Ceramics International **21** (1995) 297–301

The Influence of Processing Parameters on Microstructure and Mechanical Properties of SiC–TiC_p Ceramics

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(Received 2 November 1994; accepted 28 February 1995)

Abstract: SiC–TiC_p ceramics are considered as potential materials for high temperature composites. The influence of various processing parameters during the fabrication of hot pressed SiC–TiC_p composites on microstructure and the resulting flexural strength and fracture toughness will be discussed.

INTRODUCTION

For the fabrication of high-density silicon carbide, submicron powders and boron plus carbon sintering additives are necessary.[1,2] The use of aluminium or aluminium compounds as sintering aids has also been reported.[3,4] In general the sintering activity of ceramic composite compositions is retarded due to the presence of different phases. Therefore the hot-pressing technique has been used. Wei and Becher[5] have investigated additions of titanium carbide to silicon carbide and reported an improvement in the mechanical properties with increasing titanium carbide concentration. Janney[6] has investigated the influence of different carbon sources which were used together with aluminium as sintering additives. The addition of high surface area carbon black resulted in lower mechanical strength than in the case where carbon was incorporated in the form of a phenolic resin. The Weibull modulus however was higher for the samples containing carbon black. Hahn *et al.*[7] hot-pressed SiC–30 wt%TiC composites with additions of aluminium and carbon and also noticed improvement in the mechanical properties in comparison with silicon carbide. The flexural strength increased from 350 to 400 MPa, fracture toughness from 4 to 5 MPa $m^{1/2}$. The values remained constant up to 800°C. At higher temperatures a decrease in the mechanical strength was observed and was attributed to oxidation phenomena. In general, the thermal shock resistance of the composite material improved.

Since it can be expected that processing parameters will have a significant influence on the resulting mechanical properties, the influence of various processing steps has been analyzed in this investigation.

EXPERIMENTAL

Powder characteristics

Silicon carbide powder (Grade A10, H.C. Starck, Goslar) and titanium carbide (Grade A, H.C. Starck, Goslar) were used. The chemical composition of the powders and their particle size distribution are shown in Tables 1 and 2.

Powder processing

For the preparation of 500 g powder, 25 ml of an aluminium alkoxide solution (30–35 % Al $(OC_3H_7)_3$ in isopropanol (supplier Alfa Ventron)) was mixed with 1000 ml ethanol. The silicon carbide powder was dispersed by ultrasonic and magnetic stirring for 20 min. Then the titanium carbide powder was added and dispersion continued for an additional 40 min. For further homogenization a planetary ball mill (SiC-container and

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Table 1. Chemical composition of powders (wt%)

Element	Silicon carbide	Titanium carbide
C(ges.)	31·0	19·5
O	0·62	n.d.
N	n.d.	0·067
Fe	0·023	0·06
B	0·021	n.d.
Cr	0·001	0·027
Ca	0·002	<0·001
Si	n.d.	<0·1
W	n.d.	0·72
Ni	n.d.	0·003
Na	n.d.	0·002

Table 2. Particle size distribution of powders (μm)

	Silicon carbide	Titanium carbide
d_{10}	0·3	0·9
d_{50}	1·0	3·0
d_{90}	2·9	5·8

*Laser Diffraction (Helos Sympatec), Disp.: H_2O, 5 min. US

SiC milling media) was used. The suspension was passed through a 63 μm sieve and dried in a rotating evaporator. After addition of 3 ml ethanol per 100 g of powder, the powder was granulated and cold pressed in a steel die with two subsequent pressure steps (13 MPa and 26 MPa). The compacted discs (70 mm diameter, 10 mm high) were dried at 80°C before hot-pressing.

Sintering

The pre-pressed specimens were hot-pressed under argon in BN-coated graphite dies. BN-coated 0·3 mm graphite foils were located between the plungers and the sample. The hot-pressing temperature was 2000°C, the applied pressure 33 MPa, and the time at maximum temperature 30 min. The heating and cooling rates were 15 and 10 K min^{-1}, respectively. In Fig. 1 a typical hot-pressing cycle is shown.

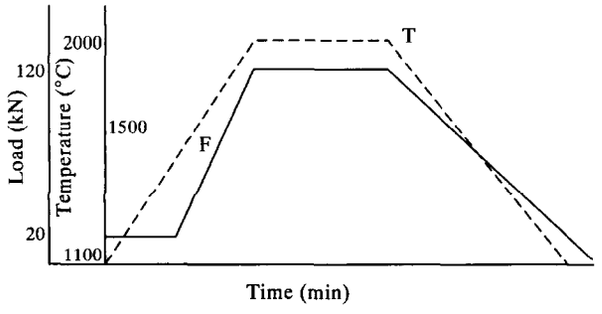

Fig. 1. Hot-pressing cycle.

Fracture test

The hot pressed discs were glued on a ceramic plate, rough ground on both sides and fine ground with a lapping wheel using boron carbide powder (Tetrabor, F320, Elektroschmelzwerk Kempten). Samples for mechanical tests (4·5 × 3·3 × 45 mm^3) were cut from the discs with diamond wheels and were tested in 4-point bending (20/40 mm). The fracture toughness was determined on notched samples (cooper wheel, 0·05 mm thickness, notch depth 1–1·3 mm). The mechanical test fixture was made of stainless steel for the RT-measurements and of silicon carbide for the HT-measurements. The loading rate was 35·5 Ns^{-1}. The high-temperature values were obtained after equilibrating the samples for 20 min at the test temperature.

RESULTS AND DISCUSSION

Mechanical properties of SiC and SiC–TiC

After optimization of different processing parameters the RT flexural strength values shown in Fig. 2 for HP-SiC, HP-TiC and HP-SiC–50TiC have been obtained. The TiC powder was hot pressed without any previous processing; the samples had densities of 97·4% th.d. The SiC powder was processed in a similar way to the SiC–50TiC powder mixtures except that 0·8 wt% boron (Grade 11, H.C. Starck) and 1.5 wt% carbon (Elftex, Cabot) were used as sintering additives. After hot-pressing

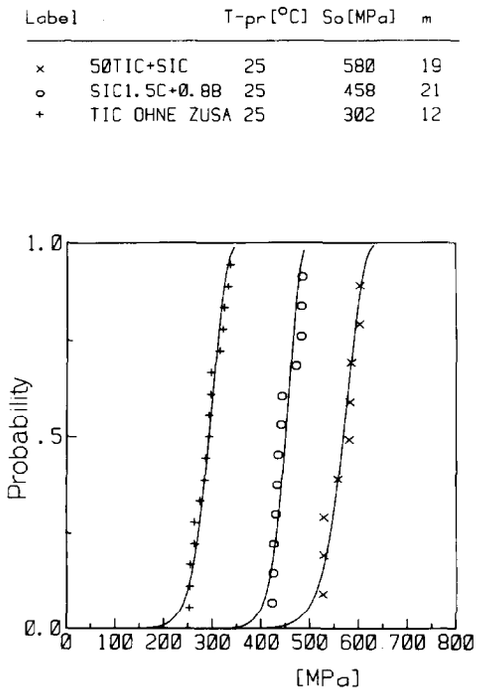

Fig. 2. Flexural strength (RT) for HP-SiC, HP-TiC and HP-SiC–50TiC.

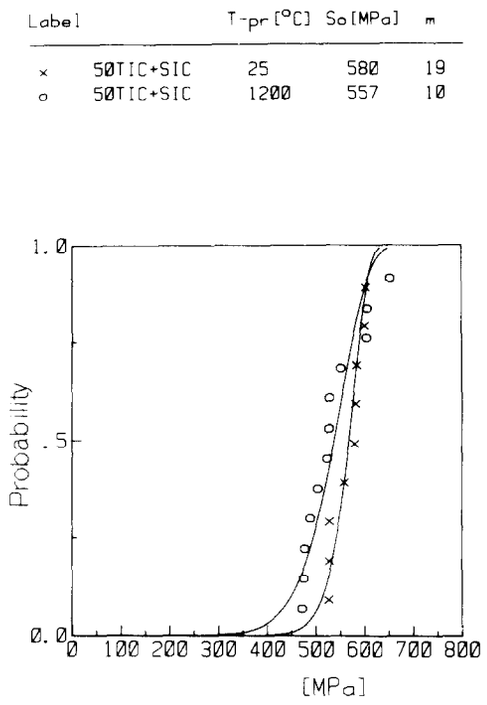

Fig. 3. Flexural strength (RT/1200°C) for HP-SiC–50TiC.

at 2100°C for 20 min, 99·2 % dense samples were obtained. The SiC–50TiC samples were fabricated as described above. The average flexural strength is 302 MPa for TiC, 458 MPa for SiC and 580 MPa for the composite material. The scatter of the strength values is rather small (high Weibull modulus). The fracture toughness is 4·1 MPa $m^{1/2}$ for TiC, 3·7 MPa $m^{1/2}$ for SiC and 5·6 MPa $m^{1/2}$ for the SiC–50TiC composite. The SiC–TiC composite shows an improvement, both in flexural strength and in fracture toughness, in comparison to the pure components. The mechanical data do not change very much at 1200°C, the flexural strength is 560 MPa, the fracture toughness has increased slightly to 6·5 MPa $m^{1/2}$ (Fig. 3). The improvement in the strength and toughness data is probably caused by the presence of internal stresses due to the different expansion coefficients of SiC ($\alpha_{25\text{-}1000}$ =4·0 × 10^{-6} K^{-1}) and TiC ($\alpha_{25\text{-}1000}$ =8·1 × 10^{-6} K^{-1}). The presence of tangential compressive forces in the matrix close to the particle could result in a crack deflection and could improve the fracture toughness.

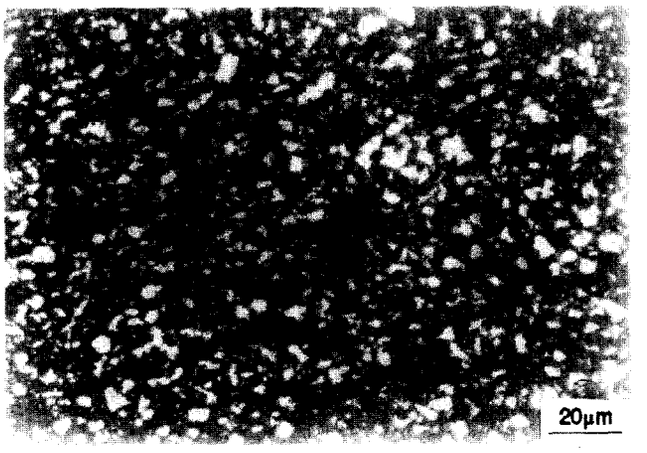

Fig. 4. Microstructure of SiC–30TiC (white phase TiC).

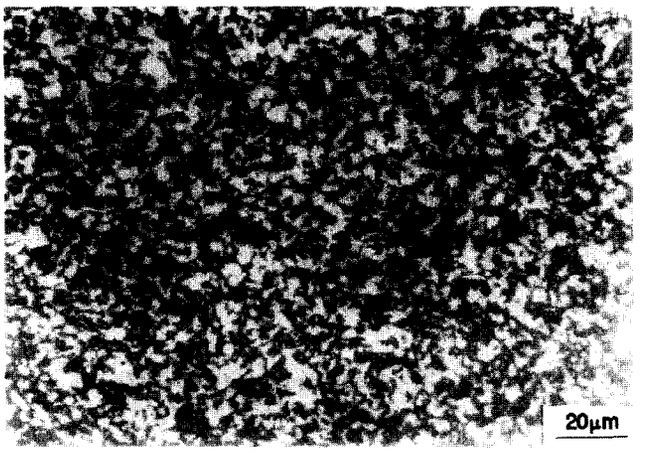

Fig. 5. Microstructure of SiC–50TiC (white phase TiC).

Influence of processing parameters on microstructure and mechanical properties

(a) Silicon carbide–titanium carbide ratio. The influence of the SiC–TiC ratio has been studied with compositions of SiC–30TiC and SiC–50TiC. The mechanical properties are summarized in Table 3, and the microstructures are shown in Figs 4 and 5. The dependence of the mechanical strength and the fracture toughness on the concentration of titanium carbide is evident and can be explained by the presence of higher internal matrix stresses in the case of SiC–50TiC.

Table 3. Influence of SiC–TiC ratio on mechanical properties

Composition (wt%)			Density (% th. d.)	σ_B (MPa)	K_{IC} (MPa $m^{1/2}$)	m (Weibull modulus)
SiC	TiC	Al_2O_3				
69·3	29·7	1·0	>99	500	5·2	15
49·5	49·5	1·0	>99	580	5·6	19

(b) Concentration of sintering additive. Al-isopropoxide as a 30–35% solution in isopropanol has been used as a sintering additive resulting in a very homogeneous distribution in the powder mixture. The influence of a variation in the isopropoxide concentration (nominal concentration of Al_2O_3 after decomposition of the alkoxide would have been 0·5 and 1·0%) is shown in Table 4. The amount of 0·5% aluminium oxide is too small to achieve sufficient densification. The samples have a sintering density of only 95% th.d. and the mechanical strength is reduced due to the presence of porosity. An addition of carbon did not result in an improvement of the microstructure. However, it must be taken into consideration that a certain amount of carbon or carbon compound is introduced by the decomposition of the isopropoxide and that in the titanium carbide 0·2 wt% free carbon is present according to the information of the supplier.

(c) Homogenization. In order to investigate the influence of homogenization, the SiC–50TiC composition was homogenized in a planetary mill particle for a period between 10 and 240 min. The particle size distribution after milling did not show a significant change after the different homogenization cycles (Table 5). The mechanical strength of the samples is shown in Fig.6, the sintering densities and the fracture toughness are indicated in Table 6. The increasing homogeneity with

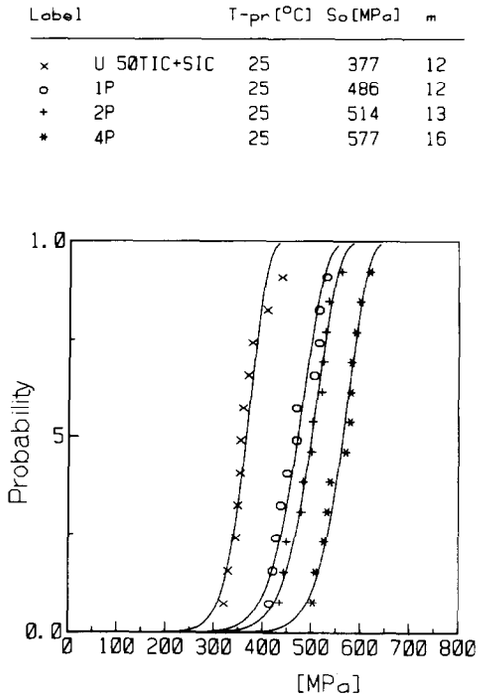

Fig. 6. Influence of homogenization on flexural strength for SiC–50TiC.

Table 4. Flexural strength of SiC–TiC composites with different concentrations of sintering additives.

Composition (wt%)			Density (% th. d.)	σ_B (MPa)	m (Weibull modulus)
SiC	TiC	Al_2O_3			
49·75	49·75	0·5	95·6	415	9
49·50	49·50	1·0	>99	514	13

Table 5. Particle size distribution of a powder mixture (49·5% SiC, 49·5% TiC, 1 % Al_2O_3) after different homogenization

Sample	Homogenization		d_{10}	d_{50}	d_{90}
	Method	Duration (min)		(μm)	
U	Ultrasonic	10	0·5	1·7	4·5
1P	Planetary mill	60	0·5	1·7	4·4
2P	Planetary mill	120	0·5	1·5	4·2
4P	Planetary mill	240	0·5	1·6	4·0

Table 6. Fracture toughness of SiC–TiC composites (49·5% SiC, 49·5% TiC, 1 % Al_2O_3) after different homogenization

Sample	Homogenization		Density	K_{IC}
	Method	Duration (min)	(% th. d.)	(MPam$^{1/2}$)
U	Ultrasonic	10	96·4	4·2
1P	Planetary mill	60	>99	5·2
2P	Planetary mill	120	>99	5·4
4P	Planetary mill	240	>99	5·6

increasing time is reflected by an increase in the Weibull modulus and is accompanied by higher flexural strength and a sligthly enhanced fracture toughness. An ultrasonic homogenization for 10 min is not sufficient and results in a lower sintering density and a decrease in mechanical strength (sample U). Large pores (Fig. 7) and TiC agglomerates (Fig. 8) which are still present in the microstructure are responsible for this result. Even after a homogenization time of two hours (sample 2 P) small pores are still present in the microstructure (Fig.9). A homogenization time of four hours is necessary in order to obtain a pore-free microstructure.

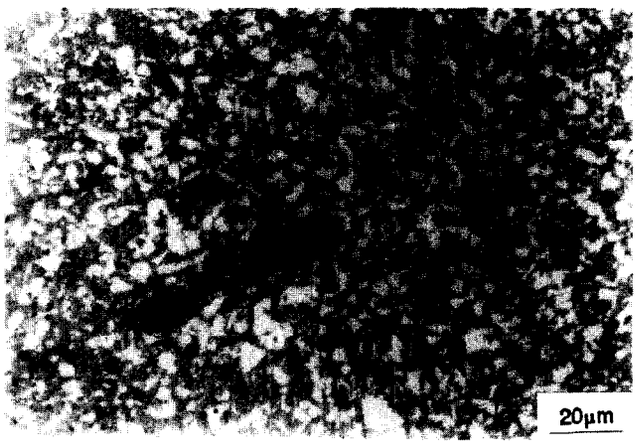

Fig. 7. Large pores in SiC–50TiC (sample U).

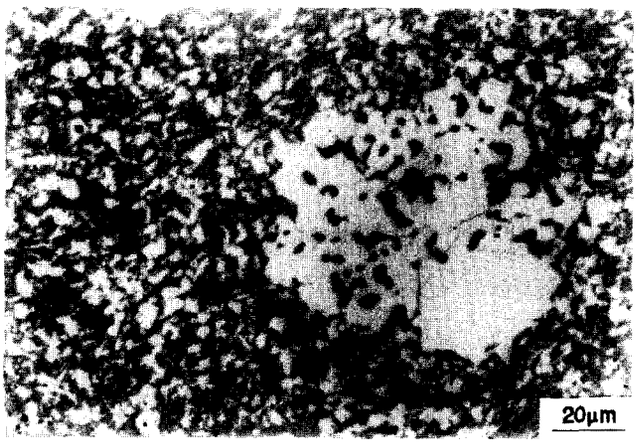

Fig. 8. TiC agglomerates in SiC–50TiC (sample U).

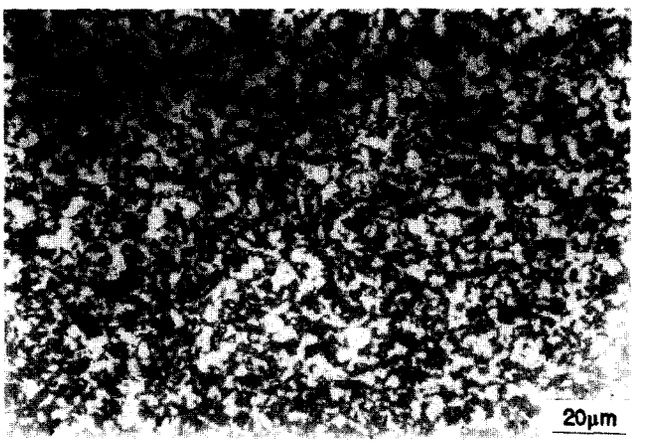

Fig. 9. Small pores in SiC–50TiC (sample 2P).

CONCLUSIONS

Besides a high sintering density a homogeneous distribution of the second phase is important to achieve high strength and fracture toughness in SiC–TiC_p composites, together with high Weibull moduli. The processing parameters must be optimized, so that full use of the material can be made.

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