

# Synthesis of Silicon Carbide Ceramics Using Chemically Modified Polycarbosilanes as a Compaction Binder

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A chemically modified polycarbosilane (PC) containing organofluorine groups (PCOCF) has been synthesized from PC and fluoroalkylmethyldimethoxysilane. PCOCF acts as an efficient compaction binder for SiC powders and as a coating material with excellent oxidation resistance in wet air. PCOCF-coated SiC powders also show excellent packing properties because of the organofluorine side chains, which give highly dense green compacts. PCOCF provides a high ceramic yield of 75% and highly dense SiC ceramics. Four-point bending strength increases and the scatter in strength values decreases significantly by PCOCF coating.

## I. Introduction

SILICON CARBIDE (SiC) is well-known for its high thermal and chemical stability, high strength, hardness, and thermal conductivity. Recently, the development of SiC-based ceramic materials has become active because of the pyrolysis of silicon-based polymeric precursors.<sup>1-3</sup> This polymeric precursor route has some important advantages with respect to conventional techniques: (1) the solubility and fluidity of the polymeric precursors afford potential routes for the preparation of SiC fibers<sup>4-6</sup> and coatings;<sup>7</sup> and (2) ceramics can be designed with specific properties using the polymeric precursors as binders.<sup>8,9</sup>

Polycarbosilane (PC) is the most typical polymeric precursor for SiC-based ceramics and can be chemically modified to yield binary ceramic systems, such as SiC-TiC,<sup>10,11</sup> SiC-ZrC,<sup>12</sup> and Si-Al-C-O systems.<sup>13</sup> If a polymeric SiC precursor is applied as a compaction binder for SiC ceramic powder compacts, the precursor-derived functions are introduced into SiC ceramic powder and powder compacts, leading to high-quality, monolithic SiC ceramics. Such a functional binder can be flexibly designed by chemical modification techniques.

Iwamoto *et al.*<sup>14</sup> successfully synthesized chemically modified PC, containing fluoroalkoxy groups, from a commercially available PC and fluoroalkyl alcohol. Iwamoto *et al.* selected a new PC with high molecular weight and fluoroalkylmethyldimethoxysilane as the starting materials to synthesize the chemi-

cally modified PC, PCOCF, with high ceramic yield. The PCOCF then was used as a compaction binder, because improvement was expected in the fluidity, the oxidation resistance, and the packing properties of SiC ceramic powder and in the mechanical properties of SiC sintered compacts.

This article describes (1) a study of the synthesis of PCOCF, (2) the effects of PCOCF coating on the properties of powders and green compacts, and (3) the mechanical properties of the sintered compact of PCOCF-coated SiC.

## II. Experimental Procedure

Figure 1 shows the synthetic route for PCOCF processing used in this study. Si-Cl bonds were introduced to a commercially available PC (10 g) (Type UH,  $M_n = 2561$ ; Nippon Carbon Co., Ltd., Tokyo, Japan) by refluxing with an excess amount of carbon tetrachloride ( $\text{CCl}_4$ ; reagent grade, Nakarai Chemical) in the presence of a catalytic amount of benzoyl peroxide (0.1 g) (BPO; reagent grade, Nakarai Chemical) for 24 h, followed by concentration in a rotary evaporator. The chlorinated PC (PCCl, 12.3 g) was dissolved in toluene and cooled to 0°C, and aqueous ammonia ( $\text{NH}_4\text{OH}$ , 6 mL) was added to this solution. The reaction mixture then was warmed to room temperature and stirred for 3 h to hydrolyze the Si-Cl bonds in the PCCl. The reaction mixture was neutralized by aqueous hydrogen chloride (HCl), followed by concentration in a rotary evaporator. The residue was suspended in water and

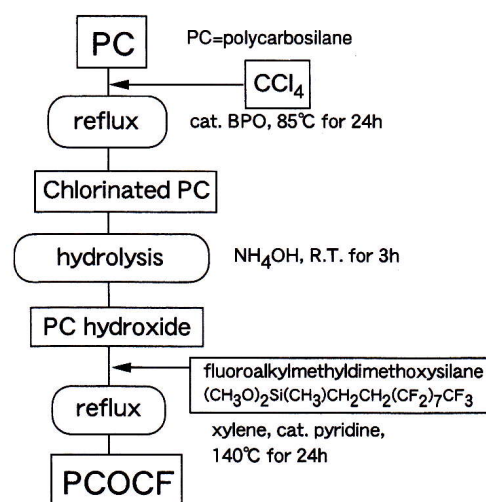


Fig. 1. Synthetic route for producing PCOCF.

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extracted with toluene. The organic extracts were washed with water, saturated aqueous sodium chloride solution, dried over anhydrous magnesium sulfate, and concentrated in a rotary evaporator to give polycarbosilane hydroxide (PCOH, 9.2 g). The synthesized PCOH (5.0 g) and fluoroalkylmethyltrimethoxysilane (3.0 g) ( $\text{CF}_3(\text{CF}_2)_7\text{CH}_2\text{CH}_2\text{Si}(\text{CH}_3)(\text{OCH}_3)_2$ ; TSL-8231, Toshiba, Tokyo, Japan) were dissolved into dry xylene (100 mL), and the mixture was refluxed at 140°C for 24 h in the presence of a catalytic amount of dry pyridine (1 mL) (reagent grade, Nakarai Chemical), followed by concentration in a rotary evaporator. The residue was washed using methanol to remove excess fluoroalkylmethyltrimethoxysilane and to give PCOCF (5.5 g). The nature of the organofluoric modification of PCOH was confirmed by  $^1\text{H}$  nuclear magnetic resonance ( $^1\text{H}$ -NMR; Model UNITYINOVA 300, Varian Japan, Ltd., Tokyo, Japan) spectra using chloroform- $d$  ( $\text{CDCl}_3$ ; Aldrich Chemical Co., Milwaukee, WI) as a solvent. Gel permeation chromatography (GPC, Model System-21, Shodex, Tokyo, Japan) analysis also was conducted to obtain the molecular-weight distribution of the modified PCOH. Thermogravimetric analysis (TGA, Model TG-DTA2000, Mac Science, Tokyo, Japan) was performed on the polymer samples up to 1000°C at a heating rate of 5°C/min under argon flow. To investigate the crystallization behavior above 1000°C, X-ray diffractometry (XRD; Model RINT 2500, Rigaku, Tokyo, Japan) measurements were performed on the heat-treated PCOCF with  $\text{CuK}\alpha$  radiation using an automated powder diffractometer equipped with a monochromator.

PCOCF-coated SiC powder was fabricated as follows. A calculated amount of toluene solution of PCOCF was mixed with SiC powder ( $\alpha$ -SiC; DU-A1, 0.47  $\mu\text{m}$ ; Showa Denko) and 0.5 mass% of boron as a sintering aid (0.85  $\mu\text{m}$ ; Mitsuwa Pure Chemicals), using a glass mortar for 1.5 h. Another sintering aid, carbon, was supplied from the PCOCF binder. The mixed slurry then was dried and sieved through a 60 mesh screen. In this way, 1, 3, 5, and 10 mass% PCOCF-coated SiC powders were prepared.

Oxidation resistance of the PCOCF-coated SiC powder was evaluated as follows. The PCOCF-coated SiC powders were stored in a sealed vessel containing liquid water (i.e., in wet air at room temperature). A comparable sample of the as-received SiC powder also was placed in the vessel. The oxygen content of each powder sample was measured by the inert-gas melting-thermal conductivity method as a function of holding time (equipment: Model EMGA-550, Horiba, Ltd., Tokyo, Japan; method: Japan Industrial Standard R-1603).  $\text{Si}_3\text{N}_4$  powder (JCRM R005; Ceramic Society of Japan, Tokyo, Japan) was used as the reference. The powder sample and nickel and tin bathing metal were melted in an inert-gas flow (argon) by impulse-type heating using a graphite crucible. Oxygen and nitrogen gases were extracted. Hydrogen and CO were moved from the gases, and they were sent to a heat conductivity analyzer with the inert gas. Oxygen content of each sample was calculated from the change of heat conductivity.

Cylindrical green compacts of the PCOCF-coated SiC powder were prepared by uniaxial pressing to pellets (15 mm in diameter and 3 mm in thickness) at a pressure of 30 MPa. PC, poly(vinyl alcohol) (PVA; SMR-10M, Shin-etsu Chemical Co., Ltd., Tokyo, Japan), and dextrin (Type 4-C, Nippon Starch Chemical Co., Ltd., Tokyo, Japan) also were coated onto the SiC powders, in a manner similar to the PCOCF coating, using ethanol or water solvent. Relative densities of the green compacts were evaluated as a function of binder addition. The green densities were calculated by measuring the volume and the weight of the green compacts. Relative density was calculated using the theoretical density of SiC (3.21  $\text{g}\cdot\text{cm}^{-3}$ ) and that of PCOCF or other binders (1.1  $\text{g}\cdot\text{cm}^{-3}$ ). The green compacts then were placed in a graphite die and heat-treated in a graphite resistance-heating furnace (Model High Multi 10000, Fujidempa Kogyo, Osaka, Japan) at 1500°C for 1 h, followed by hot pressing at 2050°C for 15 min at a hot-pressing pressure of 30 MPa under an argon atmosphere. The hot-pressing program

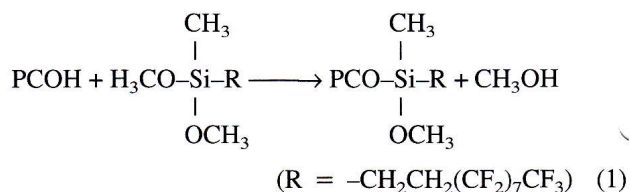
is shown in Fig. 2. Apparent density of hot-pressed and sintered compacts was measured by the Archimedes method. Four-point bending strengths were determined using a four-point rupture test on 3 mm  $\times$  4 mm  $\times$  40 mm bar samples whose surfaces and edges were polished with diamond powder (0.5  $\mu\text{m}$ ). The fracture toughness was determined by the single-edge precracked-beam (SEPB) method. Microstructure of the hot-pressed and sintered compacts was observed by scanning electron microscopy (SEM; Model S-800, Hitachi, Tokyo, Japan).

### III. Results and Discussion

#### (1) Chemical Structure of Precursors

Figure 3 shows the  $^1\text{H}$ -NMR spectra of polymerized samples and fluoroalkylmethyltrimethoxysilane. As-received PC presents a broad peak at 0.18 ppm with two shoulders at  $-0.08$  and  $-0.6$  ppm ( $\text{Si}-\text{CH}_2$ ,  $\text{Si}-\text{CH}_3$ , and  $\text{Si}-\text{CH}-\text{Si}$ , respectively) and another broad peak at 4.3 ppm ( $\text{Si}-\text{H}$ ). The ratio between the integrated intensities of C-H and Si-H is 1:0.09. In the PCOH spectrum, a new broad peak appears at 1.6 ppm assigned to  $\text{Si}-\text{OH}$  groups, and the relative peak intensity of Si-H groups decreases compared with that of as-received PC. The ratio of C-H:Si-H:Si-OH is 1:0.02:0.05. According to these spectroscopic analyses,  $\sim 56\%$  of the Si-H groups in the as-received PC have been successfully converted to Si-OH groups. However, the infrared spectrum of PCOH indicates that a few Si-Cl bonds remain in the PCOH.

Fluoroalkylmethyltrimethoxysilane displays four peaks at 0.16 ppm ( $\text{Si}-\text{CH}_3$ ), 0.82 and 2.10 ppm ( $\text{CH}_2$ ), and 3.54 ppm ( $\text{O}-\text{CH}_3$ ). The spectrum of PCOCF mainly consists of the peaks of PCOH and fluoroalkylmethyltrimethoxysilane. The peak due to  $\text{O}-\text{CH}_3$  groups at 3.54 ppm in the PCOH spectrum disappears, and new peaks due to  $\text{O}-\text{CH}_3$  appear at 3.40 and 3.50 ppm in the PCOCF spectrum. The peaks of  $\text{CH}_2$  groups in the PCOCF spectrum also become broader compared with those of fluoroalkylmethyltrimethoxysilane, indicating the influence of the polymer network. The appearance of the new peaks due to the  $\text{O}-\text{CH}_3$  group indicates that some of the  $\text{Si}-\text{OCH}_3$  groups of fluoroalkylmethyltrimethoxysilane are substituted by PCOH, as shown in Eq. (1).



One of the new peaks at 3.40 and 3.50 ppm is possibly due to the  $\text{O}-\text{CH}_3$  group in  $\text{PCO}-\text{CH}_3$ , which forms from the produced  $\text{CH}_3\text{OH}$  and unreacted PCCL.

Molecular-weight distributions of PCOCF, PCOH, and PC have been analyzed by the GPC method, as shown in Fig. 4.

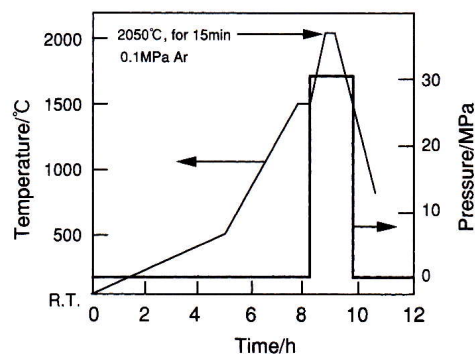


Fig. 2. Hot press sintering program.



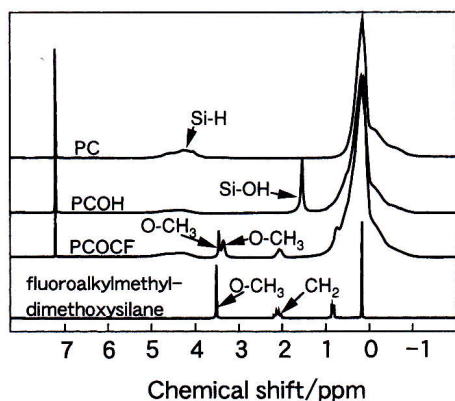


Fig. 3.  $^1\text{H}$ -NMR spectra of PC, PCOH, PCOCF, and fluoroalkylmethyldimethoxysilane.

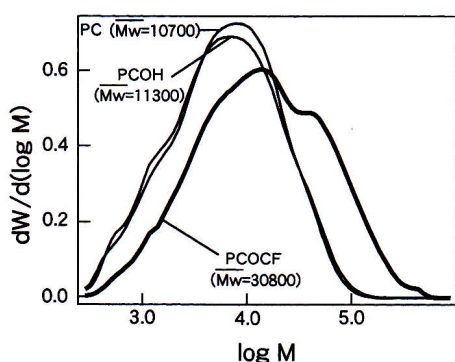
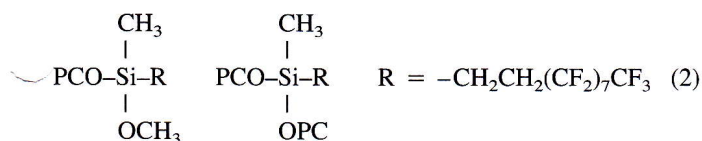


Fig. 4. Molecular-weight distribution of PC, PCOH, and PCOCF.

The mean value of  $M_w$  of each polymer sample also is given in Fig. 4. The  $M_w$  of PCOCF is almost 3 times larger than those of PC and PCOH. The larger  $M_w$  indicates that the two  $\text{OCH}_3$  sites of some of the fluoroalkylmethyldimethoxysilanes are fully substituted by PCOH. According to these chemical structural analyses, PCOCF most likely consists of the following functional groups:



The TGA curves of the as-received PC-, PCOCF-, and PCOCF-coated SiC powder are shown in Fig. 5. The weight loss of the as-received PC starts at  $100^\circ\text{C}$ , and a drastic weight loss begins at  $550^\circ\text{C}$  and ends at  $700^\circ\text{C}$ . The analyses were performed in a flowing argon atmosphere. The ceramic yield of the as-received PC is 85%. The weight loss of PCOCF starts at  $200^\circ\text{C}$  and ends at  $700^\circ\text{C}$ , again in argon. However, the large weight loss of PCOCF starts at  $400^\circ\text{C}$ , which is a relatively low temperature compared with that of the as-received PC. According to our previous study, decomposition of the organofluoric groups in PCOCF during pyrolysis causes the large weight loss of PCOCF at temperatures from  $400^\circ$  to  $700^\circ\text{C}$ .<sup>14</sup> The amount of conversion to SiC from PCOCF at  $1000^\circ\text{C}$  is reasonably high, i.e., 75%. The change in the ceramic yield of PCOH and PCOCF indicates that 1 g of PCOCF contains  $10^{-3}$  mol of organofluoric side chains. TGA behavior of PCOCF-coated SiC powder is similar to that of PCOCF itself. Thus, the organofluoric side chains of the binder of the SiC powder compact may burn out completely at temperatures  $>700^\circ\text{C}$ .

XRD patterns of the PCOCF heat-treated at  $1000^\circ$ ,  $1500^\circ$ , and  $2050^\circ\text{C}$  are shown in Fig. 6. The heat-treated PCOCF gives

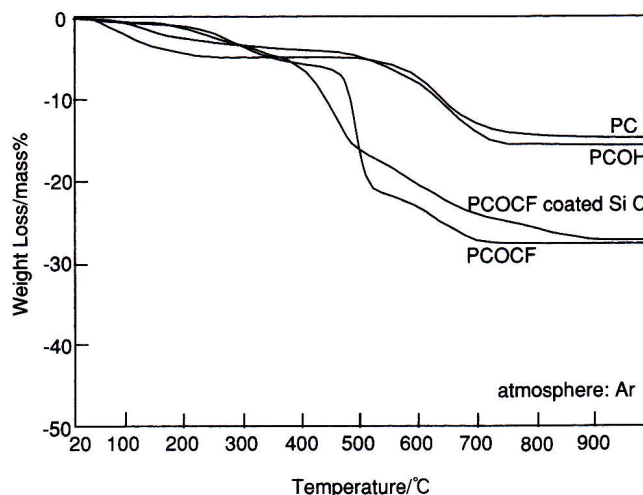


Fig. 5. TGA curves of PC, PCOH, PCOCF, and PCOCF-coated SiC powder in flowing argon.

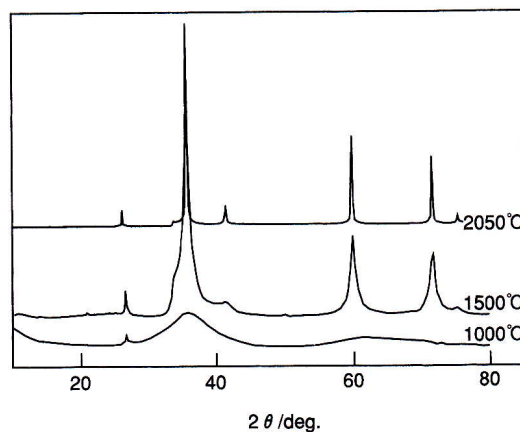


Fig. 6. XRD spectra of PCOCF heat-treated at various temperatures in pure argon.

an amorphous phase at  $1000^\circ\text{C}$ , from which crystallization to the 3C-SiC phase starts at  $1500^\circ\text{C}$  and ends at  $2050^\circ\text{C}$ . Almost all the heat-treated PCOCF converts to 3C-SiC crystalline powders.

## (2) Properties of PCOCF-Coated Powder and the Green Compacts

Figure 7 shows the oxygen contents of PCOCF-coated SiC powders stored in wet air as a function of holding time. No significant change in the oxygen content is observed in the PCOCF-coated powder samples, whereas the oxygen content of the as-received SiC powder increases to 2.0 mass% in 10 d. This means that excellent water repellency can be achieved by modifying the SiC powder surface with PCOCF.

Figure 8 shows the relationship between binder addition and relative green density of the PCOCF-coated SiC powders compared with PC- and other binder-coated SiC powders. The relative green density of the PCOCF-coated SiC powder compact is the highest among the binders. Therefore, the closest packing of SiC particles occurs when they are coated with PCOCF.

## (3) Properties of Sintered SiC Compacts

The density of the hot-pressed, sintered compacts is shown in Fig. 9. A small amount of PCOCF coating increases the density, which reflects the high green density of the SiC powders. A coating of 10 mass% of PCOCF, however, decreases the density, even if the relative density of the green compact is



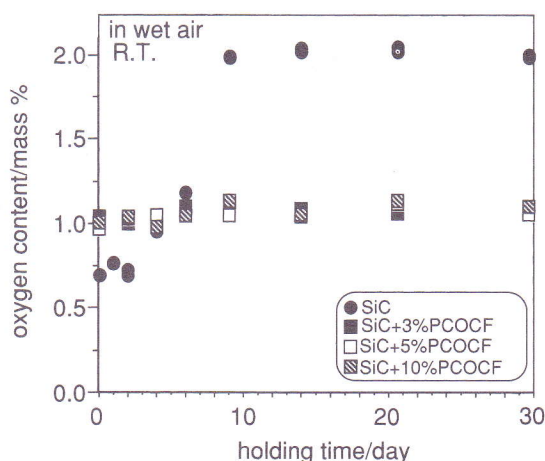


Fig. 7. Oxygen content of PCOCF-coated SiC powder stored in wet air as a function of holding time.

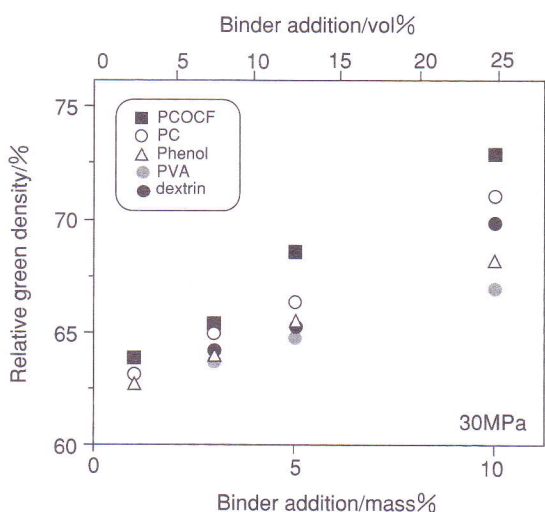


Fig. 8. Relationship between binder content and relative green density.

the highest. The decrease in density may be due to trapped gases from PCOCF during sintering.

Four-point-bend strengths of the sintered compacts also are shown in Fig. 9. The bending strength slightly increases with PCOCF coating (<10 mass%), and it undergoes a maximum (550 MPa) at 3 mass% PCOCF coating. The scatter in strength values significantly decreases with amount of coating for small amounts of PCOCF (3–5 mass%). This decrease in density indicates the sintered body of the PCOCF-coated SiC powder compact contains fewer large pores. The density may depend on the improved packing properties in the green compact of the PCOCF-coated SiC powder.

The polished and etched surfaces of the sintered compacts with different amounts of PCOCF coating have been observed by SEM (Fig. 10). The sintered compact coated with 3 mass% PCOCF consists mainly of uniform fine grains (3–10  $\mu\text{m}$ ), containing a small amount of elongated grains, and its microstructure is very similar to that of the sintered compact coated with 3 mass% phenol. Constituent grains in the sintered compact coated with 5 mass% PCOCF are larger than those of the sintered compact coated with 3 mass% PCOCF. Large, elongated grains ( $\sim 30 \mu\text{m}$ ) appear in the sintered compact coated with 10 mass% PCOCF, and the constituent grains exhibit a bimodal distribution. The difference in the carbon content of each sample, i.e., the PCOCF content, may cause the difference

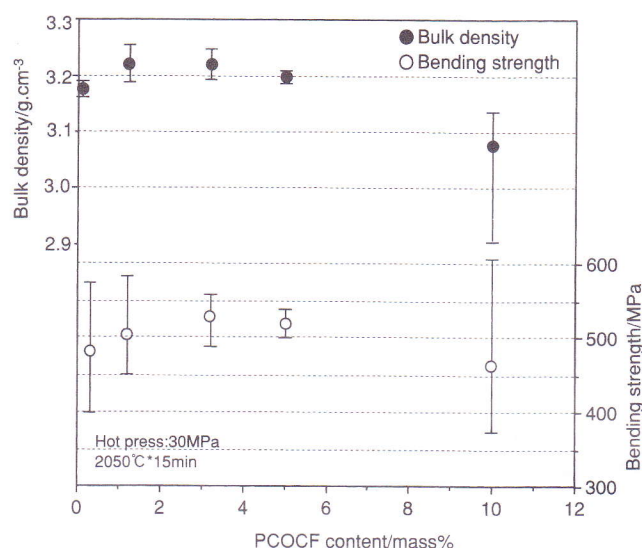


Fig. 9. Bulk density and four-point bend strength of hot-pressed, sintered SiC compact as a function of PCOCF content.

in the microstructure. Excess coating of PCOCF (>5 mass%) induces SiC grain growth.

The bending strength of the sintered compact with 5 mass% PCOCF is slightly lower than that of the sintered compact with 3 mass% PCOCF. The lower bending strength is due to accelerated grain growth, as shown in Fig. 10, even though the sintered compact is almost fully densified, as shown in Fig. 9. The bending strength of the sintered compact with 10 mass% PCOCF is lower than those of all other sintered compacts, because the bulk density of the 10 mass% PCOCF-coated SiC is the lowest, as shown in Fig. 9. The sintered compact is composed of large, elongated grains, as shown in Fig. 10.

The fracture toughness of all the sintered compacts is  $\sim 3.0 \text{ MPa}\cdot\text{m}^{1/2}$ . The bending strength increases and the scatter in strength values decreases with PCOCF coating (<10 mass%), and the fracture toughness does not decrease. SEM observation of the fracture surfaces shows that all sintered compacts have a transgranular fracture behavior. PCOCF coating does not affect the fracture properties.

The roles of PCOCF coating are (1) to densify the sintered compact without affecting the microstructure when using a 3 mass% coating, and (2) to densify and enhance SiC grain growth when using a 5 mass% coating. Excess coating of PCOCF (10 mass%), however, causes trapped pores as well as grain growth in the sintered compacts. PCOCF coating is effective to control the microstructure of SiC and to decrease the sintering temperature of SiC.

#### IV. Conclusion

In this work, a chemically modified polycarbosilane (PC) containing organofluoric groups (PCOCF) has been synthesized by the reaction of PC and fluoroalkylmethyldimethoxysilane to prepare an efficient compaction binder for SiC powders with 0.5 mass% of boron as a sintering aid. The results are summarized as follows:

- (1) PCOCF, the chemically modified PC, has been successfully synthesized from PC and fluoroalkylmethyldimethoxysilane.
- (2) A reasonably high ceramic yield of 75% has been achieved in PCOCF synthesis.
- (3) PCOCF-coated SiC powders show improved packing properties because of the organofluoric side chains.
- (4) PCOCF-coated SiC powders have excellent oxidation resistance.



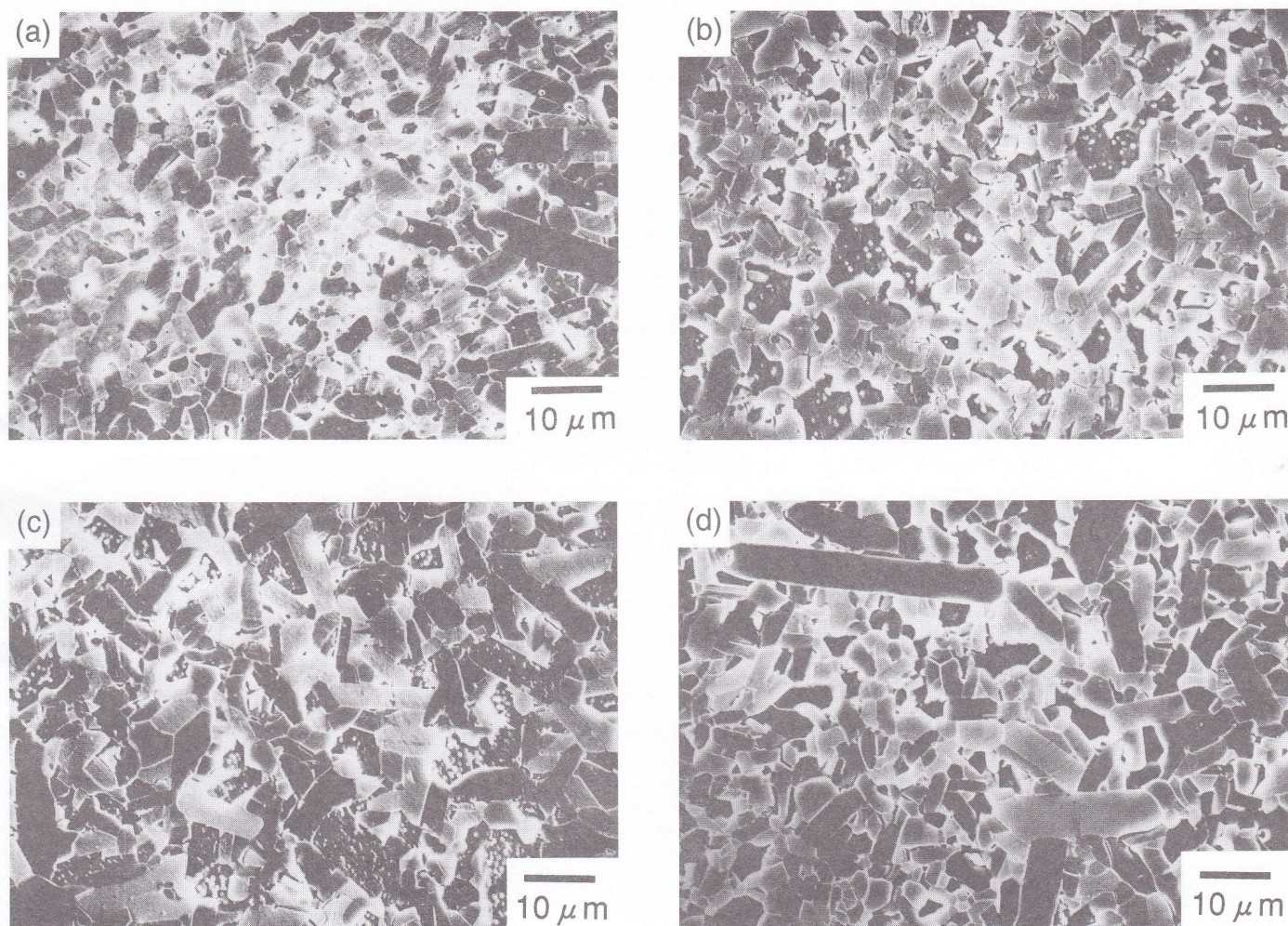


Fig. 10. SEM photographs of polished and etched surfaces of SiC + 0.5% B + (a) 3% phenol, (b) 3% PCOCF, (c) 5% PCOCF, and (d) 10% PCOCF.

(5) Four-point bend strength increases with amount of PCOCF coating, and it is a maximum (550 MPa) at 3 mass% PCOCF coating.

(6) The scatter in bending strength values of hot-pressed, sintered PCOCF-coated SiC significantly decreases with amount of PCOCF coating.

(7) Coating of PCOCF of >5 mass% enhances SiC grain growth.

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