Journal of Nuclear Materials 386–388 (2009) 631–633

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

Design and installation of DC plasma reactor for SiC nanoparticle production

I.K. Yu [a], J.H. Rhee [a,*], S. Cho [a], H.K. Yoon [b]

[a] Materials Research Team, National Fusion Research Center, Daejeon 305-333, South Korea
[b] Division of Mechanical Engineering, Dong-Eui University, Busan 614-714, South Korea

A B S T R A C T

In order to get the high quality SiC nanopowders, a DC plasma reactor system with adjustable torch has been developed. SiC nanopowders were synthesized using this system and the synthesized primary particles have nearly spherical structures, mostly β-SiC phase with a particle size of 10–30 nm. Larger aggregate particles have been produced in our system probably due to longer particle growth times and faster collisions. The synthesized particles collected from the reactor wall and cyclone bottom have some free silicon and free carbon. To produce high quality silicon carbide nanopowders, it is highly necessary to improve the experimental conditions such as lower system pressures, shorter residence time, and higher quenching rates during powder synthesis.

1. Introduction

Recently, silicon carbide (SiC)-composites produced by silicon carbide nanoparticles have been considered as one of the prominent candidates for blanket materials in nuclear fusion. SiC provides outstanding oxidation resistance and mechanical properties at high temperatures even above the melting point of steels. Also SiC powders have a wide range of industrial applications such as raw materials, abrasives, additives, etc. It is highly required for SiC-based materials to enhance the fracture toughness, high temperature creep strengths, and swelling resistance, especially for fusion applications [1,2]. To improve the mechanical properties of SiC monolithic and composites, high quality nanopowders have been used for the better sinterability and smaller grain sizes. It has been widely considered that nanopowders can provide better mechanical properties primarily due to the higher specific surface areas and surface activities compared with those of micron powders [3–5]. In this paper, we will report the results of design and installation of DC plasma reactor system and characterization of the synthesized silicon carbide nanopowders.

2. Experimental

The custom-made DC plasma reactor system used for the synthesis of SiC nanopowders is shown in Fig. 1. Basically, this synthesis system has been designed and built to use thermal decomposition of target materials and SiC nanopowder formation reactions during operation of thermal arc plasma. Under certain vacuum conditions, these activated silicon and carbon atoms in the plasma space will interact with each other in the reactor chamber and then can form SiC nanopowders to decrease total surface energy and therefore reach more stable β-phase crystal structure. This system is composed of a DC plasma torch, a DC power supply (60 kW), a cylindrical stainless steel reactor with water cooling, a cyclone, a bag filter, and a dry scroll pump for vacuum. The position of the plasma torch can be adjusted vertically up to about 20 cm. The starting materials used mainly in this experiment were silicon carbide bars with about 10 mm outer diameters and about 10 cm lengths. Typical synthesis experiments were operated at system pressures of about 300–400 Torr, Ar plasma gas flow rates of 15 lpm (L/min), Ar quenching gas flow rates of about 15–30 lpm, arc current of about 80–150 A, and distances of about 10–20 mm between the torch exit and target materials.

For SiC nanopowder synthesis, three operation parameters have been carefully decided to optimize plasma conditions and to improve crystalline nanopowders quality; arc current, working pressure, flow rates of Ar quenching gas. Higher arc current can increase plasma temperature and thus enhance thermal decomposition rates of target materials and SiC nanopowder synthesis reactions. But, sometimes, it does not work properly and make serious damages on target, such as, big cracks and fracture. Higher vacuum pressure can improve the crystallinity of as-grown SiC nanopowders but restrict the amount of synthesized nanopowders and flow rates of Ar quenching gas in our plasma conditions. The temperature of thermal arc plasma has not been experimentally measured in our reactor system due to extremely high plasma temperature. Type K thermocouple is located at about 15 cm away from DC plasma torch and temperature of plasma reactor chamber has been measured at about 290–350 °C under certain vacuum conditions during nanopowder synthesis experiments, indirectly.

* Corresponding author. Tel.: +82 42 870 1747; fax: +82 42 870 1638.
E-mail address: jhrhee@nfri.re.kr (J.H. Rhee).

0022-3115/$ - see front matter © 2009 Published by Elsevier B.V.
doi:10.1016/j.jnucmat.2008.12.230

Fig. 1. DC plasma reactor system for the synthesis of SiC nanopowders.

It takes several minutes to stabilize DC arc plasma and reactor pressure by setting Ar quenching gas flow rates and adjusting the exhaust valve in the operating procedures. After plasma ignition, thermal decomposition of target materials has been started and then evaporated silicon and carbon atoms have begun to form crystalline SiC nanopowders under vacuum pressures at about 300–400 Torr. SiC nanopowders have been synthesized for about 20–30 min in the plasma reactor, and after plasma shutdown, cooling procedure has been done for about 30 min in our vacuum chamber, and then dry-air venting procedure has been started in the final stages of operation. The quenching rates of synthesized nanopowders have not experimentally determined yet and possibly can be measured after upgrading system components and reactor performance.

After synthesis experiments, the synthesized nanopowders were collected from the reactor wall and cyclone bottom of the plasma reactor system. A dark grey or black color has been observed in the collected nanopowders probably due to excess free carbon. In the energy dispersive X-ray analysis (EDX) and Bruker D8 X-ray diffraction (XRD) measurements, the excess free carbon and free silicon have been measured and estimated about 20.8% and 10.4% in the synthesized SiC nanopowders, respectively. There are three major XRD peaks positioned at 2θ angles of about 35.6°, 59.9°, and 71.8°, assigned to the (1 1 1), (2 2 0), and (3 1 1) planes of the cubic β-SiC structures, respectively. Hitachi S4800 scanning electron microscope (SEM) image shows that primary particles have nearly spherical structures, mostly β-SiC phase with a particle size of 10–30 nm and larger aggregate particles have also been observed. Figs. 2 and 3 show the XRD patterns and SEM images of the synthesized SiC powders, respectively.

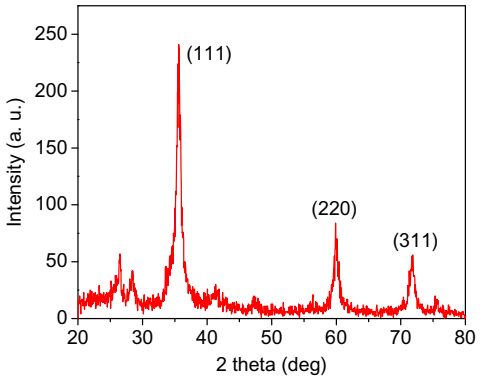

Fig. 2. XRD patterns of the synthesized SiC powders.

3 Results and discussion

Spherical polycrystalline SiC powders have been synthesized and then characterized by using XRD and SEM to evaluate the performance of the custom-made plasma reactor system. Based on these measurements, the primary particles could be formed from the evaporated silicon and carbon atoms after thermal decomposition of SiC target materials. Some of silicon and carbon atoms did not react with each other and thus not participate in the formation of polycrystalline silicon carbide nanopowders, which could induce the excess free silicon and free carbon during powder synthesis as estimated by the EDX and XRD measurements. After nucleation and growth of the primary particles, larger aggregate particles have been produced in our system probably due to lower quenching rates, faster particle-to-particle collisions, longer particle growth times, etc.

As thermal decomposition and vaporization rates increase, the atomic concentrations of vaporized silicon and carbon atoms increase around DC arc plasma, the accidental collision rates of these reactive atoms will increase rapidly, and also there is certainly enough possibility to form Si–C chemical bonds due to activation energy or driving force supplied by thermal plasma energy. Newly formed SiC nanoparticles have much higher specific surface areas and surface energy than those of micron-sized particles. Therefore it is likely to minimize their specific surface areas and surface energy by aggregating nanoparticles and forming larger aggregates during synthesis experiments. It is somewhat difficult to measure and analyze the intermediate nanoparticles in each growth stage. Therefore no experimental data has been obtained in the intermediate SiC nanoparticles of our reactor system. As shown in Fig. 3, SEM images clearly show the spherical-shaped SiC nanoparticles

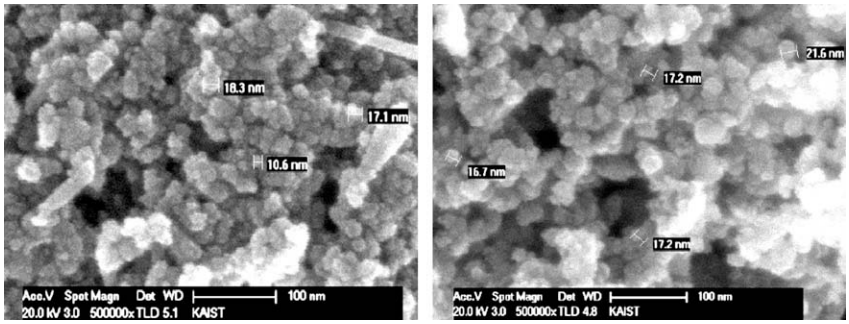

Fig. 3. SEM images of the synthesized SiC powders.

and larger aggregates at system pressure of about 350 Torr as described previously.

It is widely considered that the formation of ultrafine SiC nanopowders can be possible under higher quenching rates, shorter particle growth times, and swirling turbulent flow patterns in the nanopowder synthesis system [6–12]. To improve the SiC synthesis efficiency and minimize the larger aggregate particles, the coarsening and aggregations of the primary particles should be controlled by using the quenching methods and system pressures during nanoparticle growth. Argon gas quenching methods maybe possible in the transverse or counter-flow directions to increase the swirling turbulence and quenching rates of synthesized SiC powders under certain vacuum pressures [6,7]. Further investigations are necessary to reduce the amounts of free carbon and free silicon in the synthesized powders and thereafter to produce high quality SiC nanopowders.

4. Conclusions

Spherical polycrystalline β-SiC nanopowders were synthesized by using a plasma reactor system and the primary particles have a particle size of 10–30 nm. Larger aggregate particles have been produced probably due to longer particle growth times and faster collisions. The synthesized SiC powders collected from the reactor system have excess free silicon and free carbon. To produce high quality SiC nanopowders, it is highly necessary to improve the experimental conditions such as lower system pressures, shorter residence time, swirling turbulent flow patterns, and higher quenching rates during nanopowder synthesis.

Acknowledgements

The authors would like to thank Dr W.T. Ju and Professor Y.W. Kim for their technical advices in the plasma reactor system and nanopowder synthesis. This work was supported by the Korea Research Council of Fundamental Science and Technology, Republic of Korea.

References

[1] Y. Katoh, L.L. Snead, C.H. Henager Jr., A. Hasegawa, A. Kohyama, B. Riccardi, H. Hegeman, J. Nucl. Mater. 367–370 (2007) 659.
[2] R.H. Jones, L. Giancarli, A. Hasegawa, Y. Katoh, A. Kohyama, B. Riccardi, L.L. Snead, W.J. Weber, J. Nucl. Mater. 307–311 (2002) 1057.
[3] A.R. Raffray, R. Jones, G. Aiello, M. Billone, L. Giancarli, H. Golfier, A. Hasegawa, Y. Katoh, A. Kohyama, S. Nishio, B. Riccardi, M.S. Tillack, Fus. Eng. Des. 55 (2001) 55.
[4] Y. Katoh, A. Kohyama, T. Nozawa, M. Sato, J. Nucl. Mater. 329–333 (2004) 587.
[5] S.J. Zinkle, Fus. Eng. Des. 74 (2005) 31.
[6] J.-G. Li, M. Ikeda, R. Ye, Y. Moriyoshi, T. Ishigaki, J. Phys. D: Appl. Phys. 40 (2007) 2348.
[7] P. Kong, T.T. Huang, E. Pfender, IEEE Trans. Plas. Sci. PS-14 (1986) 357.
[8] R. Bandyopadhyaya, A.A. Lall, S.K. Friedlander, Pow. Technol. 139 (2004) 193.
[9] S.-M. Oh, M. Cappelli, D.-W. Park, Kor. J. Chem. Eng. 19 (2002) 903.
[10] N.P. Rao, N. Tymiak, J. Blum, A. Neuman, H.J. Lee, S.L. Girshick, P.H. McMurry, J. Heberlein, J. Aerosol Sci. 29 (1998) 707.
[11] N. Rao, B. Micheel, D. Hansen, C. Fandrey, M. Bench, S. Girshick, J. Heberlein, P. McMurry, J. Mater. Res. 10 (1995) 2073.
[12] J.Y. Guo, F. Gitzhofer, M.I. Boulos, Plas. Chem. Plas. Proc. 17 (1997) 219.