Effect of silicon source and carbon source on the morphology of SiC powders

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Abstract. Silicon carbide (SiC) powders were prepared by carbothermal reduction method with silica fume and carbon-white as the silicon source; glucose, phenolic resin, polyvinyl pyrrolidone (PVP) as carbon source. The effects of reaction temperature, silicon source and carbon source on the morphology of silicon carbide powder were investigated. The result shows that: The optimum temperature for preparation of silicon carbide powder was 1400 °C; When the glucose was used as carbon source, the morphology of SiC powder was greatly affected by the kinds of silicon sources; When the phenolic resin and PVP were used as carbon source, the morphology of SiC powder was greatly influenced by the carbon source.

1. Introduction
Silicon carbide (SiC) ceramic has excellent mechanical properties, oxidation resistance, wear resistance, thermal stability, thermal shock resistance and chemical corrosion resistance, and the thermal expansion coefficient is small, thermal conductivity is large [1-6]. Different size and morphology of SiC powders can be used to prepare different types of SiC ceramic products. Therefore, the study of the influence factors of SiC powder morphology has high practical significance and market value.

In this paper, SiC powder was prepared by carbothermal reduction method with different silicon source and different carbon source, the effects of reaction temperature, silicon source and carbon source on the morphology of silicon carbide powder were investigated. It has a certain guiding significance to the actual production of silicon carbide powder.

2. Experimental
2.1. Reagents and Instruments
Reagents: silica fume, Tech, Gansu Sanxin silicon industry limited company; carbon-white, Tech, Weifang Sanjia Chemical Co. Ltd.; glucose, AR, Chongqing Heping Pharmaceutical Co. Ltd.; phenolic resin, AR, Tianjin United Chemical Industry Co. Ltd.; polyvinyl pyrrolidone (PVP), AR, Henan Zheng Yao chemical products Co. Ltd.
Instruments: Analytical balance (08F106-42), Electrothermal constant temperature blower drying box(DHG-9076A), Tubefurnace(SK2-6-12A), Muffle furnace(LHT807GN4G), Horizontal vacuum sintering furnace(ZT-45-22W).
2.2. Sample Preparation
Frist of all, the carbon source was dissolved in distilled water, and then added a certain amount of silicon source, ball milled the slurry at high speed for 2 hours. Then put the slurry in the oven under 100 °C for 24 h. After the slurry was dried and pulverized, put the powder into a tubular furnace heated to 650 °C for 2 h under nitrogen atmosphere to prepare precursor, the heating rate was 5 °C /min. The precursor was heated to reaction tempeture under vacuum for 2 h. The synthesized powders were washed with hydrofluoric acid (HF) to remove unreacted silica and other impurities, then heated the powders to 650 °C in atmosphere for 2 h to remove the residual carbon in the product. Finally, the SiC powder could be obtained by washing, filtering and drying.

2.3. Sample characterization
The phase composition and crystallinity of the materials were characterized by powder XD-5A x-ray diffraction (XRD) which was produced by Japan Island Company. Microstructures of the products were observed on a JSM-5510LV Scanning Electron Microscope (SEM) which was produced by Japan.

3. Results and Discussion

3.1. Effect of reaction temperature on the morphology of silicon carbide
The X- ray diffraction (XRD) analysis of the product was carried out at as shown in Figure 1. The reaction conditions was that: The micro silica fume used as the silicon source and glucose used as the carbon source under the 1300 ºC, 1400 ºC, and 1500 ºC for 2 h.

![Fig.1 XRD patterns of the products at different temperatures](image)

At 1300 ºC, 1400 ºC, and 1500 ºC calcined 2 h, the obtained products were 3C SiC, the crystalline SiC belongs to β-SiC. From the intensity of diffraction peaks of 3 kinds of samples, the diffraction peak of the product at 1500 ºC was the strongest, followed by 1400 ºC, and the diffraction peak of the 1300 ºC product was the weakest. At 1300 ºC, the crystallinity of the product was not high, and with the increase of temperature, the diffraction peak became sharp. It was known that the crystallinity of the product was becoming better, and the crystal was gradually developed.

The SEM test on three kinds of temperature on the morphology of the products were shown in Figure 2.
As shown in Fig. 2(a), when the reaction temperature was 1300 °C, the sample consisted of some amorphous SiC particles, mutual adhesion between particles, particle size was about 100 nm. It can be seen from Fig. 2(b) that the sample particles were obvious, particles were connected to each other, high sphericity, single particle size was about 200 nm. It can be seen from Fig. 2(c) that the silicon carbide particles obviously grew up, the large particles showed sharp edges and corners, the morphology of some small particles were still nearly spherical. In contrast to 1300 °C and 1400 °C, the particles grew obviously and the sphericity became worse. At this temperature, the SiC crystal was the most complete and the particles were growing up, the above results were in agreement with the XRD data. Therefore, in this experiment, the optimum temperature for preparation of silicon carbide powder was 1400 °C.

3.2. Effect of silicon sources on silicon carbide morphology

The experiment used silica fume and carbon-white two silicon source, used glucose as carbon source, the SiC powder was prepared under the temperature 1400 °C for 2 h, and conducted SEM tests on raw materials and products, the test results were shown in Figure 3.

As shown in Fig.3, when the glucose was used as carbon source, the morphology of SiC powder was close to that of silicon source, this may be because during the experiment, the glucose could be completely soluble in water, the silicon source and the carbon source could be mixed evenly. In the carbonization process, the carbon would be wrapped in silicon source around to form a coating layer, under the reaction temperature, the silicon source gradually turned into a molten silicon source, and the carbon was gradually dissolved into the silicon source to form silicon carbide powder. In the process, silicon source to melt and the carbon source was gradually dissolved, leading to the the reaction to continue, eventually lead to silicon carbide inherited silicon source morphology.

3.3. Effect of different carbon sources on the morphology of silicon carbide

The experiment used silica fume as silicon source, changed the types of carbon source to study the effects of different carbon sources on the morphology of silicon carbide. Conducted SEM test on the product and the test results were shown in Figure 4.
glucose as carbon source
phenolic resin as carbon source
PVP as carbon source

The silicon carbide powder of different carbon sources were fabricated by SEM analysis, it can be seen that: Different carbon sources lead to different morphologies of SiC, when glucose was used as carbon source, SiC was spherical and granular; when phenolic resin was used as carbon source and SiC was flocculent; when PVP was used as carbon source, SiC was amorphous powder. This may be due to phenolic resin and PVP as carbon source, its solubility in water was not as good as glucose, resulting in mixed carbon source and silicon source was not uniform, in the process of carbonization, the carbon source could not completely cover the silicon source. In the reaction process, the silicon source was melted and flowed in the reaction system, a portion of the silicon source flowed to the surface of carbon source and reacted with the carbon to form SiC. At present, the silica fume unable to maintain its original shape, so the product could not inherit the morphology of the silicon source, but the shape of the carbon source determined the morphology of the product.

4. Conclusions
(1) The optimum temperature for preparation of silicon carbide powder was 1400 °C;
(2) When the glucose was used as carbon source, the morphology of silicon carbide inherits the morphology of silicon source;
(3) When the phenolic resin and PVP were used as carbon source, the morphology of silicon carbide is flocculent, and the morphology of silicon source is not well inherited.

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References
[1] A. Najafi, F. Golestani-Fard, H. R. Rezaie, N. Ehsani, J. Sol-Gel Sci. Technol. 59, 205-214 (2011).
[2] Solomah, A. C., Reichert, W., Rondinella, V., Esposito, L., & Toscano, E. (2010). Journal of the American Ceramic Society, 73(3), 740-743.
[3] Perham, T. Office of Scientific & Technical Information Technical Reports (1996).
[4] Chuan-Jun, T. U., et al. Transactions of Nonferrous Metals Society of China 25.3 (2015): 856-862.
[5] Okuno, Akiyasu, and M. Watanabe. US, US 4946807 A. 1990.
[6] Lee, Jin Seok, D. M. Choi, and S. C. Choi. Solid State Phenomena 124-126 (2007): 747-750.