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Combustion synthesis of high purity SiC powder by radio-frequency heating

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Abstract

High purity silicon carbide (SiC) powder was produced via combustion synthesis by radio frequency (RF) heating. The reaction described here was involved in four stages: preheating, initial combustion reaction, product formation and cooling. Significantly, the initial combustion reaction started from a low ignition temperature (T_i) in the range of 1160 °C–1200 °C, subsequently the reaction proceeded under RF heating. The C/Si ratio, reaction temperature and reaction time had a large influence on the resultant SiC powder. Particularly, based on the comparison of the ignition temperature and the reaction degree under different C/Si ratios, it was firstly proposed that the C/Si ratio with 1:1 was not the optimal condition for the initial combustion reaction. Moreover, with the increase of reaction temperature and the reaction time, the phase transitions of SiC from 3C to 6H and from 6H to 15R were found, respectively. The SiC grains grow with the reaction time due to the successive reaction processes including formation, decomposition, fusion and reformation.

Keywords: A. Combustion synthesis; C. High purity; C. Ignition temperature; D. SiC

1. Introduction

Silicon carbide (SiC) crystal is a promising material for high-temperature, high-power, high-frequency electronic, optoelectronic devices and engineering applications, owing to its superior properties, such as wide band gap, high critical breakdown electric field, high thermal conductivity, high saturation velocity [1,2], high chemical stability, low thermal expansion and high thermal shock resistance [3,4]. SiC powder can be used as a source material for the growth of high-resistivity SiC single crystals through physical vapor transport technique (PVT).

The conventional method for preparing SiC powder is the Acheson process [5–9]. It is a carbothermic reaction of SiO_2 with carbon powder, lasting for about 1 week and the reaction occurs at high temperatures (2200–2400 °C). Due to the high reaction temperature and the long reaction time, the resultant SiC powder consists of larger particles. Therefore, extensive milling is needed to reduce the particle size to micron or sub-micron level and makes the powder suitable for sintering. Furthermore, the reaction of silica and carbon is a high exothermic reduction reaction accompanying the release of carbon monoxide and carbon dioxide. Accordingly, this method features the high energy consumption and the production of the poor purity powder.

Other SiC synthesis methods include pyrolysis of the organosilicon polymer [10–12], chemical vapor deposition (CVD) by the reaction of ethylene and silane [13–15], sol–gel [16–18], thermal plasma synthesis from rice hull [19], and so on. However, all above methods cannot meet the requirement of high purity and low cost product.

At present, combustion synthesis (CS) of silicon and carbon powder [20–25] has been regarded as an economic and effective method for the synthesis of high purity SiC powder. However, the synthesis of SiC from silicon and carbon powder has a low adiabatic combustion temperature, which needs an additional heating energy source, e.g., microwave energy and radio-frequency heating [26].

In this work, in order to synthesize high purity SiC powder, a radio frequency (RF) generator was used to

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supply an additional energy for the CS process. We studied the effects of the major reaction parameters such as the C/Si ratio, reaction time and reaction temperature on the phase composition, particle size and the purity of the obtained SiC powder. Interestingly, the critical ignition temperature (T_i) of silicon and carbon synthesis reaction was found to be about 1200 °C and the procedure of the temperature increase lasted for 30 min due to the exothermic reaction. Finally, the reaction mechanism was discussed in detail.

2. Experimental procedures

The raw materials used in the experiment mainly included fine silicon powder of 99.999% purity with particle size of 5–10 μm and carbon powder of 99.999% purity with particle size of 5–20 μm.

The SiC powder was synthesized by the combustion method. The synthesis procedure was carried out in a constant pressure reactor whose power was supplied by RF inductive device. The pressure of the atmosphere was controlled and kept a constant Ar flow by butterfly valve during the reaction. The whole reaction includes four stages: (I) preheating, (II) initial combustion reaction, (III) product formation, and (IV) cooling. The detailed process is described as follows. The reactant silicon and carbon powder were sealed into a PTFE milling tank to homogeneously mix for 24 h at room temperature, where the molar ratio of C/Si was precisely controlled. Then the graphite crucible of 10% porosity, charged by silicon and carbon powder within a loose state [27], was sealed and put into the RF furnace. The reaction chamber was firstly vacuumed and then flushed to 200 mbar with a constant Ar flow. Typically, the reaction chamber was heated to about 2000 °C after preheating (I) and initial combustion reaction (II) stages. Then, the synthesis reaction of silicon and carbon (III) occurred according to the formula of $Si_{(s)}+C_{(s)}=SiC_{(s)}$. After the reaction was completed, the chamber was cooled to room temperature (IV). The obtained SiC product phases include 3C-SiC, 6H-SiC and 15R-SiC.

The SiC phase structure was analyzed by X-ray diffraction with Cu K_a radiation and Raman spectroscopy with 532 nm laser. The product morphologies were observed by scanning electron microscopy (SEM, Hitachi S-4800 field-emission) with a 15 kV acceleration voltage and confocal laser scanning microscopy (CLSM) with 405 nm laser. The glow discharge mass spectroscopy (GDMS) was used to analyze the impurity content in the SiC powder.

3. Results and discussion

The calculated adiabatic temperature (T_{ad}) for the SiC synthesis was 1800 K. Therefore, the combustion reaction of SiC is impossible to achieve a self-sustaining combustion without an additional energy. In this research, a continuous energy was supplied to ensure the reaction of silicon and carbon powder by RF heating. SEM images of the C and Si powder raw materials were shown in Fig. 1(a) and (b), respectively. Carbon powder own flake morphology whereas the silicon grains aggregate together. After reaction, the obtained SiC powder grains are polygonal and the particle size is about 10–50 μm, as shown in Fig. 1(c).

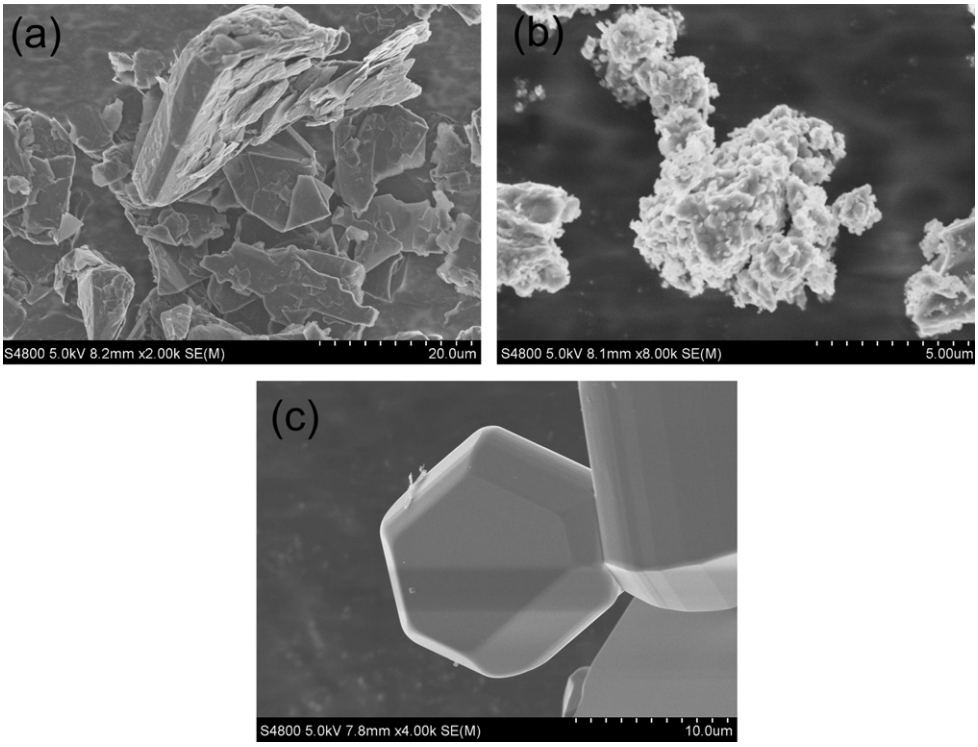

Fig. 1. SEM images of the raw materials and the synthesized SiC powder. (a) Carbon (b) Si and (c) SiC powder.

3.1. Effects of the C/Si molar ratio on the SiC powder

The influences of C/Si molar ratio on the ignition temperature and the reaction degree of the SiC synthesis were firstly studied. Fig. 2(a) shows the temperature profiles of the combustion reaction obtained on raw materials with different C/Si molar ratios, in which the partial heating program is given. A typical reaction process is described when C/Si molar ratio is fixed as 1:1. The reaction chamber was firstly preheated to 1185 ± 5 °C (I), and then the initial combustion reaction (II) was initiated and lasted about 2 h. In the stage II, after the ignition, the reaction temperature rapidly increased and passed through a transient steady state of about 20 min; as the reaction continued, the reaction temperature began to slightly go down about 15 °C accompanying the propagation of combustion wave. In the product formation stage (III), the reaction temperature was enhanced to the corresponding reaction temperatures, such as 1500 °C, 1700 °C, 1800 °C and 2000 °C, and lasted several hours to sustain the reaction and removed the metal impurities before a final cooling state (IV, not shown here).

As seen in Fig. 2(a), the ignition temperatures (T_i) appeared in a temperature range of 1160 °C∼1200 °C for all raw materials with different C/Si molar ratios after a preheating procedure. However, it seems that with the increase of the C content in the mixture, the ignition temperature increases and then passes through a maximum at C/Si ratio of 1.04:1. When the C/Si ratio increases to 1.1:1, the ignition temperature of this mixture appears at 1160 °C, which is a little lower than those of the other mixtures with lower C/Si ratios. The reaction degree in the initial combustion reaction, i.e. the transformation rate from Si/C mixture to the SiC powder, can be evaluated by the temperature difference between the ignition and the transient steady state in the initial combustion reaction stage [28]. Fig. 2b shows a direct comparison of the temperature differences (ΔT) with different C/Si molar ratios. In the Si-rich region, the temperature decreased from 135 ± 5 °C to 120 ± 5 °C with the increase of the C/Si ratio from 1:1.05 to 1:1. In the C-rich region, as the C/Si ratio increases from 1:1 to 1.1:1, the reaction undergoes the lowest temperature difference of 103 ± 5 °C at C/Si ratio of 1.02:1. Particularly, the temperature difference has a clear increase of about 35 °C over the C/Si molar ratio of 1.02:1. Therefore, it is proposed that the excessive Si and/or C content in the raw materials actually results in a larger temperature difference, further leading to a larger reaction degree especially in the initial combustion reaction stage. The relationship between reaction degree and temperature difference is described according to the following equation [28]:

$$\eta = \overline{C_P}(T - T_i)/Q \quad (1)$$

Where η is the reaction degree, $\overline{C_p}$ is the product average quality heat capacity, T is combustion temperature, T_i is the ignition temperature, and Q is reaction heat per unit mass.

Generally speaking, the C/Si ratio of 1:1 is a common parameter for the combustion synthesis to avoid the incomplete reaction. However, it is theoretically significant that a larger deviation from the C/Si ratio of 1:1 can produce a higher reaction degree in the initial combustion reaction stage. As mentioned above, the C/Si ratio has a large influence on the ignition temperature and the reaction degree for the initial combustion reaction stage. On the other hand, if the C/Si ratio is larger than 1.1:1, the residual carbon particles retain in the inner of the accumulated SiC powder, which may not be removed by heating at 700 °C under an oxygen atmosphere. Hence, an appropriate C/Si ratio is a key factor for the synthesis of high purity SiC powder.

3.2. Effects of the reaction temperature on the SiC phase structures

In order to study the influences of the reaction temperature, combustion reactions were performed at four

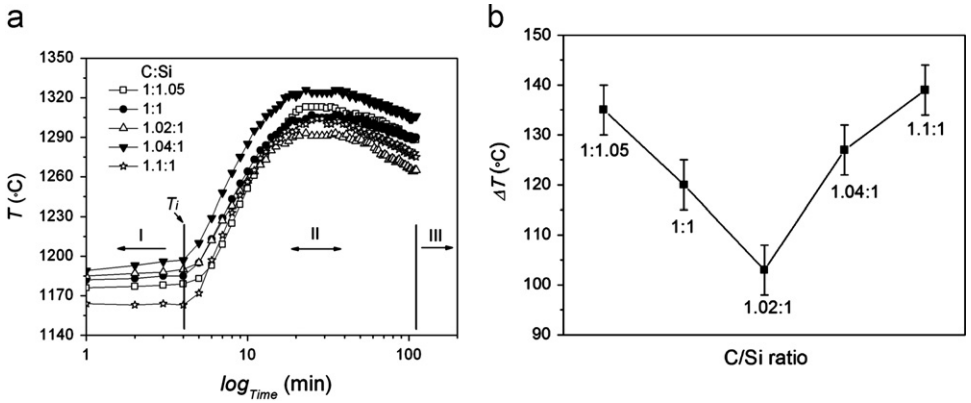

Fig. 2. Influence of C/Si molar ratio on the ignition temperature and the reaction degree. (a) Temperature profiles of the combustion reaction obtained on raw materials with different C/Si molar ratios. T_i points to the ignition temperature. I is the preheating stage, II is the initial combustion reaction stage lasting for about 2 h, III is the product formation stage for several hours in which the temperature is enhanced to reaction temperature, and cooling state (IV) is not shown here. (b) The temperature differences between the ignition and the transient steady state of the initial combustion reaction for the raw materials with different C/Si molar ratios.

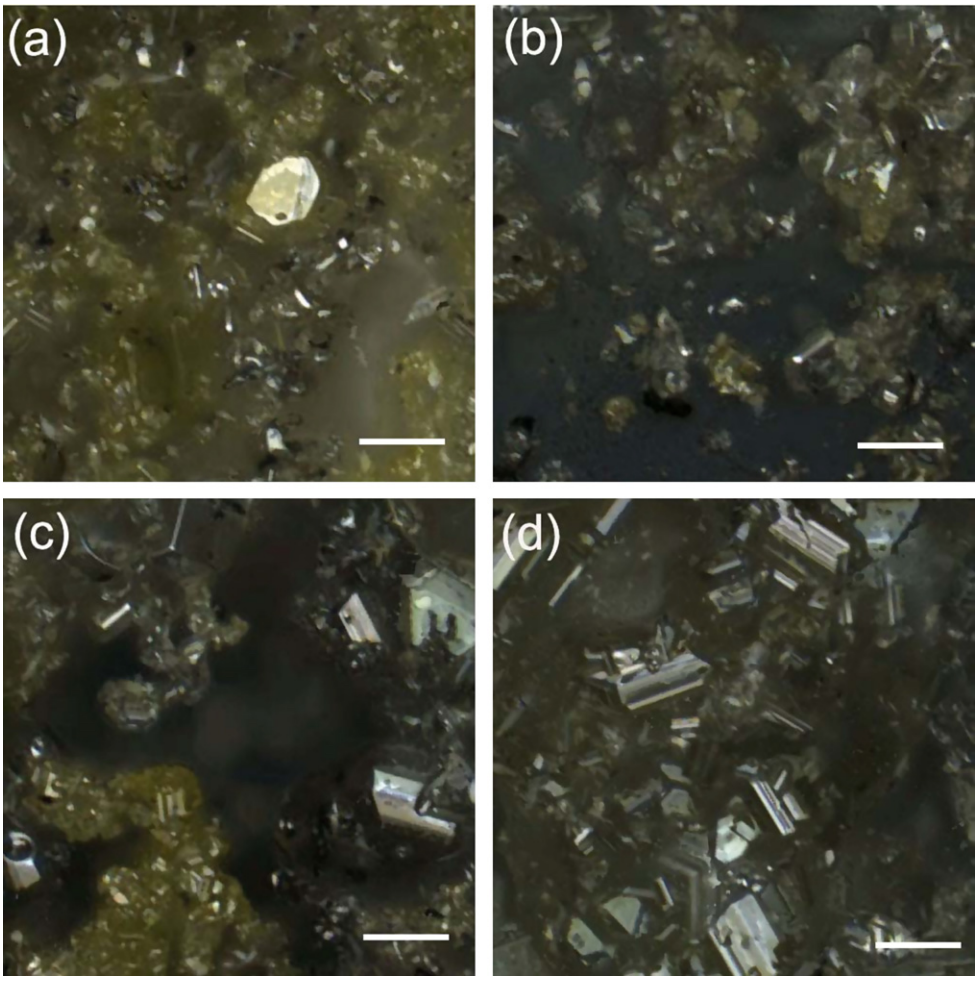

Fig. 3. Confocal laser scanning microscopic (CLSM) images of synthesized SiC powder products at different synthesis temperatures when the C/Si ratio was fixed at 1:1. (a) 1500 °C, (b) 1700 °C, and (c) 1800 °C, (d) 2000 °C. The scale bar is 50 μm.

different temperatures in the product formation stage for 4 h, and the C/Si ratio was fixed at 1:1. The morphologies of SiC powder were observed by confocal laser scanning microscopy (CLSM), as shown in Fig. 3. The color of SiC crystalline grain can be used to identify the phase structure of SiC. When the grain is yellow, the SiC product is mainly composed of β-SiC (3C), while the α-SiC (6H) grains exhibit colorless in case of without nitrogen doping [29]. At lower reaction temperatures, for instance, 1500 °C (Fig. 3a) and 1700 °C (Fig. 3b), the major phase of SiC powder is β-SiC with yellow color. At 1800 °C (Fig. 3c), the transparent grains with hexagonal structure are observed in the image, indicating the formation of α-SiC. When the temperature reaches to 2000 °C, the complete transition from β-SiC to α-SiC is achieved and very little β-SiC can be seen in the image (Fig. 3d).

Fig. 4 shows the XRD patterns of SiC powder products obtained at different temperatures. These SiC products were not further processed by the removal of excess graphite before XRD measurements. At 1500 °C, peaks of graphite, silicon and β-SiC were all observed, which indicated that the yield of SiC was rather low and the SiC synthesis reaction was not yet completed. As the reaction temperature increased to 1700 °C, the peaks of graphite disappeared and silicon still coexisted with β-SiC products, indicating a further conversion reaction with the increase of reaction temperature. When the temperature increased to 1800 °C, the XRD pattern revealed that the SiC product mainly consisted of the phase of β-SiC and only a small quantity of α-SiC was found in the product under this synthesis condition. Furthermore, the graphite impurity appeared in the product again because of the evaporation of Si at a higher temperature. It should be noted that the β-α transition is a kinetically controlled process, and a longer time or a higher temperature is usually needed to achieve complete conversion. As the temperature increased up to 2000 °C, only the phase of α-SiC was formed.

CLSM observations and XRD results together proved that, at lower temperatures such as 1500 °C, 1700 °C and 1800 °C, the β-SiC (3C) was the major phase in the SiC powder; whereas at 2000 °C, the α-SiC was the only phase of the SiC powder. α-SiC began to form at 1800 °C after heating for 4 h by radio-frequency heating when the C/Si ratio is 1:1. Meanwhile, at this temperature, the silicon was sublimated and the synthesized silicon carbide powder can decompose to form Si_2C, SiC_2, and so on. The excessive

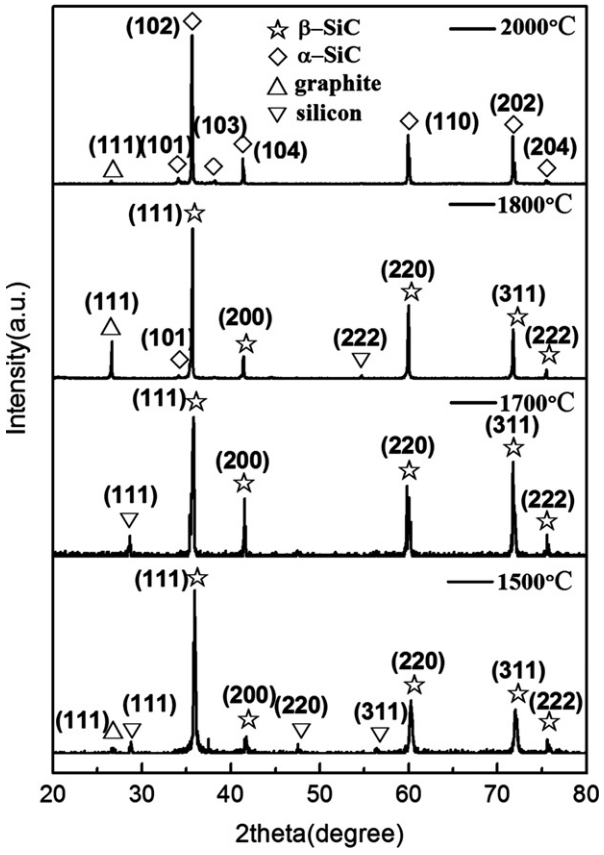

Fig. 4. X-ray diffraction patterns of SiC powder products synthesized at 1500 °C, 1700 °C, 1800 °C, and 2000 °C. The C/Si ratio was fixed at 1:1. The products were not processed by the removal of excess graphite and silicon before XRD measurements.

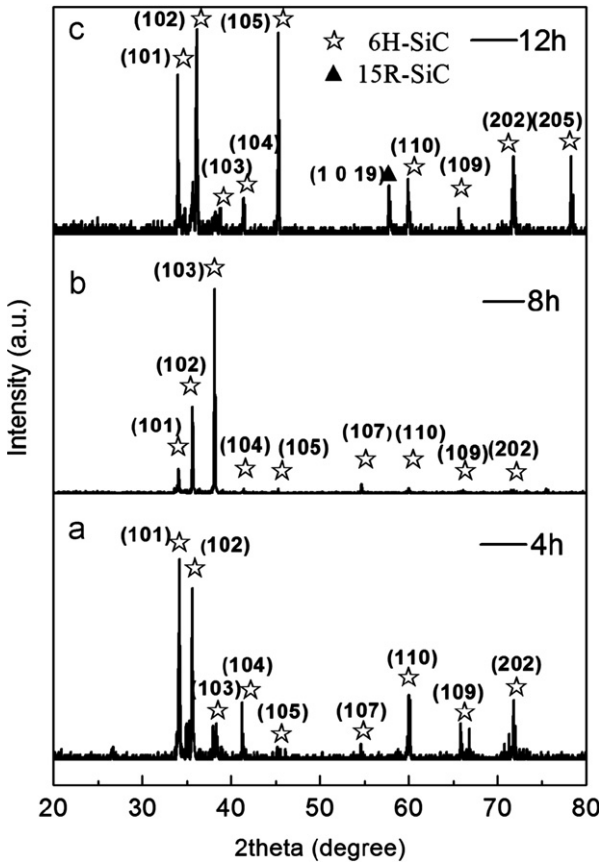

Fig. 5. XRD patterns of SiC powder products synthesized at 2000 °C with different reaction times when the C/Si ratio was fixed at 1:1. (a) 4 h, (b) 8 h and (c) 12 h. The products were further processed by heating procedure to remove excessive graphite before XRD measurements.

carbon powder was produced due to the thermal decomposition with the elapsed time. Finally, high purity SiC powder can be facilely obtained after the residual graphite was removed by means of heating at 700 °C. Anyway, the reaction temperature is an important influence factor on the purity and the phase structure of the SiC powder.

3.3. Effects of the reaction time on the structures and sizes of SiC particles

In order to study the influences of the reaction time on SiC powder products, combustion reactions were performed at 2000 °C in the product formation stage for different reaction times. The C/Si ratio was still fixed at 1:1 and all the SiC products were further processed by heating procedure to remove excessive graphite before XRD measurements. XRD patterns of SiC powder products further indicated that a complete β-α conversion occurred at 2000 °C and the product predominantly consisted of α-6H-SiC. However, the phase structure and the grain morphology of the powder product also varied with the reaction time. After 4 h reaction, the XRD pattern (Fig. 5a) of powder product exhibits several diffraction peaks that are readily indexed as (1 0 1), (1 0 2), (1 0 3), (1 0 4), (1 0 5), (1 0 7), (1 1 0), (1 0 9), and (2 0 2) of α-6H-SiC. After 8 h, the obtained SiC powder was still α-6H-SiC. From the corresponding X-ray diffraction pattern shown in Fig. 5b, it was found that (1 0 3) diffraction intensity obviously increased, which implied that the (1 0 3) plane was possibly one of the main appearance planes. When the reaction time increased to 12 h, an obvious peak located at (1 0 19) was observed, which indicated the formation of α-15R-SiC. Meanwhile, the grains grew much larger and all diffractions became much stronger.

The Raman spectra of the SiC powder synthesized at different reaction times were further used to determine the phase structures of the products, as shown in Fig. 6. All the Raman spectra show the presence of sharp peaks at 786 cm^{-1} and 965 cm^{-1}, which correspond to transversal optical (FTO) phonon mode and longitudinal optical (FLO) phonon mode of α-SiC [30], respectively. No visible characteristic peaks of β-SiC appeared in the spectra. The peaks at 147 cm^{-1} (FTA mode) were also present in each Raman spectrum, indicating the presence of the α-6H-SiC. When the reaction time was extended from 8 h to 12 h, an obvious peak at 173 cm^{-1} confirmed the formation of α-15R-SiC [31]. These results prove that the α-*15R*-SiC can

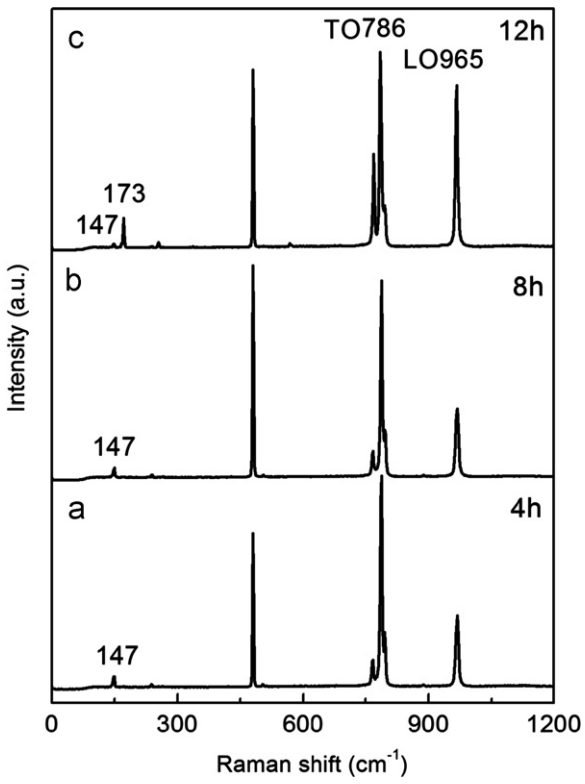

Fig. 6. Raman spectra of SiC powder products synthesized at 2000 °C with different synthesis times when the C/Si ratio was fixed at 1:1. (a) 4 h, (b) 8 h and (c) 12 h. The peaks at 480 cm^{-1} come from the disturbance of the fluorescent light.

be synthesized at 2000 °C for 12 h by RF heating-assisted combustion synthesis. More important, the phase structure of SiC is dominated by the reaction temperature and time together during combustion synthesis.

Particle sizes and morphologies of the synthesized products at different reaction times were analyzed. The SEM images of the SiC powder products were shown in Fig. 7. It is obvious that the particles of SiC powder mainly exhibit hexagonal morphology in all images. Fig. 7a and b shows that the SiC powder synthesized after 4 h has a wide size distribution ranging from 5 μm to 100 μm. Among these particles, the smaller particles with the size of about 5–30 μm occupy the majority in the products. After 8 h, the size of the SiC powder obviously increased and had a wide size distribution from 10 μm to 150 μm in which the particles with the size of about 100 μm occupied the absolute majority (Fig. 7c and d). In addition, it seems that the smaller SiC particles are composed of very fine grains that aggregate to form agglomerates. When the reaction time was extended to 12 h, the particle size further increased up to 50–200 μm (Fig. 7e). It is obvious that long time synthesis will result in partial fusion of small particles to form bigger particles (Fig. 7f).

In Fig. 7f, the small particles adhered to the bigger particles and formed much bigger agglomerate. The whole Si–C reaction process is shown in Fig. 8. SiC formation from the reaction of Si and C can occur at a lower temperature. In this case, the obtained SiC powder was not created directly from Si–C reaction. Because Si and C powder were uniformly mixed at initial stage, the particles of Si and C would be adsorbed with each other (Fig. 8 I and II) to form SiC (reaction (2)) in a solid–solid reaction at a lower temperature. With the increase of reaction time, some of the synthesized SiC solids gradually decomposed to generate C solid and gaseous phases of Si, Si_2C, SiC_2, and so on (reactions (3), (4), and (5)) as intermediate products. These intermediate products continued to re-react and fuse. Consequently, Si and SiC_2 reacted with C in a gas–solid reaction and SiC_2 reacted with Si in a gas–gas reaction to form inhomogeneous particles (reactions (6), (7), and (8)). The above reaction processes were roughly summarized as step III in Fig. 8. The Si and C reactions are:

$$Si_{(s)} + C_{(s)} = SiC_{(s)} \quad (2)$$

$$SiC_{(s)} = Si_{(g)} + C_{(s)} \quad (3)$$

$$2SiC_{(s)} = Si_2C_{(g)} + C_{(s)} \quad (4)$$

$$2SiC_{(s)} = SiC_{2(g)} + Si_{(g)} \quad (5)$$

$$Si_{(g)} + C_{(s)} = SiC_{(s)} \quad (6)$$

$$Si_2C_{(g)} + C_{(s)} = 2SiC_{(s)} \quad (7)$$

$$SiC_{2(g)} + Si_{(g)} = 2SiC_{(s)} \quad (8)$$

The successive reactions can repeatedly occur between the SiC solids and the intermediate products until the temperature is lowered down to room temperature. The above procedure directly causes the smaller particles to fuse and incorporate into bigger particles (Fig. 8 IV). The powder particle size increases with the reaction time due to the repeated processes (Fig. 8 V) of formation, decomposition, fusion and reformation. Therefore, the reaction time is a key influence factor on the phase structure and the particle size of SiC powder in the combustion synthesis.

3.4. GDMS analysis of the synthesized SiC powder

The composition of high purity SiC powder was analyzed by glow discharge mass spectroscopy (GDMS) and the results were listed in Table 1. It is well known that boron and aluminum elements are the main shallow acceptor impurities in SiC single crystals. In order to grow high purity SiC single crystals, it is necessary to reduce the boron and aluminum concentrations in the SiC powder source. The impurities could evaporate from the graphite crucible at the high reaction temperature. Compared with the results in other literatures [32–34], the contents of the boron and aluminum elements in the powder product at present work are much lower by one to two orders of magnitude. As for other metallic elements, the synthesized SiC powder also exhibits obviously comparability or even improvements in purity. Thus the synthesized powder can be directly used as a source material for high-resistivity SiC single crystal growth.

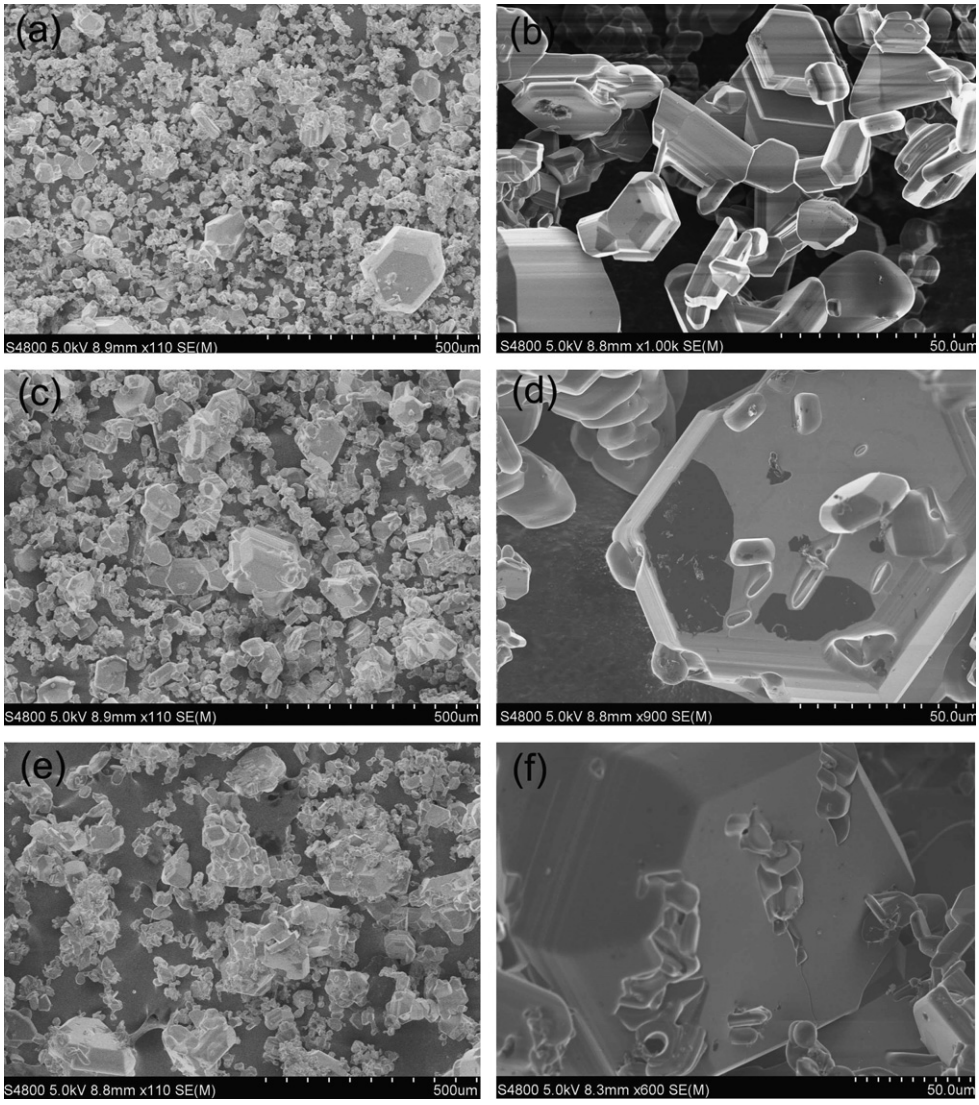

Fig. 7. SEM images of SiC powder products at 2000 °C synthesized for different synthesis times. (a) 4 h, (c) 8 h and (e) 12 h. The corresponding magnified images were shown in (b), (d), and (f), respectively.

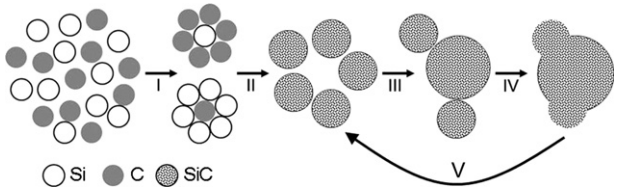

Fig. 8. Si–C reaction process of SiC formation with the elapsed reaction time.

Table 1
GDMS analysis of the synthesized SiC powder products.

	Element/ppm					
	B	Na	Al	S	Fe	Ti
Synthesized powder	0.11	0.24	0.26	0.76	0.27	0.50
Literature [32]	0.40	0.19	2.0	0.85	0.44	0.57
Literature [33]	3.90	–	19.0	–	–	–
Literature [34]	5.00	0.60	9.0	0.30	2	75

4. Conclusions

An effective combustion synthesis method for the production of high purity SiC powder product was presented in this paper. We used a radio frequency heating device to supply the energy for ignition and sustaining of combustion reaction. The influences of the C/Si ratio, reaction temperature and reaction time on the size, composition and structure of the SiC powder were investigated. The following conclusions can be drawn:

(1) Combustion synthesis of high purity SiC powder by RF heating can be performed in four stages: (I) preheating, (II) initial combustion reaction, (III) product formation and (IV) cooling.

(2) The initial combustion reaction started from the ignition temperature (T_i) at near 1200 °C following a preheating procedure. The larger increase of the C content in the mixture seemed to considerably lower T_i, e.g., the C/Si ratio of 1.1:1.
(3) A larger deviation from the C/Si ratio of 1:1 can cause a larger temperature difference, further leading to a higher reaction degree especially in the initial combustion reaction stage.
(4) The reaction temperature and the reaction time at the product formation stage together determine the crystalline phase of the synthesized SiC powder. With the increase of reaction temperature and the reaction time, the transitions from 3C to 6H and from 6H to 15R were found, respectively. As a result, high purity 6H-SiC powder without Si impurity can be synthesized at 2000 °C for 4–8 h.
(5) A kinematic reaction process was found in the combustion synthesis of SiC powder. Longer reaction time causes the growth of the SiC grains due to repeated processes of formation, decomposition, fusion and reformation.

Acknowledgments

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