

## Formation of SiC Powder from Laser Heated Vapor Phase Reactions

Kiyoshi SAWANO\*, John S. HAGGERTY and H. Kent BOWEN

( Massachusetts Institute of Technology  
Cambridge, Massachusetts 02139, U. S. A.  
\* R & D Laboratories I, Nippon Steel Corporation  
1618, Ida, Nakahara-ku, Kawasaki-shi 211 )

*SiC powder has been synthesized using CO<sub>2</sub> laser-heated vapor phase reactions in the silane-methane gas system. SiC particles form via a two-step reaction: silicon particle formation and subsequent carburization of the silicon particles. It is shown that the silicon particles form at the lower edge of the reaction zone, and carburization occurs in the hottest region, through which the CO<sub>2</sub> laser beam passes. The formation of silicon particles can be explained by silane pyrolysis and subsequent collisions and coagulation of the silicon particles. The carburization mechanism of the silicon particles is discussed in terms of diffusion through a SiC product layer and methane pyrolysis. The rate-controlling step is believed to depend on the reaction conditions such as temperature and gas stoichiometry.*

[Received July 21, 1986]

## Preparation of Ultrafine Powders of AlN and (AlN+Al) by Nitrogen Plasma-Al Reaction

Masahiro UDA, Satoru OHNO and Hideo OKUYAMA

( National Research Institute for Metals  
3-12, Nakameguro 2-chome, Meguro-ku, Tokyo 153 )

*Ultrafine powders of AlN and (AlN+Al) with particle sizes of less than 0.5 μm were produced by arc melting of Al in atmospheres of N<sub>2</sub>+Ar, N<sub>2</sub>+H<sub>2</sub>, N<sub>2</sub>+NH<sub>3</sub> at 0.1 MPa pressure. The size, surface area, chemical composition, and crystal structure of obtained powders were determined by electron microscope, BET method, chemical analysis and X-ray diffraction analysis respectively. The following results were obtained.*

- ( 1 ) AlN ratio in (AlN+Al) powders obtained in (N<sub>2</sub>+Ar) atmospheres decreases from 0.3 to zero with decreasing nitrogen partial pressure.
- ( 2 ) AlN ratio in (AlN+Al) powders obtained in (N<sub>2</sub>+NH<sub>3</sub>) atmospheres increases from 0.3 to 0.95 with increasing NH<sub>3</sub> partial pressure.
- ( 3 ) Projections were formed on molten Al surface by evolved nitrogen gas from the melt and by reaction between nitrogen and molten Al.

*The reaction between nitrogen plasma and molten Al plays an important role in the formation of ultrafine particles. The driving force for the formation of ultrafine particles can be explained as follows: In arc melting of Al in a nitrogen atmosphere, molten Al simultaneously contacts with two gas phases, one is atomic nitrogen in the arc gas phase and another is the molecular nitrogen in the non-arc gas phase. The extent of dissolution of atomic nitrogen in the molten Al is much larger than that of molecular one. Then, the dissolved nitrogen through the arc gives higher nitrogen content for the non-arc gas phase. That is, super-saturated nitrogen will evolve in non-arc gas phase carrying with Al vapor. A kind of enhanced evaporation of Al is induced. This Al vapor reacts with surrounding gas species near the arc such as N<sub>2</sub>, N, NH<sub>3</sub>, NH<sub>2</sub>, NH in high temperature ranges (above 1600 K) to form AlN or (AlN+Al) vapor which condense to ultrafine particles.*

[Received July 21, 1986]

## Preparation of Fine Silicon Nitride Powders by Vapor Phase Reaction of Nitrogen Containing Organosilicon Compound with Ammonia

Takasi SUZUKI, Takamasa KAWAKAMI, Takesi KOYAMA, Kansei IZAKI, Rieko NAKANO,  
Takuji SHITARA, Koichi HAKKEI, Toshio HIRAI\* and Koichi NIHARA\*\*

( Mitsubishi Gas Chemical Company, Inc.  
5-2, Marunouchi 2-chome, Chiyoda-ku, Tokyo 100  
\* The Research Institute for Iron, Steel and Other Metals, Tohoku University  
\*\* The National Defence Academy, Physics Department )

*Fine amorphous silicon nitride powders were prepared by vapor phase reaction between nitrogen-containing, chloride-free organosilicon compound and NH<sub>3</sub> at the production rate of ~200 g/h. Crystallization was carried out by heat treating the resulting amorphous powders in Ar atmosphere at 1500°C. High purity crystalline silicon nitride powders could be synthesized*

with metallic impurities Fe, Al, Ca < 50 ppm, carbon content < 1 wt% and oxygen content of ~1 wt%. The specific surface area, mean particle size and  $\alpha$  phase content were ~10 m<sup>2</sup>/g, 0.6  $\mu$ m and >95 wt%, respectively. Mechanical properties of hot-pressed silicon nitride from the resulting crystalline powders were comparable to hot-pressed bodies fabricated from high purity silicon nitride powders by imide decomposition and carbothermic reduction of silica.

[Received July 28, 1986]

## Preparation of SiC and Si<sub>3</sub>N<sub>4</sub> Powders by RF-Plasma

Akira KUIBIRA, Akira MITSUI, Junichi HOJO and Akio KATO

(Department of Applied Chemistry, Faculty of Engineering, Kyushu University)  
Hakozaki, Higashi-ku, Fukuoka-shi 812

By the RF-plasma method, preparation of silicon carbide and silicon nitride powders from alkoxyxilanes ( $(\text{Si}(\text{OC}_2\text{H}_5)_4)$ ,  $(\text{CH}_3)_2\text{Si}(\text{OC}_2\text{H}_5)_2$ , and  $(\text{CH}_3)_2\text{Si}(\text{OCH}_3)_2$ ) and from  $\text{SiCl}_4$ - $\text{NH}_3$  system, respectively, was studied. In the preparation of silicon carbide, the reaction product consisted of  $\beta$ -SiC, free carbon and silicon oxide. The amount of SiC in the powder product increased with rising reaction temperature or with increasing  $[\text{H}_2]$ . The amount of SiC increased with decreasing C/Si ratio in alkoxyxilanes and was estimated to reach 100% when the C/Si ratio is around 3. In the preparation of silicon nitride, the reaction product was white and non-crystalline powders with a particle size of 0.01-0.02  $\mu$ m. The composition of powders was virtually independent of  $[\text{NH}_3]$  and close to that of  $\text{Si}_3\text{N}_4$ . Heat treatment at 1550°C was necessary to obtain the crystalline  $\alpha$ - $\text{Si}_3\text{N}_4$ .

[Received July 14, 1986]

## Synthesis of Precursors for SiC-C Fibers by Copolyrolysis of Polysilane and Pitch

Yoshio HASEGAWA and Kiyohito OKAMURA\*

( The Research Institute for Special Inorganic Materials  
Asahi-mura, Kashima-gun, Ibaraki 311-14 )  
\* The Oarai Branch, The Research Institute for Iron, Steel and Other Metals, Tohoku University )

Precursors of carbon-containing SiC fibers (SiC-C fiber) were synthesized with high yield by the copolyrolysis of poly(dimethylsilane) and 3, 9-80 wt% pitch. During copolyrolysis the decomposition product of polysilane reacts with pitch molecule with no significant condensation among pitch molecules. SiC-C fibers were obtained by melt spinning of the precursors and heat-treatment up to 1400°C in vacuum. The tensile strength and Young's modulus of the SiC-C fiber decreased with increasing carbon content. The electric conductivity is two orders of magnitude larger than that of SiC fibers obtained from pure polycarbosilane.

[Received July 14, 1986]

## Preparation of Ultrafine SiC Powders by Pyrolysis of Tetramethyldisilane

Morinobu ENDO, Toshiro SANNO, Kazuhiro MORI, Nobuaki URASATO\* and Minoru SHIRAIISHI\*\*

( Faculty of Engineering, Shinshu University  
Wakasato, Nagano-shi 380 )  
\* Shin-etsu Chemical Co., Ltd.  
\*\* National Institute for Pollution and Resources )

Ultrafine silicon carbide powders of high purity have been prepared by pyrolysis of tetramethyldisilane at temperatures 700° to 1400°C. The diameter of the particles changed from 5 to 200 nm depending on the pyrolysis conditions. The ultrafine particles consist of small crystallites of  $\beta$ -type SiC, arranged randomly in the particles. The molar ratio (C/Si) of the powders can be also controlled within the range 0.9 to 1.2 by pyrolysis conditions such as pyrolytic temperature and reactant concentration. No admixtures of carbon such as carbon soot or film on the SiC particles were observed with a high-resolution electronmicroscope even in the powders having (C/Si) ratio more than 1.0. The formation mechanism of the ultrafine SiC powders is discussed in relation to the structure.

[Received July 15, 1986]