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Original Research Paper

High-purity disperse α-Al_2O_3 nanoparticles synthesized by high-energy ball milling

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ABSTRACT

The preparation of disperse fine equiaxed α-Al_2O_3 nanoparticles with narrow size distribution, high purity, and high yield is essential for producing Al_2O_3 nanocrystalline ceramic of fine grains which may exhibit a good toughness. In this work, micron-sized α-Al_2O_3 particles were directly ball-milled and subsequently washed with hydrochloric acid at room temperature. Fracture of large α-Al_2O_3 particles and cold welding of fine α-Al_2O_3 nanoparticles occur simultaneously during ball milling. It leads to the reduction of particle size with increasing milling duration below 80 h and reaches to a dynamic equilibrium with a minimal average particle size of 6.4 nm for milling durations over 80 h. Using the optimized high-energy ball milling parameters, we prepared high-purity disperse equiaxed α-Al_2O_3 nanoparticles with an average particle size of 8 nm and a purity of 99.96% (mass percent) in a high yield. After fractionated coagulation separations, disperse fine equiaxed α-Al_2O_3 nanoparticles with narrow size distribution were obtained. Finally, Al_2O_3 nanocrystalline ceramic with a relative density of 99.8% and an average grain size of 34 nm was sintered from the disperse fine equiaxed α-Al_2O_3 nanoparticles with an average particle size of 4.8 nm and a size distribution of 2–10 nm by pressureless two-step sintering.
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1. Introduction

Alumina (Al_2O_3) ceramics are essential materials of industrial importance due to their excellent physicochemical properties and wide applications in structural materials, microelectronics, optical ceramics, and dental formulations [1–4]. However, as compared to metallic materials, Al_2O_3 ceramics produced so far are brittle at room temperature, which substantially limits their wider applications. The mechanical properties strongly depend on the microscopic structure of the materials [5]. CaF_2 and TiO_2 nanocrystalline ceramics with an average grain size of 8 nm exhibit noticeable plastic deformations at 80 and 180 °C, respectively [6]. SiC nanocrystalline ceramic shows increasing toughness with the reduction of grain size [7]. In addition, the reduction of grain size in nanoscale can simultaneously improve the hardness, fracture toughness, and transparency of nanocrystalline ceramics such as cubic Si_3N_4 and $MgAl_2O_4$ [8,9]. So Al_2O_3 nanocrystalline ceramics may exhibit a combination of excellent properties such as high toughness, high hardness, high thermal and chemical stability, and transparency. Disperse fine equiaxed α-Al_2O_3 nanoparticles (NPs) with narrow size distribution exhibit a good powder flow and microstructural uniformity in green bodies, reduce sintering temperature, enhance the densification rate, and alleviate the abnormal growth of large particles at the expense of small particles during sintering [10–12]. Furthermore, high-content impurities in Al_2O_3 ceramics are usually harmful to their properties such as optical properties and undesired for high-performance Al_2O_3 nanocrystalline ceramics [13–15]. In order to densify fully dense Al_2O_3 nanocrystalline ceramics with excellent properties, high-purity, disperse, fine, and equiaxed α-Al_2O_3 NPs with narrow size distribution are essential.

Many efforts have been attempted to synthesize disperse fine equiaxed α-Al_2O_3 NPs. α-Al_2O_3 NPs were usually derived from the calcination of precursors at high temperatures above 1000 °C [16–21]. However, α-Al_2O_3 NPs below 15 nm are thermodynamically unstable with respect to γ-Al_2O_3 at room temperature and thus extremely difficult to fabricate [22,23]. The high-temperature calcination leads to the undesired sintering and growth of α-Al_2O_3 particles. So the α-Al_2O_3 particles prepared by calcination of precursors, even prepared at the low formation temperature by introducing seeds, usually have a vermicular or aggregated microstructure [16–21]. The vermicular or aggregated microstructure leads to large and non-uniform pores in green bod-

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ies which can only be eliminated at high sintering temperatures and results in coarse-grained Al_2O_3 ceramics rather than expected Al_2O_3 nanocrystalline ceramics with fine grains [11,12]. In our previous work, disperse equiaxed α-Al_2O_3 NPs with a mean size of 12 nm were prepared at 770 °C using α-Fe_2O_3 as seed and isolation phase [24]. However, the Fe impurity in the obtained α-Al_2O_3 NPs is as high as 4.26% (atomic percent). The α-Al_2O_3 NPs with a high purity above 99.9% (mass percent) reported so far are larger than 100 nm, even in micron scale [25,26].

High-energy ball milling was often employed to prepare disperse NPs. ZrO_2 NPs were synthesized by the mechanochemical reaction of $ZrCl_2$ and MgO forming ZrO_2 and $MgCl_2$ [27]. The by-products ($MgCl_2$) could effectively isolate the ZrO_2 NPs during the milling; then dispersed ZrO_2 NPs of about 6 nm were obtained by the subsequent dissolution of $MgCl_2$. Karagedov et al. [28] prepared α-Al_2O_3 NPs with a mean size of 25 nm by milling the mixture of α-Al_2O_3 powders and "grinding catalyst". However, the impurity content is as high as 2.4% (mass percent) in the obtained α-Al_2O_3 NPs, and the α-Al_2O_3 ceramic sintered from these NPs has a mean grain size as large as 0.1–0.2 μm and a relative density of only 92% [28]. In our another previous work, disperse equiaxed α-Al_2O_3 NPs with an average particle size of 14.3 nm and a size distribution of 2–250 nm were prepared by removing the Fe matrix in the α-Al_2O_3-NPs-embedded composites (synthesized by a mechanochemical method) through selective corrosion [29]. After fractionated coagulation separations, disperse fine equiaxed α-Al_2O_3 NPs with different average sizes below 15 nm and narrow size distributions were obtained. However, the mechanochemical method is complicated, and the yield of fine α-Al_2O_3 NPs is relatively low (the mass fraction of α-Al_2O_3 NPs below 10 nm is as low as 1.1%). Moreover, the purity of the obtained α-Al_2O_3 NPs (99.6 wt%) is not high enough. So it is necessary to develop a simple method to prepare higher-purity, disperse, fine, and equiaxed α-Al_2O_3 NPs in a high yield. Besides, the mechanisms for the high-energy ball milling synthesis of NPs reported mainly focused on metals, solid solutions, intermetallics, and alloys [30–32]. However, the mechanisms for the high-energy ball milling synthesis of ceramic NPs have not been explored.

In our present work, a new simple high-energy ball milling process was developed to prepare high-purity, disperse, fine, and equiaxed α-Al_2O_3 NPs. In this new process, micron-sized α-Al_2O_3 particles were directly ball-milled and subsequently washed by hydrochloric acid (HCl) at room temperature. The high-energy ball milling parameters were optimized. The formation mechanism of disperse fine equiaxed α-Al_2O_3 NPs during high-energy ball milling was analyzed. Using the optimized ball milling parameters, disperse equiaxed α-Al_2O_3 NPs with an average particle size of 8 nm and a purity of 99.96% (mass percent) were prepared. Then disperse fine equiaxed α-Al_2O_3 NPs of different average sizes and narrow size distributions were obtained by fractionated coagulation separation, especially the disperse fine equiaxed α-Al_2O_3 NPs with average particle sizes below 10 nm and narrow size distribution suitable for densifying Al_2O_3 nanocrystalline ceramic. Finally, the green compacts of disperse fine equiaxed α-Al_2O_3 NPs with an average particle size of 4.8 nm and a narrow size distribution of 2–10 nm were sintered to an almost fully dense Al_2O_3 nanocrystalline ceramic with an average grain size of 34 nm and a relative density of 99.8% by a pressureless two-step sintering.

2. Experimental section

Commercial aluminium nitrate (99.0 wt%, Tianjin Kemiou) was used as raw material. The aluminium nitrate powders were calcined at 1150 °C for 2 h to obtain α-Al_2O_3 powders for high-energy ball milling.

High-energy ball milling was performed on a Fritsch P4 planetary ball mill with two 80 mL stainless steel vials and stainless steel balls of 10 mm in diameter. Then 10 g calcined α-Al_2O_3 powders were loaded in each vial along with balls in air and milled at different milling parameters. The milling parameters were optimized by adjusting the main disk rotation speed from 150 to 300 revolutions per minute (rpm), the ball-to-powder weight ratio (BPR) from 10:1 to 50:1, and the milling duration from 2 to 100 h. The relative rotation speed ratio of the vial to the main disk was −2. The ball-milled powders were washed with 12 mol/L HCl at room temperature. The resulting powders collected by centrifugation were washed several times with 4 mol/L HCl in ultrasonic bath and centrifuged at 10000 rpm for 5 min.

Before fractioned coagulation separations, α-Al_2O_3 NPs were suspended in deionized water in ultrasonic bath and centrifuged at 10,000 rpm for 5 min to remove α-Al_2O_3 particles larger than 100 nm. α-Al_2O_3 NPs smaller than 100 nm were used for fractioned coagulation separations. The α-Al_2O_3 NPs smaller than 100 nm were suspended in 3 mol/L HCl. Larger α-Al_2O_3 NPs coagulated and smaller α-Al_2O_3 NPs stayed in supernatant after centrifugation. All α-Al_2O_3 NPs suspending in the upper supernatant of 3 mol/L HCl settled down by adding enough 12.0 mol/L HCl and were collected by centrifugation at 10,000 rpm for 5 min. So α-Al_2O_3 NPs separated at 3 mol/L HCl were obtained. In similar ways, α-Al_2O_3 NPs were separated at 1.8, 1.0, and 0.6 mol/L HCl, respectively.

The α-Al_2O_3 NPs with an average particle size of 4.8 nm and a size distribution of 2–10 nm obtained by high-energy ball milling-acid washing-fractioned coagulation separation process were uniaxially pressed into green compacts (8 mm in diameter, 0.4 mm in thickness, and 48% in relative density) at 500 MPa with a dwell time of 6 h. The green compacts were sintered in air by two-step sintering. They were heated at a rate of 10 °C/min up to 1150 °C without hold, cooled at a rate of 10 °C/min down to 1000 °C, kept for 40 h at 1000 °C, and finally cooled at a rate of 10 °C/min down to room temperature.

The crystal structure and grain sizes of the obtained α-Al_2O_3 NPs were analyzed by X-ray diffraction (XRD) using a Rigaku D/Max-2400 diffractometer with Cu K_α radiation and a scanning speed of 5°/min working at a voltage of 40 kV and a current of 150 mA. XRD peak broadening analysis was used to evaluate the grain sizes and lattice strain by the Williamson-Hall analysis [33]. Transmission electron microscopy (TEM) observations, selected area electron diffraction (SAED), and energy dispersive X-ray spectroscopy (EDS) analyses were conducted on an FEI Tecnai G^2 F30 field-emission transmission electron microscope operated at an acceleration voltage of 300 kV to analyze the microstructure, lattice structure, and elemental compositions of the obtained α-Al_2O_3 NPs. Furthermore, an accurate elemental analysis was conducted using an inductively coupled plasma-atomic emission spectrometry (ICP-AES) on a TJA IRIS ER/S ICP-AES spectrometer. The specific surface areas of α-Al_2O_3 NPs were measured by N_2 adsorption at 77 K using the Brunauer-Emmett-Teller (BET) method on a Micrometrics ASAP 2020 M surface area analyzer. Before the BET measurements, the powder samples were degassed at 300 °C for 12 h. Relative densities of sintered bodies were determined by Archimedes' method. The sintered bodies were fractured and then thermally etched to analyze their microstructure by scanning electron microscopy (SEM) on a Tescan MIRA 3 field-emission scanning electron microscope operating at an acceleration voltage of 15 kV. The average particle or grain sizes and size distributions of the α-Al_2O_3 NPs or sintered bodies were statistically determined from several thousands of particles or grains observed by TEM or SEM in the different areas of the samples.

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