Rapid Communication

Property Modification of Ball-milled Slurries using Wet-jet Milling

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SYNOPSIS

Wet-jet milling process was employed to modify the properties of Al_2O_3 slurry after ball milling. Particle size distribution of slurry wet-jet milled after ball milling corresponded to that of ball-milled slurry. However, the viscosity of ball-milled slurry increased rapidly with time, whereas the viscosity of the slurry wet-jet milled after ball milling was stable for long times. Al_2O_3 particles after ball milling yielded more OH groups on the surface. These OH groups were reduced or removed by wet-jet milling following ball milling. Moreover, wet-jet milled slurry after ball milling had lower re-flocculation properties and higher packing density of above 60% than ball-milled slurry. Hence, Al_2O_3 particles in slurry wet-jet milled after ball milling is packed well as compared to ones of ball-milled slurry before wet-jet milling treatment.

KEY WORDS

ball milling, wet-jet milling, slurry, viscosity, packing density

1 Introduction

Ceramic slurries consisting of dispersed solid phase in an aqueous solvent are widely used in the ceramic shaping process, such as slip casting¹⁻⁷⁾ which is one of the most widely manufacturing methods in ceramics industry because it allows preparation of compacts with large sizes and complex shapes. The ceramic slurry must satisfy the following conditions to fabricate dense green body: low viscosity, high solid content and good dispersion^{8,9)}. A good dispersion of ceramic particles in the aqueous phase is achieved by strong electrostatic, steric repulsion and weak attractive interparticle van der Waals force.

In general, ceramics are manufactured via a lot of process such as pulverization of raw materials, dispersion, mixing, shape forming, sintering and machining. In the processes of pulverization, dispersion and mixing, ball milling process is widely used for preparation of a great deal of slurries. However, it is known that the ball milling requires 24 h or more to produce pulverized and dispersed slurry¹⁰⁾. Moreover, ceramic particles after ball milling have tendency to re-flocculate by an increase of the attractive force which is caused by an increase of the active sites induced by the excess collision energy with ball medium^{11,12)}. The re-flocculation of particles causes a

viscosity increase and thixotropic property¹³⁾ and leads inhomogeneous structures and low relative density of green bodies^{14,15)}.

Recently, wet-jet milling process has been demonstrated as a new method of mixing and dispersion in chemical engineering and food technology fields¹⁶⁾. In this process, the particles in the suspension or solution collide mutually at a high pressure and a high speed. In addition, the cavitation and shear flow are induced by the high speed. Using these effects, the grain refinement, emulsion, and homogenization are achieved within a short period of time. In our previous works^{17,18)}, wet-jet milling was used as a new method to prepare the concentrated ceramic slurry and it was shown that the wet-jet milled slurries had very low viscosity and low re-flocculation property.

In the present work, effects of wet-jet milling on property modification of slurries prepared by ball milling process with properties of a great deal of producibility were investigated. The properties of slurries, such as viscosity, particle size distribution and sedimentation behavior, were characterized. We will demonstrate that wet-jet milling is employed to modify the properties of ball-milled slurry.

2 Experimental procedure

A commercially available high purity $\alpha\text{-Al}_2\text{O}_3$ (AKP-20; Sumitomo Chemical, Japan) with an average particle

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size (D_{50}) of $0.57\mu\text{m}$ was used in this work. A commercially available NH_4^+ salt of poly(acrylic acid) (PAA-NH_4^+ , Aron A-6114; Toagosei Co. Ltd., Japan) was used as the dispersant. The aqueous suspension containing 30 vol.% of Al_2O_3 powder with 0.12 wt.% dispersant was prepared to obtain a dispersed slurry. The suspension was ball-milled for 24 h using high grade Al_2O_3 balls with a diameter of 10 mm. Ball milling was carried out at 60 rpm. The slurry to ball ratio is 1 : 2 in volume relation. After then, the ball-milled slurry was wet-jet milled at 200 MPa.

A schematic illustration of wet-jet milling system (PRE03-20-SP; Genus, Japan) used for this study is presented in Fig. 1. Slurry was pumped into the collision unit at 200 MPa where the mutual collision of the particles took place.

The viscosity of slurry was measured using Sine-wave vibro viscometer (SV-10; A&D, Japan) at 20°C . The viscosity was calculated from the current to move transducers placed in the slurry (a frequency of 30 Hz and an amplitude of 0.2 mm). Acoustic Spectrometer (DT-1200; Dispersion Technology, USA) was employed for particle size distribution analysis. The sedimentation tests were carried out by putting the slurries into borosilicate glass tube and allowed them to stand undisturbed for one month. The thickness of saturated sedimentation cake was measured. Slurries were dried at room temperature for 24 h in air and 110°C for 24 h under vacuum. IR measurement of the Al_2O_3 particles following milling was carried out by Fourier transformed infrared spectroscopic analysis (Spectrum GX; PerkinElmer, USA), whose sample chamber could be purged with nitrogen gas to diminish the influence of water vapor and carbon dioxide in the atmosphere. The KBr pellet method was used for spectral analysis. A transmission electron microscopy (TEM, JEM-

2010, JEOL, Japan) were used to observe the surface morphologies of Al_2O_3 particles after milling.

3 Results and Discussion

Fig. 2 shows the particle size distribution of ball milling, wet-jet milling, and wet-jet milling after ball milling. The particle size distribution of slurry prepared by wet-jet milling after ball milling is identical with that of ball-milled and wet-jet milled slurries. The mean particle size (D_{50}) as well as the primary particle size of raw Al_2O_3 materials was 570 nm. This indicates that it is possible to pulverize raw materials to the primary particle size by ball milling, wet-jet milling, and wet-jet milling after ball milling process.

Fig. 3 shows apparent viscosity of slurries prepared by ball milling, wet-jet milling and wet-jet milling after ball milling as a function of time. The viscosity of ball-milled slurry increases rapidly with time (Fig. 3 (a)). It is known that the increase in viscosity of dispersed slurry is caused by re-flocculation of ceramic particles and the flocculated slurry has thixotropic property. Thus, this increase of slurry viscosity indicates re-flocculation of Al_2O_3 particles in the ball-milled slurry. On the other hand, the viscosity of slurry wet-jet milled after ball milling (Fig. 3 (b)) is very stable and almost constant for long times as well as the wet-jet milled slurry (Fig. 3 (c)), indicating non-flocculation and low thixotropic properties.

Fig. 4 shows the sedimentation test of ball-milled slurry (Fig. 4 (a)) and wet-jet milled slurry after ball milling (Fig. 4 (b)). After one month, the slurries are separated into the two regions with the supernatant on the top and the sedimentation cake. The height of the sedimentation cake

Fig.1 Schematic illustration of wet-jet milling.

Fig.2 Particle size distribution of Al_2O_3 powder after ball milling, wet-jet milling, and wet-jet milling following ball milling.

Fig.3 Apparent viscosities of slurries prepared by (a) ball milling, (b) wet-jet milling and (c) wet-jet milling after ball milling as a function of time.

of the slurry wet-jet milled after ball milling is lower as compared to that of ball-milled slurry. Table 1 represents the normalized height percent and the relative packing density estimated from sedimentation test for ball-milled slurry and wet-jet milled slurry after ball milling. The cake heights of ball-milled slurry and slurry wet-jet milled after ball milling are 62.9 and 47.0% of the initial slurry height, respectively. The relative packing density of sedimentation cakes was calculated from the following Eq. (1);

$$\text{Relative packing density} = \text{Initial solid loading} / (\text{Cake height} / \text{Initial slurry height}) \quad (1)$$

From this result, the relative packing densities of sedimentation cakes on ball-milled slurry and slurry wet-jet milled after ball milling were estimated to be 47.7 and 63.8%, respectively. It is known that the packing density of sedimentation cake increases with decrease in the amount of flocculated particles¹⁹⁾. The packing density of

Fig.4 Appearance photographs of slurries left undisturbed for 4 weeks after (a) ball milling and (b) wet-jet milling after ball milling.

ball-milled slurry is lower than that of slurry wet-jet milled after ball milling. These results indicate that Al_2O_3 particles in slurry wet-jet milled after ball milling are packed well as compared to ones of ball-milled slurry before wet-jet milling treatment. Hence, wet-jet milling following ball milling decreases the re-flocculation property and improves the properties of ball-milled slurries.

Fig. 5 shows Fourier transform infrared diffuse reflection spectra of raw Al_2O_3 powder, Al_2O_3 powder wet-jet milled after ball milling and ball-milled Al_2O_3 powder. 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 ($\alpha\text{-Al}(\text{OH})_3$)²⁰⁾. In the case of ball-milled Al_2O_3 (Fig. 5 (c)), the intensities of these peaks increase as compared to raw Al_2O_3 (Fig. 5 (a)). However, the intensities of these peaks decrease by wet-jet milling after ball milling (Fig. 5 (b)), and the spectra is identical with that of raw Al_2O_3 . These results indicate that more OH groups are induced on the Al_2O_3 surface by ball milling, since the particle surface is damaged by high collision energy of the ball milling, and these OH groups

Table 1 Normalized height percent and the relative packing density estimated from sedimentation test for ball-milled slurry and wet-jet milled slurry after ball milling.

Method	Normalized height (%)	Relative packing density (%)
Ball milling	62.9	47.7
Wet-jet milling after ball milling	47.0	63.8

Fig.5 FTIR diffuse reflection spectra of (a) raw Al₂O₃ powder, (b) Al₂O₃ powder wet-jet milled after ball milling and (c) ball-milled Al₂O₃ powder.

are reduced or removed by cavitation and shear flow effects on the wet-jet milling following ball milling. We believe that the increase of OH groups on the surface of Al₂O₃ particles after ball milling affected the slurry properties such as viscosity, sedimentation and packing density. On the other hand, the surface of Al₂O₃ particles after wet-jet milling following ball milling is identical with raw Al₂O₃. This suggests that the Al₂O₃ particles surface damaged by

ball milling is recovered by wet-jet milling and the interaction force between particles wet-jet milled after ball milling is weak. The weak interaction contributes to the stable viscosity and high packing density.

Fig. 6 shows TEM micrographs of Al₂O₃ particles after ball milling and wet-jet milling following ball milling. After ball milling, irregularity of crystal structure was seen on the surface of Al₂O₃ particle (Fig. 6 (a)). It is suggested that the irregularity were induced on the Al₂O₃ particle surface by ball milling. Moreover, FTIR spectrum from ball-milled Al₂O₃ powders (Fig. 5 (c)) also supports the result of TEM observation (Fig. 6 (a)) and the generation of many hydroxyl (OH) groups on the surface of particles is occurred by ball milling. Therefore, the ball-milled slurry was unstable (Fig. 3 (a)). On the other hand, the surface of Al₂O₃ particle after the wet-jet milling following ball milling was clear and irregular structure could not be observed on the particle surface (Fig. 6 (b)). Thus, wet-jet milling process removes the irregular structure that is fabricated on particle surface by ball milling and leads to the stable slurry.

These results indicate that wet-jet milling is useful to modify the slurry properties after ball milling.

4 Conclusion

Al₂O₃ particles after ball milling had more OH groups on the surface, which enhances the particle-particle interaction which in effect accelerate re-flocculation. Wet-jet milling following ball milling decreased the re-flocculation property and improved the stability of the ball-milled slurry, and reduced the OH groups on the surface of Al₂O₃ particles. The slurry properties, such as viscosity, particle size distribution and sedimentation behavior were

Fig.6 TEM micrographs of Al₂O₃ particles after (a) ball milling and (b) wet-jet milling following ball milling.

measured. Particle size distribution was identical to ball milled slurry and wet-jet milled slurry after ball milling. Relative packing density of sedimentation cake produced by the slurry wet-jet milled after ball milling was estimated to be 63%, indicating less flocculation and high packing between particles. These results indicate that wet-jet milling is useful to modify the slurry properties after ball milling.

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