



Original Research Paper

Non-firing ceramics: Activation of silica powder surface for achieving high-density solidified bodies



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ABSTRACT

Sintering of most ceramic materials requires high temperatures over 1000 °C due to their high melting point. Sintering is a high-cost process, which can take several hours or days. Also, sintering at high temperatures can affect the microstructure of ceramics by causing undesired grain coarsening or changing the chemical stoichiometry of final product when volatile elements are present. In this study, a so-called “non-firing sintering” of silica particles is proposed, without using thermal energy. This method involves the chemical activation of powder surface via ball milling, where the surface of particles is rubbed against balls, and friction between particle/ball breaks the bonds of functional groups like $\equiv\text{Si}-\text{O}-\text{Si}\equiv$ on the surface of particles. Results show that water adsorption increases as milling time is increased, while particle size is unchanged after ball milling according to nitrogen adsorption isotherms. Scanning electron microscopy results confirm the trans granular fracture of pellets prepared by ball milling, and transmission electron microscopy micrographs show a necking-like bonding between silica particles.

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1. Introduction

Sintering of ceramics is realized as a symbol of human civilization and refers to a process that particles consolidate through an atomic diffusion event by increasing the temperature and/or pressure [1,2]. On the one hand, sintering is the most important part of ceramic processing in terms of reliability of the final product because pore-free and high-strength materials can be obtained only by a proper sintering process [3,4]. On the other hand, sintering is a complicated process because many factors such as particle size, size distribution, particle morphology, green body density, diffusion coefficient, vapor pressure, and viscosity can easily affect the whole sintering process [2,5]. Most of the ceramic materials possess a high melting point and therefore sintering of ceramics is usually conducted at high temperatures over 1000 °C, approximately 70% of the melting point.

Lowering the sintering temperature in ceramic materials has been desired during the past decades [6,7]. High-temperature sintering is an energy-consuming and high-cost process, which can take several hours to several days in order to achieve a dense sintered body. In addition, sintering at high temperatures may change

the chemical stoichiometry of the final product when volatile elements like Li, Na, K and Pb are present in the system [8–10]. Addition of liquid phase additives [11], high-pressure sintering [12], two-step sintering [13], spark plasma sintering [14], flash sintering [15], and microwave sintering [16] are some well-known methods applied for lowering the sintering temperature. However, temperatures usually higher than 500 °C are still necessary in these methods. Recently, a cold sintering process is reported [17,18], in which inorganic compounds can be sintered at temperatures less than 300 °C, under pressures in the MPa range.

In this study, a facile “non-firing sintering” process will be proposed without heating the sample and also without applying an external pressure. The underlying idea of non-firing sintering is to activate the surface of particles by ball milling and consolidate the particles via the joining of activated surfaces. By ball milling, the surface of particles is rubbed against balls, and the friction between particle/ball breaks the bonds of functional groups like $\equiv\text{Si}-\text{O}-\text{Si}\equiv$ on the surface of particles. This study shows how non-firing sintering is effective for solidifying silica (SiO₂) powders, where high relative densities are obtained. Activation of particle surface after ball milling is investigated systematically by water adsorption measurements because more active surfaces can adsorb more water. In order to make sure that particle sizes are not reduced after ball milling, nitrogen (N₂) gas adsorption is con-

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ducted for all samples and surface areas are estimated based on the Brunauer-Emmett-Teller (BET) method. Microstructure of samples is observed by scanning electron microscopy and transmission electron microscopy methods.

2. Materials and methods

SiO₂ powder (average particle size: 550 nm, purity: >99.8%) was ball milled in a 500 mL polypropylene vessel at 300 rpm. The ball mill vessel was filled with 400 g zirconia balls with a diameter of 13 mm, and 75 g SiO₂ powder. Seven samples were prepared with different milling times as 0 h (raw powder), 1 h, 3 h, 6 h, 12 h, 24 h, and 48 h at room temperature. The temperature of powders during ball milling is measured every 1 h, and it is confirmed that the temperature of powders after milling do not increase significantly, being around room temperature even after 48 h milling. After milling, powders were passed through a 50-mesh polyethylene sieve and uniaxially compressed at 5.3 MPa to prepare pellets of 13 mm diameter and 10 mm height. The densities of all the samples were measured using the Archimedes method using distilled water as the displacement liquid. For each experiment, the average density of two pellets was used and the accuracy of bulk densities was within ± 0.03 g/cm³. Scanning electron microscopy (SEM; JSM-7600F, JEOL, Japan) was carried out to estimate the average particle sizes after ball-milling and observing fracture surfaces of each pellet. Water adsorption measurements were carried out at 298 K in an automatic volumetric gas adsorption instrument (Belsorp-max, MicrotracBEL, Japan). The adsorption isotherms of N₂ at 77 K were measured in the same surface area analyzer and the active surface area of samples was measured by Brunauer-Emmett-Teller (BET) method. Prior to measurements, the samples were outgassed at 473 K for 2 h under vacuum less than 10⁻² kPa. The particle-particle interface of sample ball-milled at 6 h was observed by transmission electron microscopy (TEM; JEM-ARM200F; JEOL, Japan) at 200 kV. TEM samples were prepared by using a focused ion beam (FIB). Vickers hardness test was conducted for 0 h, 3 h, and 24 h samples with a load of 9 N and dwell time of 10 s. For each sample 20 tests were performed to make sure that data were reproducible.

3. Results and discussion

The water adsorption isotherms at 298 K are plotted in Fig. 1a for samples with various milling time. A type II isotherm is observed for all samples, suggesting a good water-silica interaction. It is clear that as the milling time increases, the volume of water adsorbed on the surface of silica particles increases. The interaction of water and silica surface has been widely studied [19,20], and it is proposed that a freshly formed amorphous silica contains large amounts of active sites on the surface [21,22]. These active sites readily react with atmospheric moisture, leading to the formation of surface silanol (Si-OH) groups [19]. Surface OH groups are subdivided into isolated, geminal, and vicinal. In addition, surface siloxane groups ($\equiv\text{Si}-\text{O}-\text{Si}\equiv$) with oxygen atoms on the surface are also present. In this study, adsorption of water is increased with increasing the milling time because ball milling effectively generates new active sites on the surface of silica powders by breaking the Si-O bonds in siloxane groups on the surface of silica. As a consequence, more water can be adsorbed to the newly formed sites. Fig. 1b shows the volume of water adsorbed on the surface of silica versus the milling time. It is shown that water adsorption volume increases with increasing the milling time up to 12 h, however, saturation occurs after further milling time. A schematic illustration of surface activation and water adsorption is shown in Fig. 2.

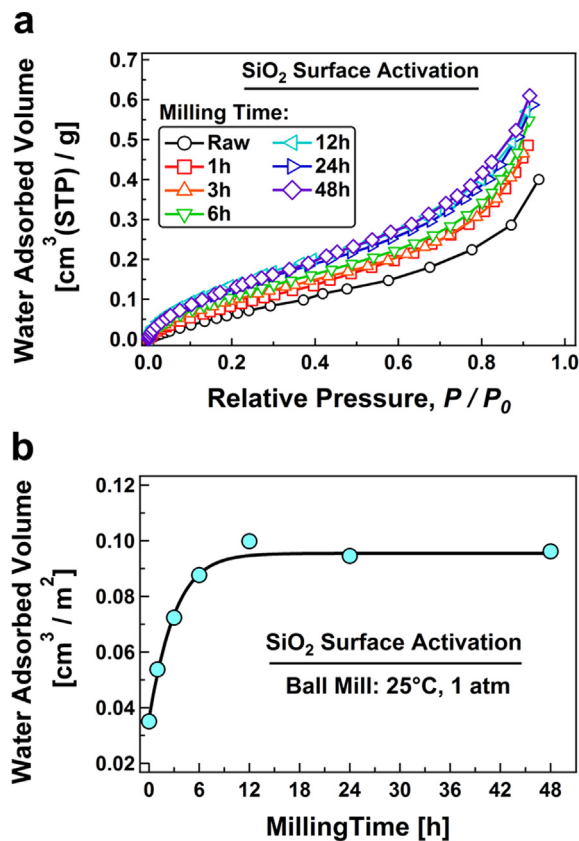


Fig. 1. (a) Water adsorption isotherms at 298 K for samples ball milled at various periods. (b) Water adsorbed volume as a function of milling time.

In order to make sure that the amount of increased water adsorption after ball milling is because of surface activation but not due to the particle size reduction, the surface area of all samples are estimated by nitrogen gas adsorption. Nitrogen adsorption isotherms of samples ball milled at various periods are shown in Fig. 3a. All samples possess a type II nitrogen gas sorption isotherm according to the IUPAC classification [23]. All isotherms represent a continuous increase at low relative pressure, indicating the adsorption of nitrogen gas on the surface of silica particles. The increase in the nitrogen sorption at higher relative pressure is due to the gas adsorption in pores and voids. It is clear from Fig. 3a that the behavior of all samples regardless of milling time is almost the same. BET specific surface areas are derived from isotherm data as shown in Fig. 3b. Although a slight increase in the specific surface area of particles can be confirmed with increasing the milling time, it can be concluded that the surface area of silica particles even after 48 h of milling is almost unchanged with respect to that of raw powder. The surface area of raw powder is 6.31 m²/g, and that of silica milled for 48 h is 6.60 m²/g.

Fig. 4 shows the average bulk density of each sample. The average bulk density of pellets prepared by raw powder is as low as 1.41 g/cm³. However, the average bulk density increases as the milling time of powders increases, reaching 2.00 g/cm³ at milling time of 12 h. After 48 h of ball milling, the average bulk density slightly increases with respect to that of 12 h, reaching 2.05 g/cm³. The enhanced bulk density by ball milling indicates that non-firing method proposed in this study is effective in preparing dense bodies without applying temperature and pressure. The fracture surface of some representative samples is observed by SEM, as shown in Fig. 5. Samples prepared by raw powder and ball milled for 1 h show an intergranular fracture, suggesting a weak particle-

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