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Grinding performance of pellet prepared using nanosize ceria particles

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Abstract

The cerium oxide particles of single crystal were obtained only by the combination of precipitation method and hydrothermal treatment at lower temperature than 200 °C. The particles showed plate-like shape even in the particle size of about 4 nm, and the plate-like plane was identified to be (1 1 1) from the observation of lattice images. By heating at 100–700 °C in air, the particle size was increased from about 4 to 60 nm maintaining the plate-like shape with elevating the temperature. These cerium oxide particles were applied as a fine abrasive grain for mirror grinding. We manufactured a grinding pellet, consisting of the plate-like cerium oxide particles, using electrophoretic deposition (EPD) phenomenon. It is called "ceria EPD pellet". As a result of grinding of quartz-crystal wafer using the ceria EPD pellet, fine mirror surface with roughness less than 1 nmRy was realized. Grinding efficiency of the ceria EPD pellet was fourth time as high as a silica EPD pellet.

Keywords: Nanostructures; Precipitation; Chemical synthesis; Crystal growth; Transmission electron microscopy

1. Introduction

Nanosize oxide particles are technologically important materials because of their anticipated new characteristics [1–5]. Cerium oxide particles have been used in chemical mechanical polishing (CMP). The oxide particles are usually produced by a ceramic process involving high temperature solid-state treatment. The oxide particles obtained by this process are rather large and non-uniform in size. These non-uniform particles result in scratching or rough surfaces in polishing. In order to overcome difficulties arising out of the ceramic process, wet chemical methods like hydrothermal treatment at supercritical conditions [6,7], sol–gel method [8–13], etc. have been developed for production of fine and reproducible particles. The oxide particles with the particle size larger than 10 nm are obtained by the sol–gel method. The hydrothermal synthesis with the use of supercritical water is suitable to produce pure and fine oxides and to control the particle size over a wide range while maintaining a narrow particle size distribution. However, the hydrothermal synthesis with supercritical water has the problem of high production cost due to the use of high temperature and high pressure. We reported various kinds of nanosize oxide particles synthesized by the combination of precipitation method and hydrothermal treatment at temperature lower than 200 °C [14,15].

In this study, we report the analysis of the particle shape and structure of the nanosize cerium oxide particles synthesized by the combination method, and the particle size control by the method. We manufactured a grinding pellet, a ceria EPD pellet, consisting of the nanosize plate-like cerium oxide particles. Mirror processing of quartz-crystal wafer using the ceria EPD pellet is discussed.

2. Experimental

2.1. Synthesis of cerium oxide particles

Cerium oxide particles were synthesized as follows. An aqueous solution of cerium chloride was added dropwise to the solution containing equivalent mole of sodium hydroxide

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while stirring the solution to make the precipitant of cerium hydroxide. After ageing the suspension at room temperature for 1 day, the pH of suspension was adjusted depending on the directing particle size. The suspension so obtained was treated in hydrothermal condition using an autoclave at the temperature of 180 °C for 4 h. The slurry was washed with distilled water and dried at 90 °C in air. After disassembling the aggregation of the dried particles, the particles were heated at various temperatures in air to convert to cerium oxide particles.

The crystallite size and structure were measured using X-ray diffraction (XRD) both for the particles made only by the hydrothermal treatment and for the cerium oxide particles as final products. The morphology of these particles was analyzed using a high resolution transmission electron microscope (TEM). The mean particle size was calculated from the value of BET surface area.

2.2. Fabrication method of EPD pellet

In general, the oxide particles adsorb various kinds of ions in solution by the surface charge. Therefore, these particles are adsorbed to either electrode by electric field. By utilizing electrophoretic deposition phenomenon, it is possible to fabricate EPD pellet having homogeneous organization structure in which the particles are uniformly dispersed [16].

The fabrication condition of the ceria EPD pellet is shown in Table 1. When the cerium oxide particles were dispersed in water solution of sodium alginate which is a polymer electrolyte, alginate ion was adsorbed to the cerium oxide particles as a protective colloid. When the electric field was applied in this dispersion, the cerium oxide particles were adsorbed to anode pole and the adsorption layer consisted of the cerium oxide particles and sodium alginate was formed. This adsorption layer was torn from the anode pole and dried for 24 h to obtain the ceria EPD pellet.

Table 1
Grinding condition

Grinding pellet	
Composition	
Ceria particles (mass%)	25
Sodium alginate (mass%)	3
Pure water	Balance
Applied voltage (V)	10
Deposition time (min)	30
Grinding method	Infeed grinding
Grinding fluid	Non (silica) or water (ceria)
Revolution of grinding wheel (rpm)	1000
Revolution of work table (rpm)	30
Infeed rate (μm/min)	1.5
Depth of cut (μm/pass)	10
Pass number	2
Spark-out time (min)	2

2.3. Grinding of quartz-crystal wafer

Grinding of a quartz-crystal wafer using the ceria EPD pellet was performed by high-precision grinding machine (TOYODA MACHINE WORKS, SG-30). Grinding condition is shown in Table 1. The quartz-crystal wafer polished by the conventional method was used. In the case of the ceria EPD pellet, the quartz-crystal wafer was ground during flowing water at 200 ml/min as grinding fluid. In the case of the silica EPD pellet, the quartz-crystal wafer was ground without using grinding fluid. The grinding efficiency was calculated from the weight decrease of quartz-crystal wafer, and the surface roughness was measured by AFM observation.

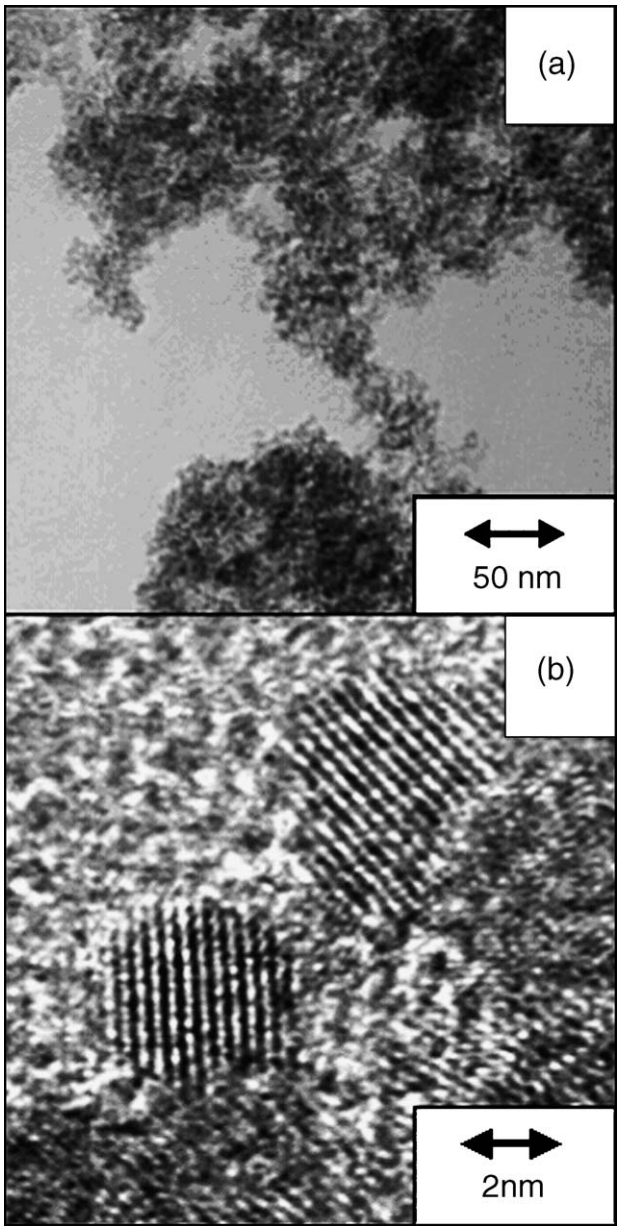

Fig. 1. (a) TEM photograph of cerium oxide particles obtained by hydrothermal treatment. (b) High resolution TEM photograph of the same cerium oxide particles obtained by hydrothermal treatment.

3. Results and discussion

3.1. Analysis of particle shape and structure of cerium oxide particles

The XRD pattern of the particles made only by the combination of precipitation method and hydrothermal treatment is identified with that of cerium oxide particles with the fluorite type structure. The mean particle size calculated from the BET surface area (180 m^2/g) is 4.5 nm.

As shown in Fig. 1(a), TEM observation of the cerium oxide particles made only by the combination method indicates an aggregation of fine particles. Fig. 1(b) shows the observation for the individual particles using the high resolution TEM. Clear lattice images are observed for each particle, and the lattice planes were identified to be composed of almost all of (1 1 1) and a few (2 0 0) planes. This preferential orientation of (1 1 1) plane indicates that each particle has plate-like shape even in the particle size of about 4 nm.

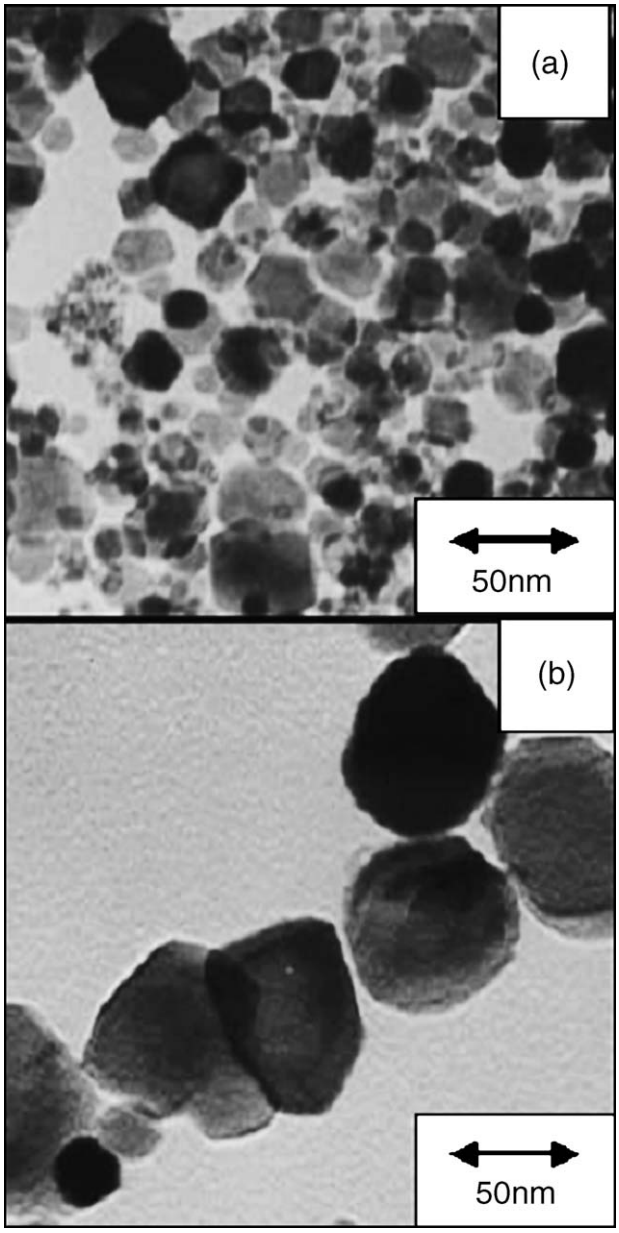

Fig. 2. TEM photographs of cerium oxide particles heated at (a) 600 °C and (b) 700 °C.

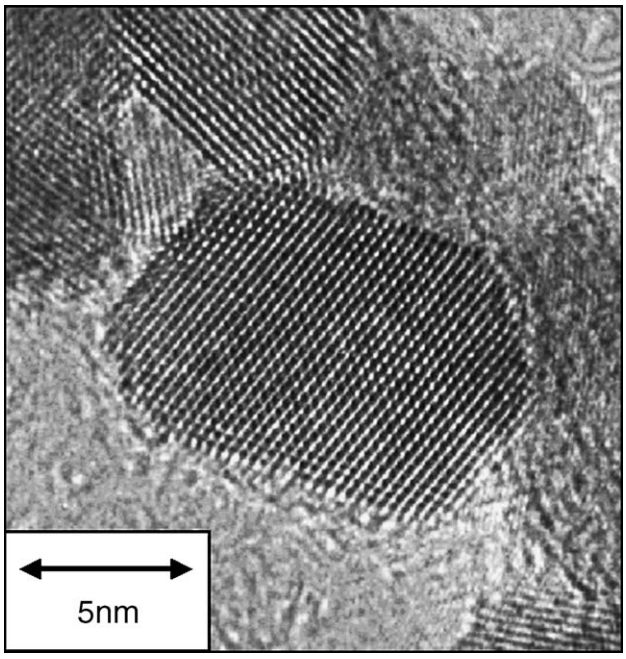

Fig. 3. High resolution TEM photograph of cerium oxide particles heated at 600 °C.

The crystallite diameter of the particles determined from the linewidth of the (1 1 1) peak in the XRD patterns using the Scherrer formula was 4.6 nm which was nearly the same value as the mean particle size calculated from BET surface area. The lattice constant determined from XRD patterns

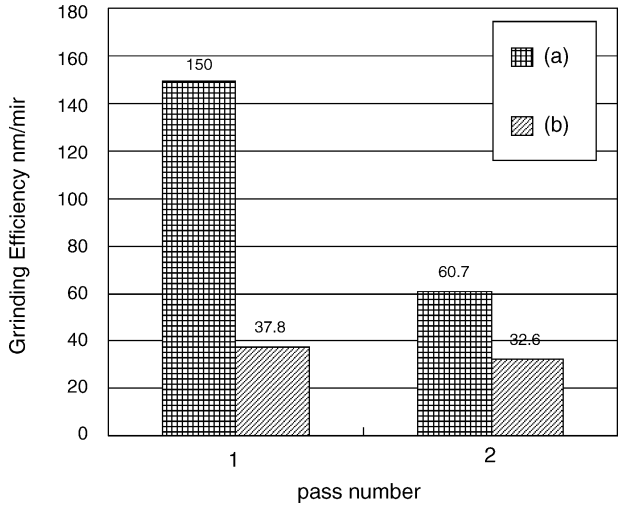

Fig. 4. The grinding efficiency of (a) the ceria EPD pellet on wet condition and (b) the silica EPD pellet on dry condition for the quartz-crystal wafer.

was 0.541 nm which agreed with the lattice constant of bulk of cerium oxide. These results show that the particles obtained only by the combination of precipitation method and hydrothermal treatment at 180 °C have single crystal structure with plate-like shape even in the fine particle size of about 4 nm.

3.2. Size control of cerium oxide particles

The particles obtained by the combination method were heated in the temperature range of 100–700 °C for 1 h in air. Fig. 2(a and b) shows TEM photographs of these particles heated at 600 and 700 °C, respectively. As the heating temperature is elevated from 100 to 700 °C, the particle size increases from about 4 to 60 nm.

Fig. 3 shows the high resolution TEM photograph of the cerium oxide particles obtained by heating at 600 °C. The particle shape is similar to hexagon with the mean particle diameter of about 13 nm. The lattice images were identified to be based on (1 1 1) planes, indicating that the cerium oxide particles have the plate-like shape and show the preferential orientation of the (1 1 1) plane. The plate-like shape of the cerium oxide particles is possibly related to the plate-like shape of the original particles made only by the combination method.

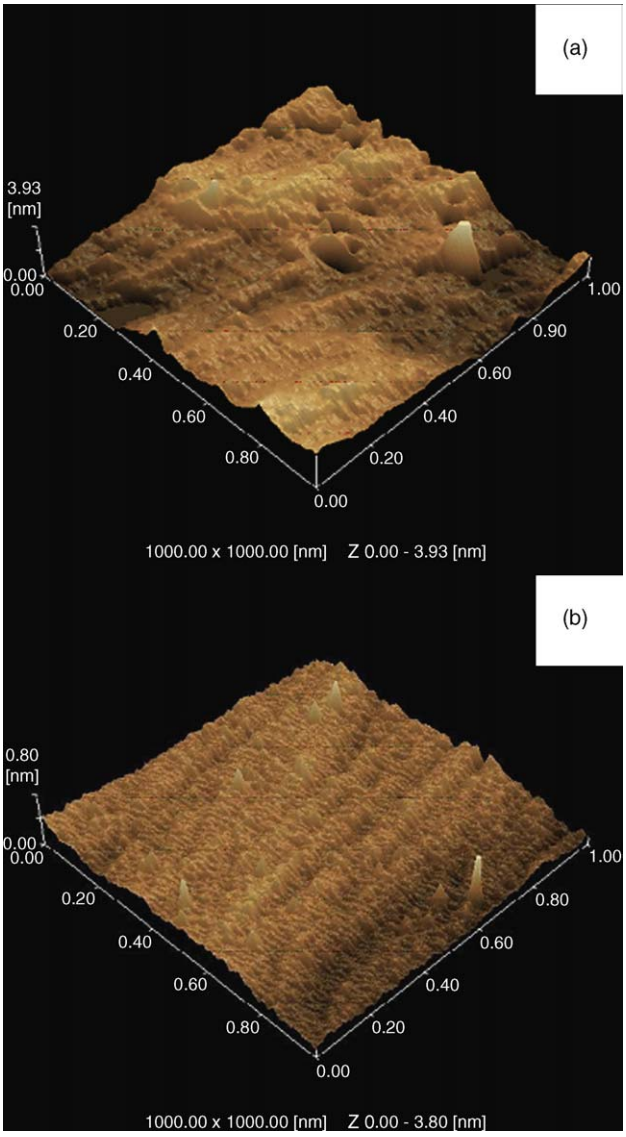

Fig. 5. AFM observation for the surface of the quartz-crystal wafer (a) before and (b) after grinding by the ceria EPD pellet on wet condition.

3.3. Grinding performance of ceria EPD pellet

Fig. 4 shows the grinding efficiency of the ceria EPD pellet, as compared with the silica EPD pellet. On the first pass, the grinding efficiency of the ceria EPD pellet was 150 nm/min. It is about four times larger than that of the silica EPD pellet. On the second pass, the grinding efficiency of the ceria EPD pellet was decreased 60.7 nm/min. It was considered that the lower grinding efficiency in the second pass was due to the decrease of hardness of the ceria EPD pellet by grinding fluid.

Fig. 5(a) shows the AFM observation for the surface of the quartz-crystal wafer before the grinding. As shown in Fig. 5(b), the fine mirror surface of 0.056 nmRa was realized by grinding using the ceria EPD pellet. These results indicate the possibility of processing the crystal wafer by angstrom order, using such EPD grinding.

4. Conclusions

We synthesized nanosize cerium oxide particles utilizing the combination of precipitation method and hydrothermal treatment lower than 200 °C. The cerium oxide particles obtained only by the combination method showed single crystal with plate-like shape even in the particle size of about 4 nm. By heating at 100–700 °C in air, the particle size was increased from about 4 to 60 nm with increasing temperature, while maintaining the plate-like shape. The ceria EPD pellet was prepared using plate-like cerium oxide particles of 60 nm. The grinding efficiency of the ceria EPD pellet was 150 nm/min for the quartz-crystal wafer, which was about four times larger than that of the silica EPD pellet. The fine mirror surface of 0.056 nmRa was realized by grinding using the ceria EPD pellet.

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