410±27	27.2	2.4±0.10

Table 1: Microstructural parameters and mechanical data of SiC samples annealed at 2160°C for different times.

The initial fracture strength of samples (ring-on-ring method) sintered at 2160 °C for 10 min was 367 MPa which increases significantly to 443 MPa by post annealing for 30 min. For longer annealing times a slight decrease of the strength (405 MPa) was determined. For the fracture toughness,  $K_{IC}$ , a similar behaviour was observed with increasing annealing times. The maximum value  $K_{IC}$  is 3.5 MPa·m<sup>0.5</sup> which was determined on samples annealed for 30 min. Prolonged annealing times caused a significant decrease of fracture toughness. Considering the proposed toughening mechanisms for ceramics, the results obtained for fracture toughness are unexpected. A platelet-like grain morphology (Fig. 2d) should lead to a high fracture toughness by, for example, crack deflection.

The observation of fracture surface of sintered SiC samples indicated the fracture mode to be transgranular as shown in fig. 4 on a sample with microstructural parameters similar for the sample in Fig. 2b (D = 0.986, G = 5.2 μm). This result was confirmed by the observation of cracks produced by Vickers indentation. As shown in Fig. 4b, the propagating cracks present for all impinge angle no deflection, even on large SiC platelets. An improvement of the fracture toughness by crack deflection does not seem to be possible for the crack propagation mode. Therefore, another mechanisms should be responsible for the observed increase of the fracture toughness after intermediate annealing times and the significant decrease after long annealing time. One possibility may be the effect of local stress fields caused by the anisotropy of thermal expansion coefficient of SiC between different crystallographic directions.



4a: Fracture surface  
4b: Crack propagation  
Fig. 4: Fracture surface of a SiC sample with a relative density of 0.987 (a) and crack propagation in a coarse grained SiC sample (sample from Fig. 2d)

## CONCLUSION

It could be proved that the homogeneous distribution of carbon black throughout the green microstructure is a key parameter for the fabrication of pressureless sintered SiC parts with good properties. The high sintering densities above 98.5% and the retarded grain growth up to high densities (98.9%) provide new possibilities for microstructure tailoring by post-sintering heat treatment and consequently for tailoring material properties.

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