Synthesis of SIC(β) Ultrafine Particles From Si, SiO, or SiO_2 Powder and CH_4

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A process for synthesis of ultrafine ceramic particles using a high-temperature flame is described. Thermodynamic calculations for the Si-C-O-H system were performed using a computer program for calculation of complex chemical equilibrium compositions. Possible synthesis of ultrafine SiC particles using Si, SiO, and SiO_2 powders (1.0–3.0 μm) as Si sources was investigated using an electric furnace (5 kW) at 2,018 K. Particle products were characterized mainly by X-ray diffraction analysis, field emission scanning electron microscopy, and thermogravimetry-differential thermal analysis. Ultrafine particles (18–29 nm) of cubic SiC (β-SiC) could be produced through $Si + CH_4$, $SiO + CH_4$ and $SiO_2 + CH_4$ reaction systems, respectively. The effect of hydrogen addition on the synthesis of SiC was also investigated. Ultrafine particles of 96.0% SiC selectivity were obtained when H_2 was added to the $SiO + CH_4$ reaction system, and the CH_4/SiO molar ratio in this case was 2.0.

Introduction

The outstanding strength of silicon carbide (SiC) under oxidizing and high-temperature conditions, in addition to its excellent corrosive resistance, makes it an attractive material for industrial applications. For these reasons, SiC formation processes have been extensively studied by many researchers over the past few decades. The most representative process among them is the chemical vapor deposition (CVD) process. Comprehensive models (Schlichting, 1980; Weimer et al., 1993; Kim and Davis, 1986) including details of gas-phase and surface chemistry coupled with reactor fluid mechanics of Si-C-H, Si-C-Cl-H systems and others have been developed. However, because it involves the use of expensive and ignitable silicon (Si) source gases such as monosilane (SiH_4), trichlorosilane ($HSiCl_3$) and methyltrichlorosilane (CH_3SiCl_3), the CVD-SiC process is costly and considered to be unsafe. Furthermore, no industrial scale-up and optimization method for this process has been developed to date. Kong et al. (1986) proposed synthesis of ultrafine SiC particles using silica (SiO_2) as a Si source. However, this process would also be costly because it would involve the use of a DC plasma jet (900 A, 25 V) for achieving steep temperature gradients of over 1×10^4 K/m.

A low-cost and safe process for synthesis of ultrafine SiC particles using Si or SiO_2 powder as a Si source and a high-temperature (2,500 K) methane-oxygen (CH_4-O_2) flame was developed and investigated in our laboratory (Xu et al., 1996). It consists of four steps: continuous supply of raw powder, evaporation of the source powder over a high-temperature flame, synthesis of SiC, and collection of ultrafine particles. The advantages of this process are as follows:

(1) The cost of the Si and SiO_2 powders used in the synthesis is very low, compared with the cost of other Si sources such as SiH_4, H_3SiCl_3 and CH_3SiCl_3;

(2) It is a very economical means of realizing a high-temperature field since no electric power input such as in a plasma or laser method is required;

(3) Ultrafine particles can be produced by the reaction of gas phases at atmospheric pressure; and

(4) Use of expensive refractory materials for the reactor wall is unnecessary, because the high-temperature zone can be formed by the flame itself.

The process of ultrafine ceramic particle formation should be such that the Si source powder melts and subsequently evaporates, and then reacts with carbon-bearing species formed by CH_4 decomposition to form ultrafine SiC particles. However, a CH_4-O_2 flame has a complex chemical reaction system involving more than one-hundred chemical

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Table 1. Species Considered for a Si-C-O-H System

Equilibrium Gas Phases						
C	C_2H_2	C_4	HSiOH	H_3SiOH	$SiCH_2$	SiOH
CH	C_2H_3	C_4H_2	H_2	O	$SiCH_3$	SiO_2
CH_2	C_2H_4	C_4H_3	H_2O	OH	SiC_2	Si_2
CH_2O	C_2H_5	H	H_2O_2	OSiHO	$Si(CH_3)_2$	Si_2C
CH_3	C_2H_6	HCO	H_2SiC	OSiOH	$Si(CH_3)_3$	Si_2H_2
CH_3O	C_2O	HO_2	H_2SiO	O_2	SiH	Si_2H_3
CH_4	C_3	HSiC	H_2SiOH	O_3	SiH_2	Si_2H_5
CO	C_3H_2	HSiCCH	H_2SiO_2	Si	SiH_3	Si_2H_6
CO_2	C_3H_3	$HSiCH_2$	H_2SiC	SiC	SiH_4	Si_3
C_2	C_3H_4	$HSiCH_3$	H_3SiO	SiCH	SiO	Si_3H_8
C_2H		HSiO		SiCCH		
Equilibrium Condensed Phases						
	Graphite		Si	β-SiC	SiO_2	

species including CH_4, C_2H_2, CH_3, CH_2, CH_2O, CHO, CO, CO_2, H_2O, H and OH. It is difficult to control the operating parameters such as residence time and concentration of species (Beer and Chigier, 1972; Ulrich, 1971), precisely.

In our study, thermodynamic analyses in which complex chemical equilibrium compositions were considered were performed first for prediction of general characteristics of chemical reactions. Then, an elementary reaction system of Si, silicon monoxide (SiO) and SiO_2 powders as Si sources, and CH_4 gas as a carbon source was investigated in an electric furnace. The experiments of ultrafine SiC particle synthesis through $Si + CH_4$, $SiO + CH_4$ and $SiO_2 + CH_4$ reaction systems were performed in an argon (Ar) inert-gas atmosphere. The influence of hydrogen (H_2) addition on the probability of SiC particle formation was also investigated. The experimental results were compared with the thermodynamic equilibrium calculation results. Finally, the SiC formation process was discussed based on the experimental results and reaction kinetics considerations.

Thermodynamic Consideration

A computer program for calculation of complex chemical equilibrium compositions, rocket performance, incident and reflected shocks, and Chapman-Jouguet detonations (Gordon and McBride, 1971) is often used for calculating the chemical equilibria in a system containing a gaseous phase, solid solutions, liquid solutions, and compounds of invariable composition. Thermodynamic analysis has also been performed using a Gibb's free energy minimization program, SOLGASMIX-PV (Besmann, 1977), in some detail by Eriksson (Eriksson, 1971; Eriksson and Rosen, 1973; Eriksson, 1975) and Kingon et al. (1983) for the Si-C-O-H system, and Fischman and Petuskey (1985) for the Si-C-Cl-H system. The calculations comprise system composition calculations with certain constraints, and for a minimum of free energy. The constraints refer to the conservation of the mass of each element present at either constant pressure or constant volume. This process is known as the direct minimization of the system total free energy.

In our investigation, the probabilities of occurrence of three reaction systems of $Si + CH_4$, $SiO + CH_4$, and $SiO_2 + CH_4$, as the Si-C-O-H system for SiC formation in Ar atmosphere were calculated. 73 gas species and four condensed phases (graphite, cubic SiC, Si and SiO_2) containing Si, C, O and H were considered for a temperature range of 500 to 4,000 K and are listed in Table 1. The free energies, enthalpies, and entropies of formation of each were taken from the *JANAF Thermochemical Tables* (1971). The calculation results for the three reaction systems are relatively similar and are shown in Figures 1a, 2a, and 3a. It was noted that the cubic form β-SiC(s) was more stable than the hexagonal polytypes in the

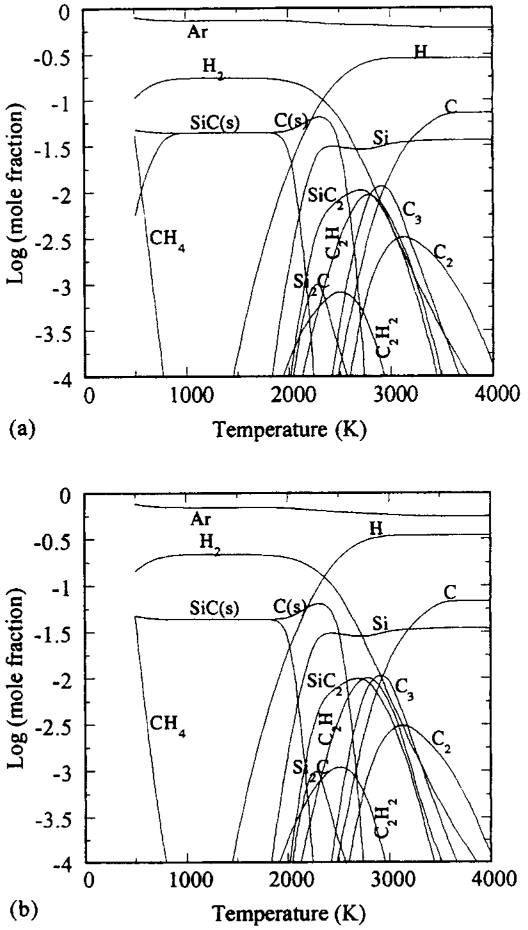

Figure 1. Thermodynamic equilibrium compositions for reaction systems: (a) $Si:CH_4:Ar = 0.05:0.10:0.85$; (b) $Si:CH_4:H_2:Ar = 0.05:0.10:0.05:0.80$.

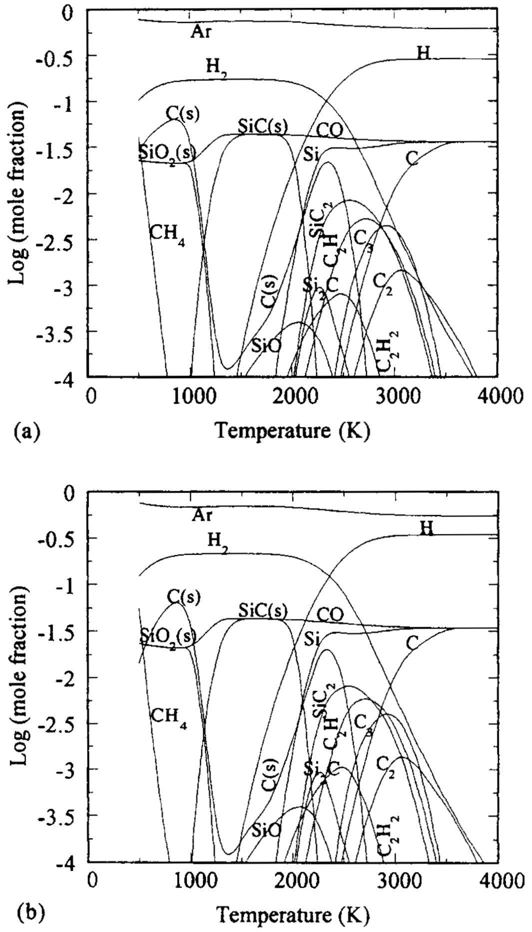

Figure 2. Thermodynamic equilibrium compositions for reaction systems: (a) SiO:CH_4:Ar = 0.05: 0.10:0.85; (b) SiO:CH_4:H_2:Ar = 0.05: 0.10:0.05:0.80.

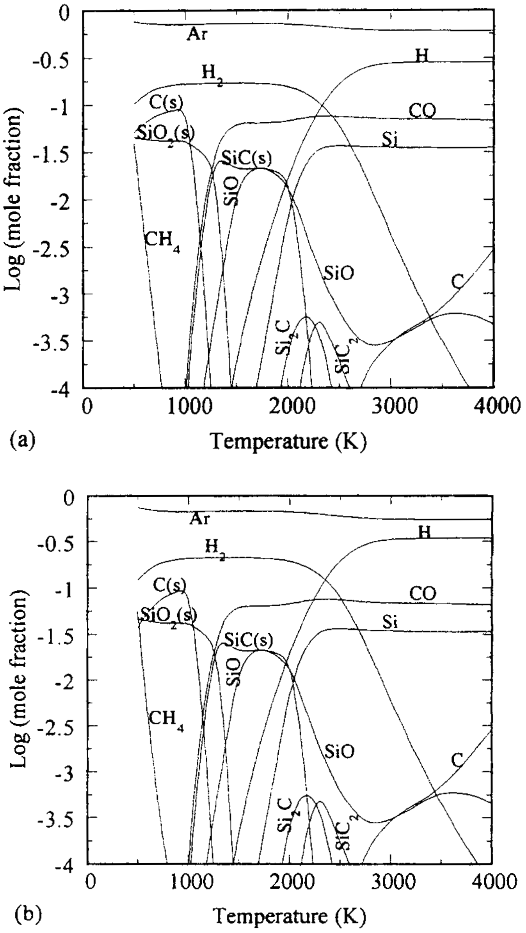

Figure 3. Thermodynamic equilibrium compositions for reaction systems: (a) SiO_2:CH_4:Ar = 0.05: 0.10:0.85; (b) SiO_2:CH_4:H_2:Ar = 0.05: 0.10:0.05:0.80.

temperature range of 1,000 K to 2,300 K. Therefore, only the polymorph should be considered in the polytypes of SiC (Guo et al., 1995; Kingon et al., 1983; Harris et al., 1971). The thermodynamic analysis results indicate that the highest potential for SiC formation lies in the temperature range of 1,000 to 2,300 K. The other species which should be considered in this temperature range are H(g), H_2(g), C(s), C_2H(g), C_2H_2(g), Si(g), SiC_2(g), and Si_2C(g) in the Si-CH_4-Ar system; H(g), H_2(g), C(s), C_2H(g), C_2H_2(g), CO(g), Si(g), SiO(g), SiO_2(s), SiC_2(g) and Si_2C(g) in the SiO-CH_4-Ar system; and H(g), H_2(g), C(s), CO(g), Si(g), SiO(g), SiO_2(s), SiC_2(g) and Si_2C(g) in the SiO_2-CH_4-Ar system, depending on the molar ratios of Si/CH_4, SiO/CH_4 and SiO_2/CH_4, respectively. From the equilibrium diagrams, it is known that the main products of CH_4 decomposition vary with temperature. At 500–2,000 K, they are C(s), CO(g) (except in the Si-CH_4-Ar system), H_2(g) and H(g); and at 2,000–3,500 K they are C(s), CO(g) (except in the Si-CH_4-Ar system), H_2(g), H(g), C_2H(g), C_2H_2(g), C(g), C_2(g) and C_3(g) (except in the SiO_2-CH_4-Ar system). Figures 1b, 2b, and 3b show the thermodynamic equilibrium diagrams for the three reaction systems of $Si + CH_4 + H_2$, $SiO + CH_4 + H_2$ and $SiO_2 + CH_4 + H_2$, respectively. Apparently, no distinct increase in the probability of SiC formation via the three reaction systems occurs upon H_2 addition. This suggests that the influence of H_2 addition on the probability of SiC particle formation induces no changes in the chemical equilibrium compositions of the reaction systems.

Since the calculations are based on equilibrium thermodynamics, the final result is independent of the initial form of the elements. It is also assumed that the reactions in the gas phase rapidly reach equilibrium, yielding molecules which represent the most stable distribution of chemical components. Thermodynamic investigation results provide useful general guidelines for the prediction of which equilibrium species are formed at various temperatures.

Experimental Procedure

From a thermodynamic viewpoint, SiC formation can occur over a wide range of temperatures (1,000–2,300 K) in a Si-C-O-H system. The temperature is most important for the formation of SiC, if the molar ratio of Si/CH_4, SiO/CH_4, or SiO_2/CH_4 is fixed. An overall picture of the reactions was obtained and the equilibrium compositions of the final products were predicted, but the actual kinetics and mechanisms of the reactions remain unclear. Moreover, the synthesis of

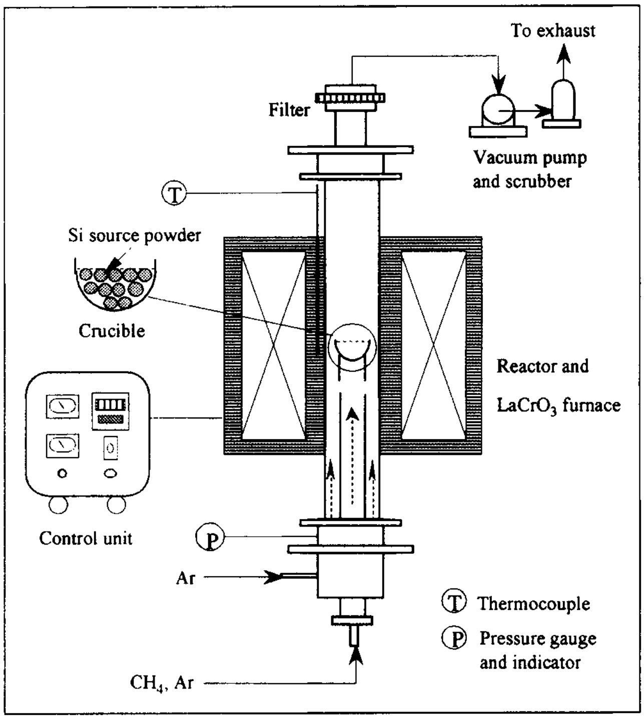

Figure 4. Experimental apparatus including an electric furnace.

ultrafine ceramic particles by the CH_4-O_2 flame method is very complex, because over 80 element reactions occur and more than 100 chemical species are present in the CH_4-O_2 flame (Beer and Chigier, 1972). Therefore, in the present study, the possibility of SiC synthesis in an elementary Si-C-O-H system was investigated experimentally using an electric furnace system.

Figure 4 shows the experimental apparatus. It consists of an electric resistance furnace equipped with one set of $LaCrO_3$ heaters (total power: 5 kW, Nippon Kagaku Togyo Co., Ltd.) and a program control unit (model EC5600, Okura Electronic Corp.). The maximum heating temperature of the heaters is 2,073 K and the temperature of the hot zone inside the alumina reactor tube (ID: 30 mm) is at least 2,018 K. The heating rate was 4.0 K/min as controlled by the program and measured using a Pt40Rh20 thermocouple between the heaters and the reactor tube. An alumina crucible loaded with Si source powder was kept in the hot zone of the furnace. Three Si source powders, crystal Si (avg. size: 1.2 μm, purity: 99.5%, Vacuum Metallurgical Corp.), amorphous SiO (avg. size: 2.5 μm, purity: 99.9%, Huruuchi Chemistry Corp.), and amorphous SiO_2 (avg. size: 1.7 μm, purity: 99.9%, Mizusawa Chemical Industry Corp.) were used. The weight of the Si source powder supplied in each experiment was 500–580 mg.

The gases were introduced into the bottom of the reactor tube and exhausted from the top. For prevention of oxidation of the Si source or SiC during the reaction, Ar gas was fed as a purge gas along the wall of the reactor tube. CH_4 (conc.: 0.5–3.0 vol. %) diluted with Ar carrier gas was fed into the hot zone of the reactor tube from an alumina sample tube (ID: 16 mm); the feed time was 5–15 min. The mole ratios of CH_4/Si, SiO and SiO_2 were determined by the changes in the CH_4 feed time. In the final experimental stage, H_2 (conc.: 10 vol. %) balanced by Ar was added to the CH_4 for examination of its effect on the probability of the SiC formation. The total gas linear velocity was 14.0 cm/s at 2,018 K. The experiments were carried out under pressures slightly higher (10–50 mm H_2O) than atmospheric pressure. The operating conditions employed in the SiC synthesis experiments are summarized in Table 2.

A paper filter was placed in the downstream region of the reactor tube to trap ultrafine particles. After the completion of the synthesis experiments, the specimen collected on the filter was oxidized (time: 20 min) in atmosphere using another electric furnace (1 kW) at about 1,073 K to remove the byproducts such as soot formed by condensation of free carbon. The specimens collected from the filters and those after the oxidation treatment were examined using X-ray diffraction analysis (XRD) (model RAD-C, Rigaku Co., Ltd.) with Ni-filtered CuKα radiation at a scanning rate of 4° min^{-1} and an acquisition range of 10–80° for 2θ. Field emission scanning electron microscopy (FE-SEM) (model S-900, Hitachi Co., Ltd.) were employed for determination of particle shape. The free carbon content in the specimens was determined by thermogravimetry-differential thermal analysis (TG-DTA) (model TGD-7000RH, Vacuum Science and Engineering Corp.) in an oxidizing atmosphere (air). The heating rate was 5.0 K/min and the maximum temperature was set at 1,273 K.

Results and Discussion

Evaporation of source powders

Si, SiO and SiO_2 powders were evaporated in the electric furnace in Ar atmosphere. The fine particles obtained on the filters were whitish brown (from Si), yellowish brown (from SiO), and white (from SiO_2). Figure 5 shows a FE-SEM image of fine particles formed by evaporation and condensation of SiO powders. The relative and cumulative distributions of ultrafine particles formed from the Si, SiO and SiO_2 powders were calculated from the results of measurement of several hundreds of particles in various areas of the FE-SEM images and are shown in Figure 6. The average sizes of the ultrafine

Table 2. Operating Conditions for the Synthesis of Ultrafine SiC Particles

Source Particle	Avg. Particle Size (μm)	Furnace Power		Furnace Temp. (K)	Source Particle Wt. (mg)	CH_4 Conc. (vol. %)	H_2 Conc. (vol. %)	Feed Time (min)	CH_4/SiO_x ($x = 0, 1, 2$) Molar Ratio	Balance Gas
		Voltage (v)	Current (A)							
Si	1.2	59.0	41.0	2018	500–530	0.5–3.0	10	5–15	2.0–5.0	Ar
SiO	2.5	59.0	41.0	2018	550–580	0.5–3.0	10	5–15	1.5–4.5	Ar
SiO_2	1.7	59.0	41.0	2018	520–560	0.5–3.0	10	5–15	2.3–5.0	Ar

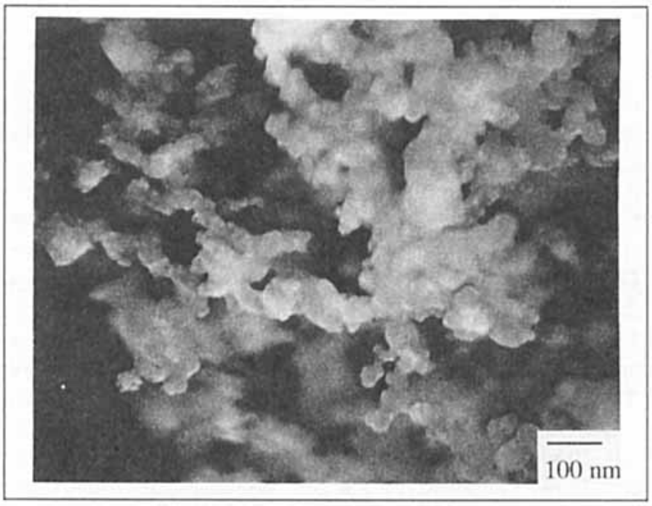

Figure 5. FE-SEM image of ultrafine particles formed by evaporation and condensation of SiO powder.

particles formed from the Si, SiO and SiO_2 powders were about 27.0 nm, 19.0 nm, and 39.0 nm, respectively. It was thus concluded that at the high temperatures, the Si, SiO, and SiO_2 powders were vaporized and condensation of the vapor resulted in formation of the ultrafine particles. Furthermore, the vaporization rate was 0.94×10^{-4} g/s for Si, 2.37×10^{-4} g/s for SiO, and 1.25×10^{-4} g/s for SiO_2, as determined from the weight changes of the crucibles containing the Si source powders. This indicates that the SiO powder evaporated more quickly than the Si and SiO_2 powders at the same high temperature. The XRD patterns for all of the samples of ultrafine particles collected on the filters showed peaks attributed to Si, that is, at 28.4°, 47.3°, and 56.1° (JCPDS No. 27-1402; Pennsylvania, 1985). This suggests that some of the amorphous SiO and SiO_2 source powders decomposed and were converted to Si crystals at the high temperatures of the reaction system.

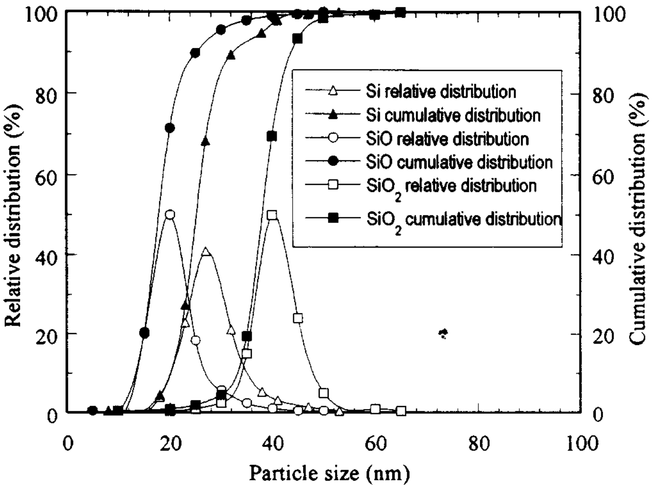

Figure 6. Size distributions of ultrafine particles formed by the evaporation and condensation of Si, SiO and SiO_2 powders.

Synthesis of ultrafine SiC particles

Experiments of ultrafine SiC particle synthesis through the reaction systems of $Si + CH_4$, $SiO + CH_4$ and $SiO_2 + CH_4$ in Ar atmosphere were performed. Figure 7 shows the XRD analysis results for the products of the reaction systems of (a) $Si + CH_4$; (b) $SiO + CH_4$; and (c) $SiO_2 + CH_4$ in the electric furnace reactor. Each of the patterns shows the peaks attributed to β-SiC, that is, at 35.6°, 60.0°, and 71.8° (JCPDS No. 29-1129) and Si, that is, at 28.4°, 47.3° and 56.1° (JCPDS No. 27-1402). The sizes of the β-SiC crystallites calculated using Scherrer's equation were 18.3 nm, 27.4 nm, and 28.9 nm for Si, SiO and SiO_2, respectively. This indicates that ultrafine β-SiC particles (18–29 nm) were synthesized in this experimental system. In addition, XRD patterns for free carbon (C) which might have been formed by CH_4 decomposition and alumina (Al_2O_3) possibly formed by Al_2O_3 present as an impurity in SiO_2 powder were observed. The free carbon content in the specimens was determined by TG-DTA in air atmosphere at a 1,273 K heating temperature. Figure 8 (trace (a)) shows a typical TG analysis curve for the product of the $SiO + CH_4$ reaction system. It was found that the oxidation of the free carbon in air atmosphere began at around 700 K and ended at about 950 K with release of heat. With increasing temperature above 950 K, this rate of oxidation of the ultrafine Si and SiC particles increased.

The SiC selectivities (SiC/(Si + SiC)) of the products obtained via the three reaction systems were calculated from the calibration curves for the weight percentages of SiC vs. the ratios of relative intensities of SiC-Si powder X-ray diffraction, based on the XRD patterns of the SiC-Si standard powder mixtures and the TG-DTA results. These results and the results of theoretical curves in the thermodynamic equilibrium calculations of the Si-C-O-H system at

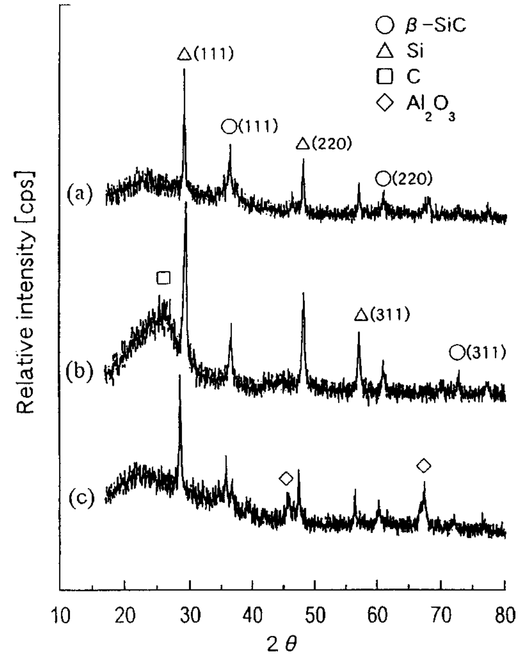

Figure 7. XRD patterns for products of reaction systems: (a) $Si + CH_4$; (b) $SiO + CH_4$; (c) $SiO_2 + CH_4$.

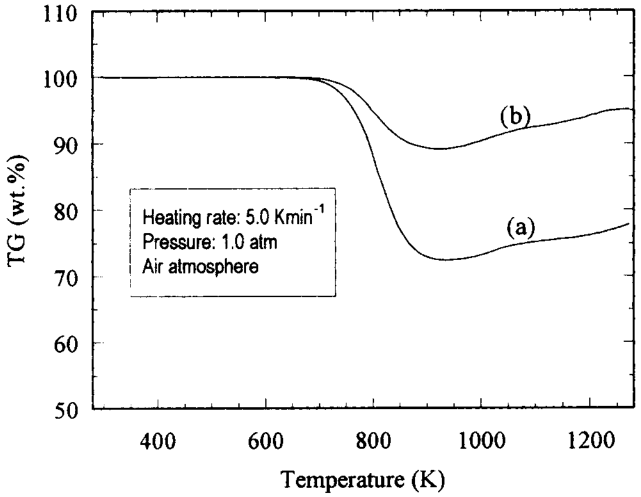

Figure 8. TG analysis curves for the products of reaction systems: (a) $SiO + CH_4$ and (b) $SiO + CH_4$ in an H_2 addition atmosphere.

2,000 K are illustrated in Figure 9. The theoretical curves show that the SiC selectivities depended on the reaction system used. The maximum calculated values were determined by the CH_4/Si, SiO and SiO_2 mole ratios, that is, 1.0 for CH_4/Si, 2.1 for CH_4/SiO, and 2.8 for CH_4/SiO_2. The values decreased markedly with decreases in the CH_4/Si, SiO and SiO_2 mole ratios. The experimental results show that the tendencies fit the theoretical selectivity curves fairly well and indicate that the SiC synthesis reactions in this experimental system approach to chemical equilibriums.

Effect of H_2 addition

The experiments of SiC synthesis through the reaction systems of $Si + CH_4$, $SiO + CH_4$ and $SiO_2 + CH_4$, respectively, were performed in an H_2 addition atmosphere balanced by Ar. The XRD patterns for the products obtained also show peak characteristics of β-SiC and Si, and the influence of H_2 addition on the synthesis of ultrafine SiC particles of the SiO $+ CH_4$ reaction system was stronger than that on the other two reaction systems. Figure 10 shows the XRD patterns for (a) the SiO source powder and (b) the product of the $SiO + CH_4$ reaction system. The peaks of β-SiC crystal, that is, at 35.6°, 60.0°, and 71.8° (JCPDS No. 29-1129), were mainly observed in the product. The free carbon content of the product was determined by TG-DTA in air atmosphere at a 1,273 K heating temperature, and the TG analysis curve for the product of the $SiO + CH_4$ reaction system is shown in Figure 8 (trace (b)). It was found that the rate of oxidation of free carbon in the air atmosphere was much lower than that in the case of the $SiO + CH_4$ reaction system without H_2 addition. Moreover, the SiC selectivity for the CH_4/SiO mole ratio of 2.0, which was 54.0% for the $SiO + CH_4$ reaction system without H_2 addition, was increased to 96.0% (shown in Figure 9) by H_2 addition. The experimental results for the $SiO + CH_4$ reaction system in the case of the H_2 addition atmosphere show that the tendency was higher than the theoretical selectivity curves shown in Figure 9.

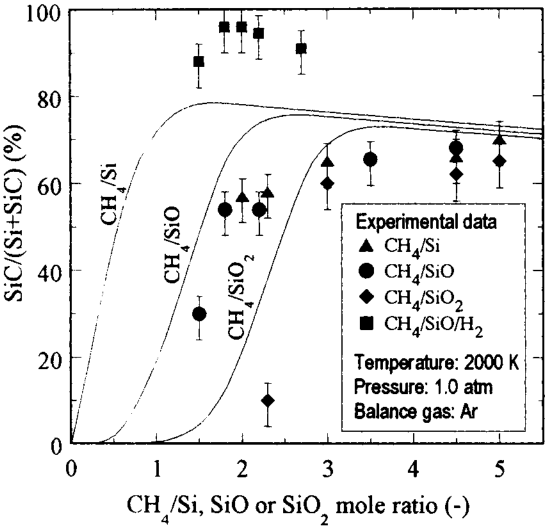

Figure 9. SiC selectivity as a function of CH_4/Si, SiO or SiO_2 mole ratio as determined in thermodynamic calculations and experiments.

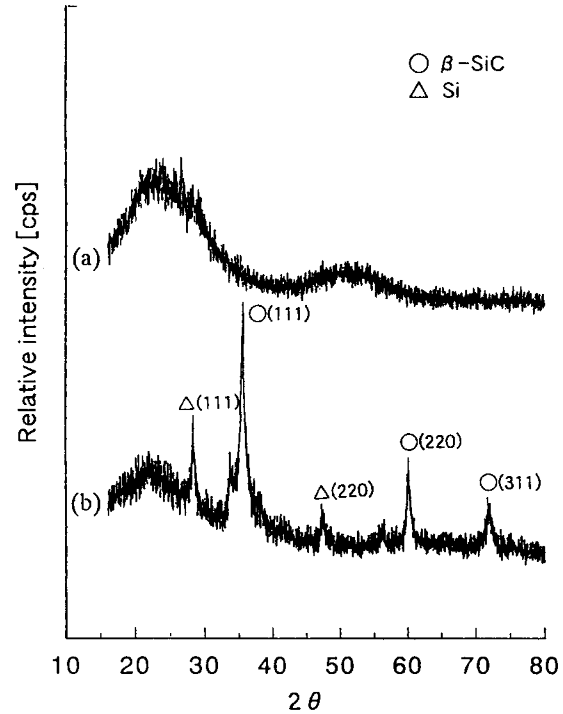

Figure 10. XRD patterns for (a) SiO powder; (b) product of the reaction system of $SiO + CH_4$ in an H_2 addition atmosphere.

From a kinetics viewpoint, it is considered that H_2 addition has two effects on the SiC synthesis. One is the inhibition of CH_4 cracking in the temperature range of 1,000–2,000 K and consequently of free carbon production. The other is the stimulation of the formation of active Si precursors such as Si, SiO, and Si_2C. It is also inferred that the higher SiC selectivity for the $SiO + CH_4$ reaction system in the case of the H_2 addition atmosphere is the result of the free Si precursor and CH precursor, such as the C_2H_2, ratio being near the optimal one in the experimental system, while the CH precursors are present at a relative excess in the other cases. The validity of this consideration has been tested by further experimentation.

Conclusions

Ultrafine β-SiC particles (18–29 nm) were synthesized from Si, SiO, and SiO_2 source powders (1.0–3.0 μm), respectively, in an electric furnace system at about 2,018 K. The experimental results were similar to the theoretical predictions based on the results of thermodynamic equilibrium calculations. Ultrafine SiC particles with high selectivity (96.0%) were synthesized in the $SiO + CH_4$ reaction system in an H_2 addition atmosphere.

These results indicate the feasibility of the proposed low-cost and low-energy-input process of synthesis of ultrafine SiC particles using SiO_2 source powders and a CH_4-O_2 flame.

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