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Journal of the European Ceramic Society 27 (2007) 733–737
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Wet jet milling of Al_2O_3 slurries

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Available online 23 May 2006

Abstract

A wet jet milling process was employed as a novel method to prepare ceramic slurries. The wet jet milling pulverized raw materials to primary particle size within a short time. The wet jet-milled slurries showed very low viscosity, as comparing to ball-milled slurries. Moreover, the viscosity of wet jet-milled slurries was stable for long times, whereas that of ball-milled slurries increased rapidly with increase in the time. Al_2O_3 particles after wet jet milling kept initial surface conditions, though Al_2O_3 particles after ball milling yielded more OH groups on the surface. The relative density of the green bodies prepared from the wet jet-milled slurries was about 65% or more without depending on the solid content of slurry. On the other hand, the relative density of the green bodies fabricated by the ball-milled slurries increased with the slurry solid content, and it reached more than 60% at 50 vol% of solid content.

Keywords: Milling; Slip casting; Suspensions; Al_2O_3; Viscosity

1. Introduction

Ceramic slurries for fabrication of ceramic products are usually prepared by ball milling,[1–7] planetary ball milling[8] and beads milling.[9,10] The ceramic slurry which disperses ceramic powders in an aqueous solvent with dispersant must satisfy the following conditions to fabricate dense green body: (1) high solid content and (2) good dispersion.[11,12] A good dispersion of ceramic particles in the aqueous phase is achieved by ensuring strong electrostatic and steric repulsion compared to the attractive interparticle van der Waals force. However, it is known that ceramic particles after milling have tendency to re-flocculate[13,14] by an increase of the active sites such as lattice defect and radicals on the particle surface. These active sites are induced by the excess collision energy with grinding media such as ball and bead.[15] The re-flocculation of particles causes a viscosity increase and thixotropic property.[16] The flocculated particles result in inhomogeneous and loose structured green bodies, i.e. broad pore size distribution and low relative density.[17,18] It is also known that these milling requires a long time to prepare good slurries[19] and induces impurities contamination from grinding media.[15]

Recently, in chemical engineering and food technology fields, the wet jet milling, in which particles in the suspension or slurry collide mutually at high pressure and high speed, has been developed as a new method of mixing and dispersion.[20] In this system, turbulent flow, shear flow and cavitation are generated by the high speed flow and rapid pressure release. Using these effects, the grain refinement, emulsifying and homogenization are achieved within a short period of time with no grinding media. Since wet jet milling is a media less process, it is expected that the slurry with no re-flocculation property can be prepared.

In the present work, wet jet milling process is employed to prepare ceramic slurries. The usefulness of wet jet milling is investigated by characterizing the properties of wet jet-milled slurries, such as particle size distribution and viscosity. Furthermore, the casting property of wet jet-milled slurries was also investigated. We will demonstrate that the good slurries with low viscosity and non-flocculate property can be produced by wet jet milling and the green bodies with high relative density can be fabricated using these slurries.

2. Experimental procedure

2.1. Slurry preparation

A commercially available high purity α-Al_2O_3 (AKP-20; Sumitomo Chemical, Japan) with an average particle size (D_{50})

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0955-2219/$ – see front matter © 2006 Elsevier Ltd. All rights reserved.
doi:10.1016/j.jeurceramsoc.2006.04.001

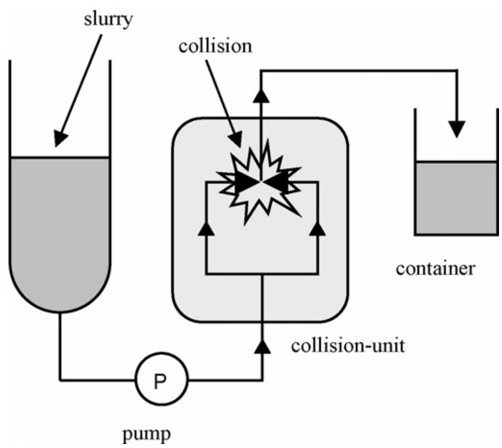

Fig. 1. Schematic illustration of wet jet mill system.

of 0.5 μm was used. A commercially available NH_4^+ salt of poly(acrylic acid; PAA, Aron A-6114; Toagosei, Japan) was the dispersant of choice. The aqueous suspensions containing 10, 30 and 50 vol% of Al_2O_3 powder were prepared in 0.12 wt.% dispersant solution. The suspensions were stirred for 2 h before milling.

A wet jet mill system (PRE03-20-SP; Genus, Japan) was used for this study. A diagram of wet jet mill system is shown in Fig. 1. Slurry was pumped into the collision unit at 200 MPa where the mutual collision of the particles took place. The processing time depends on the treatment amount of slurry because this system is continuous process, it takes only 3 min for 100 ml. For the purposes of comparison, the same Al_2O_3 slurries (10, 30 and 50 vol%) were also prepared by ball milling. The ball milling was performed for 24 h using high grade Al_2O_3 balls with a diameter of 10 mm.

2.2. Characterization

Acoustic Spectrometer (DT-1200; Dispersion Technology, USA) was employed for particle size distribution analysis. The rheological characteristics of the slurries were measured using a rotational viscometer (RE550L; Toki Sangyo, Japan) and a sine-wave vibro viscometer (SV-10; A&D, Japan) at 20 °C.

IR measurements of the Al_2O_3 powders following milling were carried out by Fourier transformed infrared spectroscopic analysis (Spectrum GX; Perkin-Elmer, USA), whose sample chamber could be purged with nitrogen gas to diminish the influence of water vapor and carbon dioxide in the atmosphere.

2.3. Slip casting

Slip casting was carried out with a gypsum mold to form green bodies of 20 mm in diameter. The green bodies were dried at room temperature after slip casting and removed from the mold. Relative densities of the green bodies were estimated using Archimedes' method after calcination at 800 °C. Both the heating and cooling rates were 100 °C/h, respectively.

3. Results and discussion

Fig. 2 shows particle size distribution of Al_2O_3 particles after wet jet milling and ball milling. The particle size distribution of the wet jet-milled Al_2O_3 is identical with that of ball-milled Al_2O_3. The mean particle size (D_{50}) of the wet jet-milled Al_2O_3 is about 570 nm as well as the primary particle size. This indicates that it is possible to pulverize raw materials to the primary particle size by wet jet milling. Fig. 3 shows SEM photographs of Al_2O_3 particles before and after milling. Before milling, Al_2O_3 particles aggregate and form secondary particles. It can be seen that the starting Al_2O_3 particles are crushed to the primary particles after wet jet milling and ball milling. Thus, it is found that wet jet milling is very effective to crush the raw material into the primary particle within a short time.

Fig. 4 shows the shear stress versus shear rate curves of the wet jet-milled and the ball-milled slurry at the solid content of 30 vol%. The flow curve of wet jet-milled slurry is nearly Newtonian, whereas that of ball-milled slurry shows thixotropy. Fig. 5 shows relationship between time and apparent viscosity of slurries at different solid content prepared by wet jet milling (Fig. 5(A)) and ball milling (Fig. 5(B)). The viscosity of ball-milled slurries increases rapidly with time (Fig. 5(B)). In general, it is known that aggregation and re-flocculation of particles causes an increase of slurry viscosity and the flocculated slurry has thixotropic property.[16] Hence, these results indicate that Al_2O_3 particles in the ball-milled slurry re-flocculate because the attractive force between particles increases by the excess collision energy with ball. On the other hand, the viscosity of wet jet-milled slurries is almost constant for a long time (Fig. 5(A)). Furthermore, the viscosity of wet jet-milled slurries is very lower than that of ball-milled slurries. These results indicate that Al_2O_3 particles in the wet jet-milled slurries disperse well for long times and have no tendency to flocculate. In wet jet mill process, mixing and dispersion are carried out only by mutual collision of particles without grinding media. Since the weight of particles is very light compared with that of grinding media, the collision

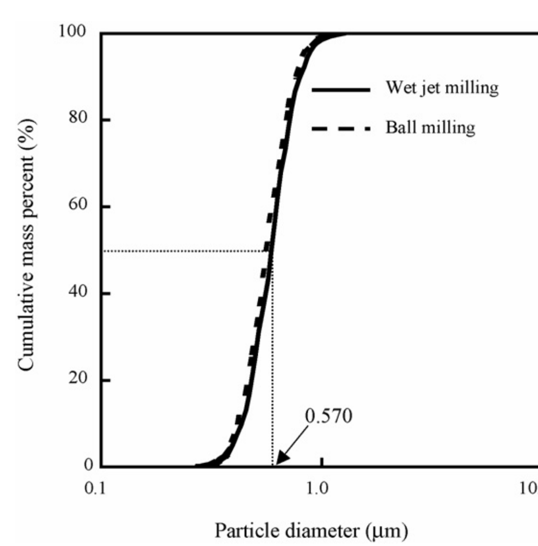

Fig. 2. Particle size distribution of Al_2O_3 particles after milling. Solid content is 30 vol%.

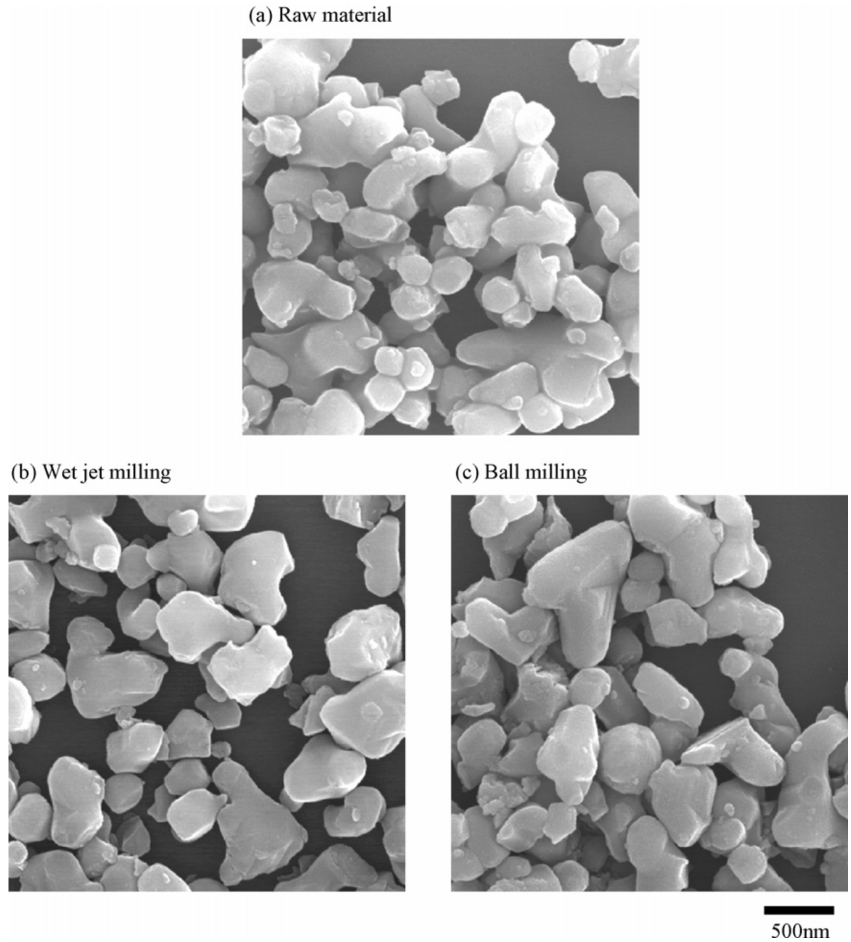

Fig. 3. SEM photographs of Al_2O_3 particles (a) before milling and after, (b) wet jet milling and (c) ball milling.

energy given to particles becomes very small. Thus, the Al_2O_3 particles in the wet jet-milled slurries maintain initial attractive force and disperse for long times after milling.

Fig. 6 presents FTIR diffuse reflection spectra of Al_2O_3 particles before and after milling at different solid contents. All spectra possess several sharp bands at around 3652, 3548 and 3480 cm^{-1}. These peaks are assigned to hydroxyl (OH) stretching vibrations (ν_{OH}) of a bayerite-type species (α-$Al(OH)_3$).[21] Spectra of the wet jet-milled Al_2O_3 powders are identical with those of the raw Al_2O_3 powders. This result indicates that Al_2O_3 particles after wet jet milling keep initial surface conditions. On the other hand, the intensities of the peaks associated with ν_{OH} of α-$Al(OH)_3$ increase and the peak at 3480 cm^{-1} shifts to 3469 cm^{-1} in the case of ball milling. Furthermore, in the spectra of the ball-milled Al_2O_3, some other new peaks appear at around 3621 and 3529 cm^{-1}. These peaks are assigned to ν_{OH} of a gibbsite-type species (γ-$Al(OH)_3$).[21] These results suggest that more OH groups are induced on the Al_2O_3 surface since the

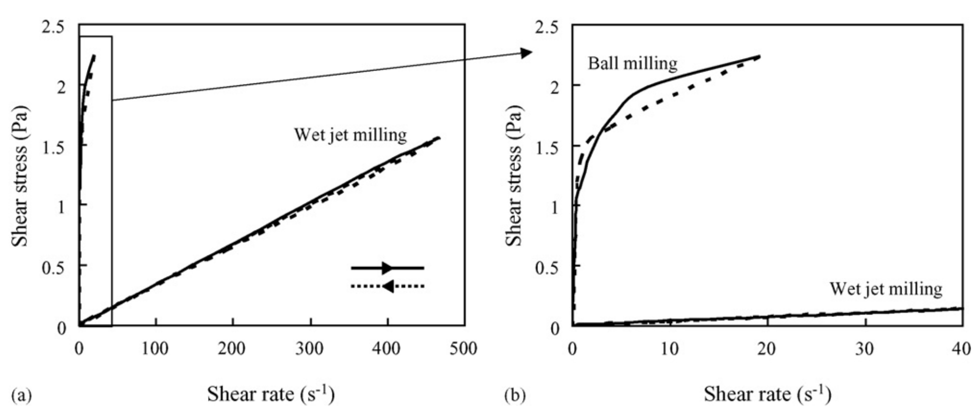

Fig. 4. (a) Shear stress–shear rate relationship of wet jet-milled and ball-milled slurries at solid contents of 30 vol%. (b) Enlarged view enclosed by the rectangle in (a).

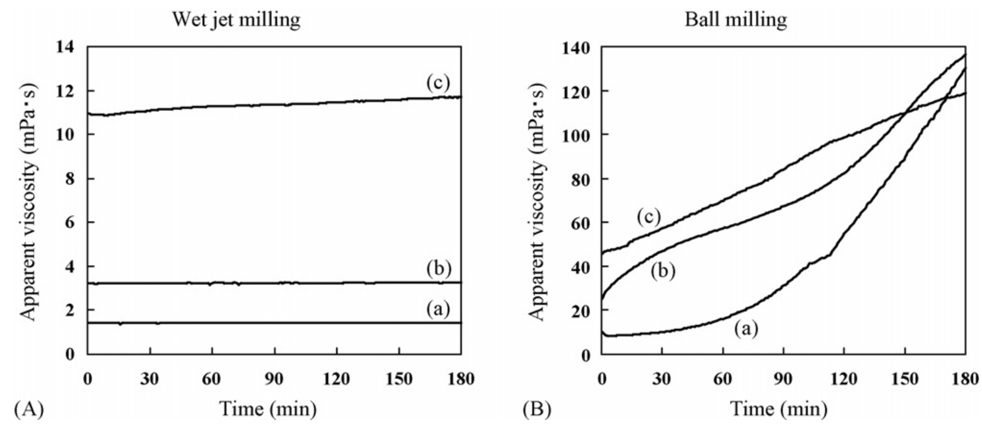

Fig. 5. Apparent viscosity of slurries prepared from (A) wet jet milling and (B) ball milling. Solid content is (a) 10 vol%, (b) 30 vol% and (c) 50 vol%.

active sites increase by the excess collision energy during ball milling.

Fig. 7 shows relative density of green bodies measured by Archimedes' method as a function of solid content. The relative densities of the green Al_2O_3 bodies prepared from wet jet-milled slurries are significantly higher than those of the green bodies prepared from ball-milled slurries. Green bodies from wet jet-milled slurries have a relatively constant relative density of about 65% or more without depending on the solid content of slurry. On the other hand, relative density of green bodies from ball-milled slurry is very sensitive to the solid content. When 10 vol% slurry is used, the relative density of green body is less than 50%, whereas the relative density is more than 60% when 50 vol% slurry is used. It is very interesting that the green body fabricated by wet jet-milled slurry with low solid content (10 vol%) shows a higher relative density than that prepared from the ball-milled slurry with a much higher solid content (50 vol%). It is known that high packing density of green body fabricated by slip casting is achieved by rearrangement of particles toward a denser packing configuration.[22,23] Tsetsekou et al.[24] also reported that the

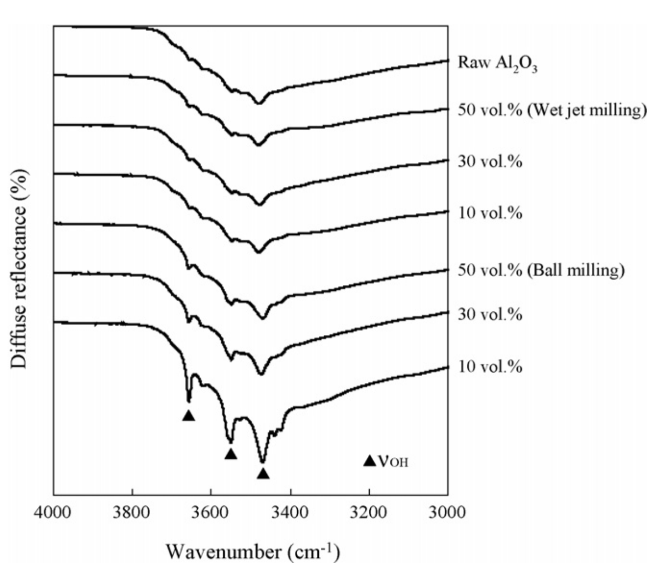

Fig. 6. FTIR diffuse reflection spectra of Al_2O_3 powders before and after milling.

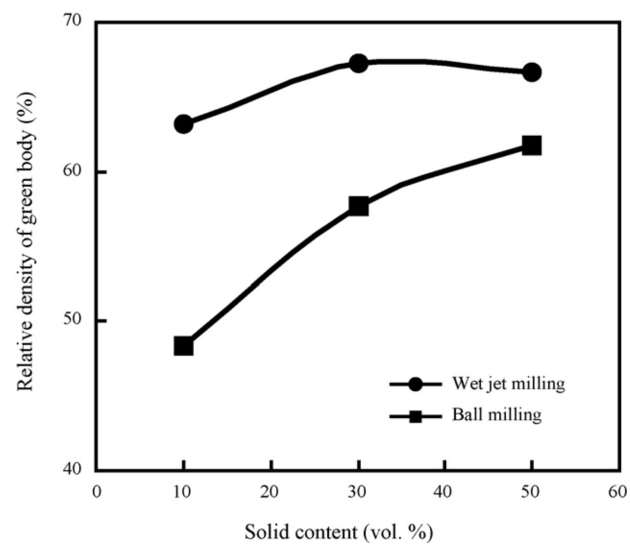

Fig. 7. Relative density of the green body as a function of solid content.

flocculated network structure impeded particle sedimentation behavior. Since Al_2O_3 particles after ball milling re-flocculate by strong attractive force, the rearrangement of particles during slip casting are disturbed. Therefore, relative density of the green body prepared from the ball-milled slurries is not so high. On the other hand, Al_2O_3 particles in the wet jet-milled slurries maintain initial attractive force and disperse well. Since good dispersion facilitate moving the particles to stable sites, Al_2O_3 particles after wet jet milling rearrange to denser packing configurations and high relative density of the green bodies about 65% or more is achieved.

4. Conclusion

Particle size distribution of Al_2O_3 particles after wet jet milling was identical with that of Al_2O_3 particles after ball milling. The mean particle size was about 570 nm as well as the primary particle size. Wet jet-milled slurry was nearly Newtonian, whereas the ball-milled slurry showed thixotropy. The viscosity of wet jet-milled slurries was very low and constant for long times indicating non-flocculation and good dispersion of particles. On the other hand, the viscosity of ball-milled slurries increased rapidly with increase in the time. Relative density of the green bodies prepared from the wet jet-milled slurries was about 65% or more without depending on the solid content of slurry, whereas that fabricated by the ball-milled slurries increased with the solid content. The relative density of green body prepared from the wet jet-milled slurry with 10 vol% Al_2O_3 was higher than that prepared from ball-milled slurry with 50 vol% Al_2O_3.

Wet jet milling process can prepare the slurry with low viscosity and flocculation property, and pulverize raw material into primary particle within a short time. This process will give a large advantage for ceramic manufacturing and product.

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