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Comminution of silicon carbide powder in a planetary mill

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Abstract

Silicon carbide powder (SiC) was comminuted in a planetary mill during time intervals of 0.5, 2, 4 and 6 h. The wet milling media was ZrO_2 spheres in isopropyl alcohol. The powders were then characterized with respect to chemical composition, particle size distribution, surface area and density for each milling time. The average particle size was reduced from 1.8 μm to 0.4 μm in 30 min and the amount of ZrO_2 increased linearly with milling time. The result was a homogeneous combination of submicrometer and nanometric SiC+ZrO_2 powder, which possessed good sinterability in liquid phase and high fracture toughness.

Keywords: Comminution; Silicon carbide; Planetary mill; Ultra-fine powder; Nano-powder

1. Introduction

The properties of an advanced ceramic product depend upon the purity of the raw materials, the green body density and the sintering process used. Highly covalent ceramics, such as carbides and nitrides, have inherent low sinterability as a consequence of the atomic bond. A very efficient way to increase density is through the reduction of the particle size, tight distribution and increase of surface area, which strongly influence the overall processing route and the final properties of the product [1,2].

A large number of mechanical methods are available for preparing fine and ultra-fine powders. Typical high-energy processes include planetary, attritor and jet milling [2]. The jet grinding process is the collision among particles propelled by a high-pressure gas jet, which has the main virtues of preventing contamination, precise mean particle size and tight distribution. The drawbacks are elevated loss of product; requires a high-quality classification system for fine particles; high equipment costs; and the presence of electrostatic charges can be a problem [3,4]. On the other hand, the planetary and attritor milling processes are via mechanical shock between the grinding bodies and powder particles in a wet media. The characteristic of both processes are low material loss; the cost of the equipment is significantly lower than jet milling; very good homogenization and particles with high superficial area are easily generated in a very short time; however, contamination may occur [4,5].

The work of Brito [6], which compares the planetary and attritor mill in reducing silica particle size, shows the planetary mill to be more efficient, even though no data on contamination have been reported. Tavares et al. [7], studying the influence of seven types of milling media on the comminution of a silicon carbide powder using a planetary mill, showed that sodium pyrophosphate was the most efficient media and ethyl alcohol was the least efficient, and for short periods of time (about 2 h) the mechanical effect was preponderant over the chemical one, but no data concerning contamination and surface area were reported.

Dense silicon carbide is an advanced ceramic in high demand and very small particles are needed to fulfill this goal. The understanding of the comminution process of SiC is very important but data in literature are difficult to find; for instance, the ISI WEB of Science database shows approximately 300 articles published about high-energy milling since 1960, but only one involving silicon carbide. To contribute to the knowledge of milling SiC, this study was done in a planetary mill and the results showed the process to be very fast in reducing the

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Table 1
Chemical composition of the received powder

Composition	wt.%
SiC	98.17
$Si+SiO_2$	0.60
Fe	0.30
Al	0.19
Free carbon (CL)	0.20
S, Ca, V, Ni, Cu and Zr	Trace

particle size and increasing the surface area, with a tight distribution; on the other hand, improvements must be made to reduce contamination.

2. Materials and methods

The silicon carbide grade SiC-1000 (Alcoa S.A.- Brazil) produced by the Achesson process was used as the raw material. The chemical composition is specified in Table 1 and the physical characteristics measured were mostly alpha phase, bimodal particle size distribution (d_{99} and d_{50} are, respectively, equal to 14.22 and 1.77 μm), density of 3.211 g/cm^3 and superficial area of 3.483 m^2/g.

The milling process was done in a planetary mill (PM-4 model, Retsch). The jars used were specially fabricated to support the elevated wear and to comminute 120 g of material. They were made of stainless steel with an inner diameter of 100 mm, 500 ml capacity and internally coated with tungsten carbide (WC–Co) by HVOF (High-Velocity Oxygen Fuel). The milling conditions were 300 rpm, isopropyl alcohol P.A. (IPA), zirconia milling balls stabilized with ceria (80% ZrO_2+20% CeO_2, Zirconox®, Netsch) with a diameter in between 0.7 and 1.2 mm, density of 6.1 g/cm^3 and Vickers hardness of 1200 kgf mm^2.

The milling media occupied half of the jar volume (250 ml), where 60% of this volume (150 ml) was filled by the grinding balls and the other 40% (100 ml) composed of the pulp (100 g of SiC powder and IPA). The amount of IPA varied a little, since the milling balls had to be slightly covered by the IPA. The milling was done in batches for time periods of 0.5, 2, 4 and 6 h. For periods longer than 6 h, the milling efficiency does not compensate the energy spent, as verified by Matos [8]. The materials were then poured into a Pyrex container, dried in an oven at 70 °C for 24 h, deagglomerated in a mortar and sieved.

The powder of each batch was then characterized by X-ray fluorescence spectrometer (Philips PW 2400), particle size distribution (Malver Mastersizer Micro Plus, MAF 5001), superficial area by BET (Gemini III 2375, Micromeritics) and density (Helium Pycnometer, ACCUPYC 1330 Micromeritics).

3. Results and discussion

The main object of this study was to produce a highly sinterable submicrometer silicon carbide powder. The first step to look at was the milling efficiency, since as the particles approach submicrometric (<1 μm) or nanometric (<0.3 μm) dimensions, there is a reduction in the milling efficiency as a result of two competing phenomena, namely, comminution and agglomeration [1]. The second step was to observe if the conditions of the high-energy milling process used generated contamination.

The comminution process was executed in a planetary mill, operated at 300 rpm with milling media composed of SiC powder, spheres of ZrO_2+CeO_2 and isopropyl alcohol as the dispersant. These processing characteristics resulted in a fast size reduction, but slow change in distribution, as shown in Fig. 1. The as-received powder (Fig. 1a) had a bimodal distribution, with peaks centered at 0.2 and 5 μm. The peak situated at 5 μm

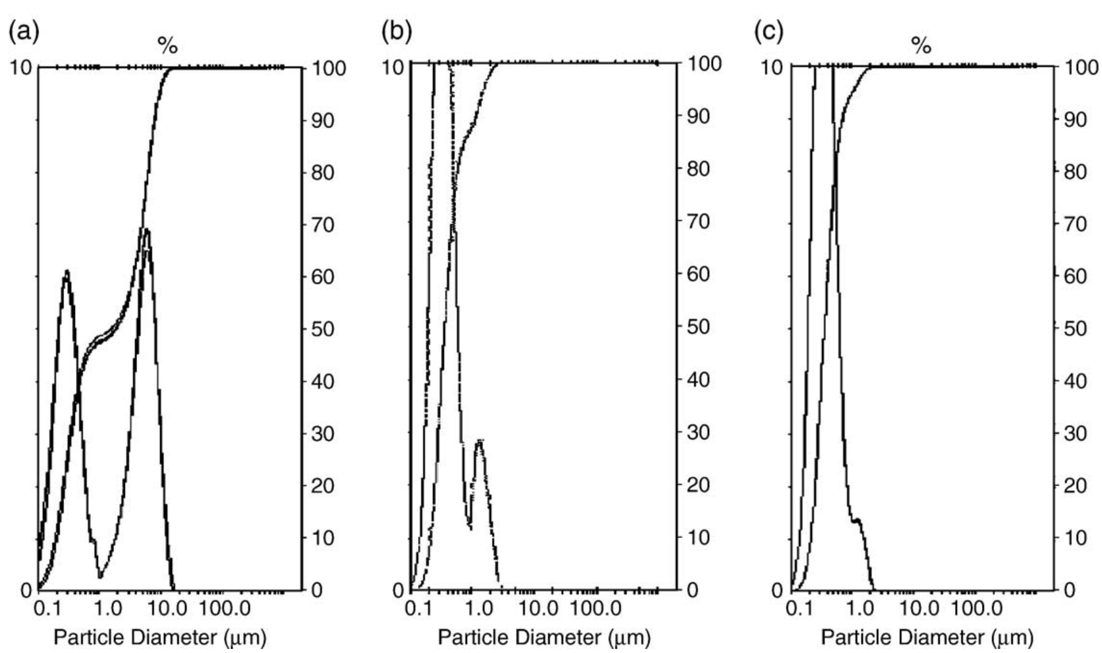

Fig. 1. Particle size distribution: (a) as-received SiC, (b) after 4 h and (c) after 6 h of milling.