Improved Ceramics through New Measurements, Processing, and Standards

Edited by
Minoru Matsui
Said Jahanmir
Hamid Mostaghaci
Makio Naito
Keizo Uematsu
Rolf Wäsche
Roger Morrell

Ceramic Transactions
Volume 133

PARTICLE SIZE CHARACTERIZATION OF HIGHLY CONCENTRATED ALUMINA SLURRIES BY ULTRASONIC ATTENUATION SPECTROSCOPY

Shin-ichi Takeda, Hiroyuki Harano, Isao Tari
Faculty of Engng. Okayama University, 3-1-1 Tsushimanaka, Okayama 700-8530, Japan

ABSTRACT

Acoustic attenuation spectroscopy can characterize the particle size distribution (PSD) of highly concentrated slurries. This technique can directly measure the PSD of dispersions in highly concentrated slurries without considering the possible change of the PSD by diluting prior to measurement. The present study investigated a correlation between macroscopic and microscopic properties of alumina slurry when the added amount of dispersant was changed.

It is found that the zeta potential of the Al_2O_3 particles is not a factor of controlling the viscosity of alumina slurry, but particle size distribution width, represented as log (d_{84}/d_{50}), and the amount of large particle fraction (LPF) affect the viscosity of alumina slurry when all the dispersant was adsorbed on to the alumina surface. When non–adsorbing dispersant was present in the slurry, the PSD and log (d_{84}/d_{50}) cannot be related with the behavior of viscosity. The obtained results demonstrated that the acoustic attenuation spectroscopy can open the way for correlating between macroscopic property such as viscosity and microscopic properties of slurry.

INTRODUCTION

Ceramics processing based on powder technology includes many steps from preparation of raw materials to sintering of cast body. Each step is important for producing the products with desired microstructure and properties. In liquid based forming process of ceramics, such as slip casting, it is reported that the control of viscosity in the slurry is one of the important factors for preparing highly reliable and reproducible ceramics [1]. Thus, the characterization of slurries is critical to allowing insight into slurry properties and how they can be manipulated into generating the desired slurry characteristics and the resulting properties of the cast body. Many techniques have been used to characterize slurries. Techniques that measure macroscopic slurry properties include rheological measurements, settling test, and measurement of packing density of cast bodies [2]. But insight into the microscopic slurry properties such as particle size distribution and surface charge has not been obtained, because most of the techniques for particle sizing and determination of surface charge were restricted to very diluted suspensions.

Recently, acoustic and electroacoustic spectroscopy are developing rapidly as an alternative to light scattering methods [3-5]. The ability to characterize concentrated disperse systems provides much of the impetus for these developments. Therefore, this study investigated a correlation between macroscopic and microscopic properties of slurry when the added amount of dispersant was changed.

EXPERIMENTAL PROCEDURE

Commercially available Al_2O_3 powder was used in this study; AL160SG4 (Showa Denko Co., Yokohama, Japan) with a BET specific surface area of 7.0 m^2/g. The median particle size reported by the manufacture was 0.50 μm, which is the size measured by the laser diffraction method in a well-dispersed diluted suspension. The chemical analysis results give: Al_2O_3 99.89%, SiO_2 0.026%, Fe_2O_3 0.020%, CaO 0.015%, MgO 0.004%, Na_2O 0.041%, K_2O 0.001%. The polymer dispersant used in this study was ammonium polyacrylate (D-134, Dai-ichi Kogyo Seiyaku Co., Ltd., Japan) with an average molecule weight of 10,000.

Al_2O_3 powder was mixed into water with different amount of added dispersant (0.25～1.0 mass% on the basis of Al_2O_3 weight), then ball milled for 4h. The solid loading in the slurry was fixed at 46vol%. The viscosity of prepared slurry was determined by a cone-and-plate viscometer at a shear rate of 192s^{-1}. The pH of the slurry was measured with a pH meter. Particle size distribution in highly concentrated slurry was measured by ultrasonic attenuation spectroscopy (DT-1200; Dispersion Technology, Inc., NY, USA) and the zeta potential of Al_2O_3 particle with various amounts of polymer dispersant were measured by colloidal vibration current. The volume fraction occupied by flocculated particles relative to all the particles in the slurry (referred to as Large Particle Fraction (LPF)) was evaluated from the analysis of measured attenuation curves using the system software. LPF was calculated only for the case where the better curve fitting can be obtained for attenuation curve if the bi-modal distribution was assumed. Bi-modal distribution is defined as the normalized sum of the two lognormal distributions. The mode with the smaller size is referred to as the first mode whereas the mode with the larger size is the second one. LPF equals to the ratio of second mode volume to the volume of the total solid content. The adsorbed fraction of polymer dispersant on Al_2O_3 particle was determined by measuring the amount of free polymer in the supernatant solution, which was obtained from alumina slurry by centrifugal sedimentation at 10,000 rpm for 15 min. The amount of free polymer was measured using by total carbon analyzer (TOC-5000, Shimadzu Co., Ltd., Japan).

RESULTS AND DISCUSSION

The rheological behavior of alumina slurries represented by apparent viscosity and the pH of the slurry as a function of the added amount of dispersant are shown in Figure 1. Prepared mixtures of powder and solution were so viscous that measurements could not be made at less than about 0.2mass% dispersant (referred to as region Ⅰ). It is also observed that apparent viscosity does not change monotonically and changes in a complicated manner when the added amount of dispersant was increased; apparent viscosity has two minima when added amount of dispersant was at 0.25mass% and 0.50mass%. The pH of the slurry decreased slightly (from pH 9.1 to pH 8.5) with increasing the added amount of dispersant.

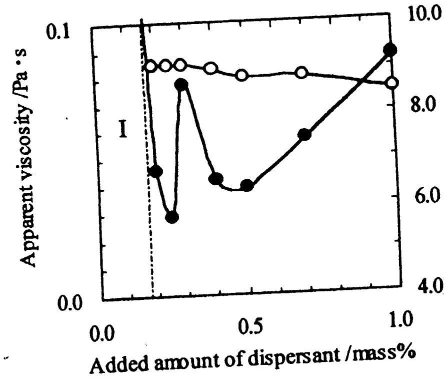

Fig.1 Apparent viscosity (●) and pH (○) of alumina slurry as a function of the added amount of dispersant.

In order to clarify the correlation between the viscosity and the amount of polymer dispersant adsorbing on the alumina particle, the adsorption density of dispersant was determined. The adsorption density of polymer dispersant on to alumina surface is shown in Figure 2. The 100% adsorption line means that all the dispersant added to the slurries adsorbed on to alumina surface. The result is that adsorption density of dispersant increases with increasing the added amount of dispersant and the adsorption density of dispersant deviates from the 100% adsorption line when the added amount of dispersant is at about 0.3mass%. It is also observed that all the dispersant added to the slurries adsorbed on to alumina surface when the added amount of dispersant ranged from 0.2 to 0.3mass% (referred to as region Ⅱ), on the other hand non-adsorbing dispersant was present in the slurry when the added amount of dispersant ranged from about 0.3 to 1.0mass% (referred to as region Ⅲ). It was proved that the complicated changing manner of the viscosity of alumina slurries couldn't be correlated with the adsorption density of dispersant. From this

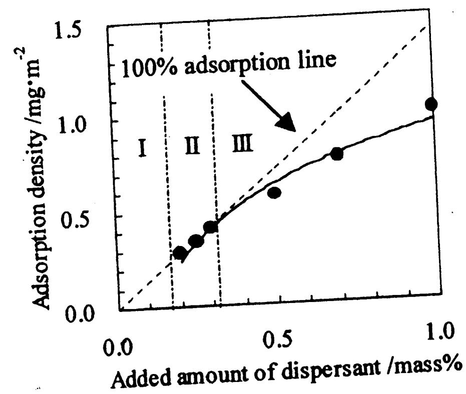

Fig.2 Adsorption density of dispersant on to alumina surface as a function of the added amount of dispersant.

observation, discussion hereafter on the macroscopic and microscopic properties of slurry will be made for each region after dividing into three regions of Ⅰ, Ⅱ and Ⅲ. Zeta potential of alumina particles and the PSDs (median size, distribution width, LPF) of alumina slurries with various amount of dispersant were measured in order to clarify a correlation between macroscopic property such as apparent viscosity and microscopic properties such as *in situ* PSD and zeta potential. The zeta potential of the Al_2O_3 particles as a function of the added amount of dispersant was measured by colloid vibration current, as shown in Figure 3. It is clearly found that measured zeta potential of Al_2O_3 particles does not change with increase in the added amount of dispersant. This result demonstrates that the change in viscosity of slurry cannot be explained by considering the change in zeta potential of alumina particles. This implies that other factors affect the values of viscosity.

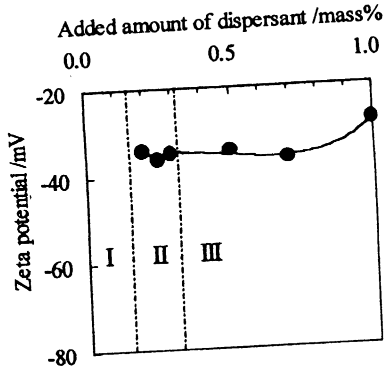

Fig.3 Zeta potential of alumina particle as a function of the added amount of dispersant.

Figure 4 shows particle size distribution of slurries at various amount of dispersant. The PSDs in alumina slurry show a bi-modal distribution. The smaller peak corresponds to the primary size of the alumina particle (0.50 μm) as reported by the manufacture, and the larger peak should be corresponding to the flocculated size (2.3 μm～3.3 μm). Figure 5 shows a median size of primary particle, flocculated particle size and distribution width of PSD as a function of the added amount of dispersant. The primary particle size as a function of the added amount of dispersant was found to remain constant. The flocculated particle size increased from 2.5 μm to 3.3 μm in region Ⅱ and decreased with increasing the added amount of dispersant in region Ⅲ. The changing manner of median size with added amount of dispersant cannot be related with that of the viscosity.

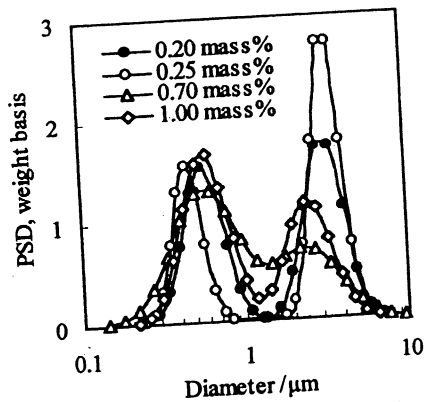

Fig.4 Particle size distribution of alumina slurry as a function of the added amount of dispersant.

It is observed that the changing behaviors of distribution width is similar to the behavior of viscosity in region Ⅱ. This result indicates that distribution width is one of the factors affecting the viscosity. On the other hand, in region Ⅲ the change of distribution width with increasing of the added amount of dispersant shows a slightly different manner of change in viscosity. This difference might imply that non-adsorbing polymer has an effect on the viscosity. The same conclusion has been already drawn and reported in the literature by Hackley [6]. In the case of a slurry having a bimodal distribution, it is important for characterizing the microscopic properties to take into consideration the fraction of occupied by primary particles and flocculated particles. Figure 6 shows LPF in alumina slurry as a function of the added amount of dispersant. LPF represents the volume fraction occupied by flocculated particles relative to all the particles in the slurry. It is observed that the LPF has a maximum when the added amount of dispersant is at 0.25mass% where distribution width and apparent viscosity has a minimum. This result suggested that apparent viscosity of alumina slurry has a minimum when LPF has a maximum and PSD distribution width has a minimum. A correlation between macroscopic property such as apparent viscosity and microscopic property such as *in situ* PSD is summarized in Figure 7.

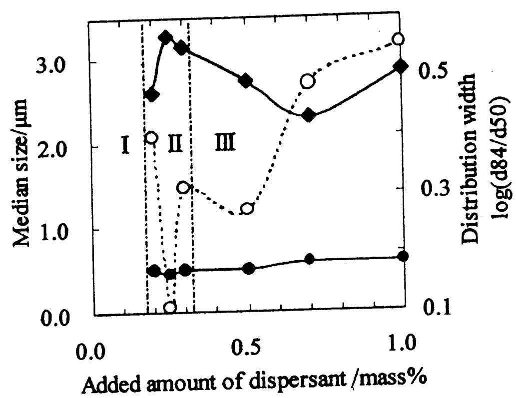

Fig.5 Median size (primary particle size (●), flocculated particle size (◆)) and distribution width (O) of alumina slurry as a function of the added amount of dispersant.

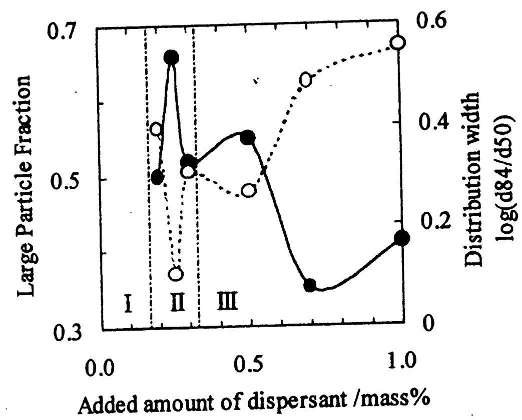

Fig.6 LPF (●) and Distribution width (O) of alumina slurry as a function of the added amount of dispersant.

SUMMARY

This study investigated a correlation between macroscopic and microscopic properties of alumina slurry when the added amount of dispersant was changed. The results clarify the factors controlling the viscosity of highly concentrated alumina slurries, and are summarized as follows:

The zeta potential of the Al_2O_3 particles is not a factor for controlling the viscosity of alumina slurry, but particle size distribution width, represented as log (d_{84}/d_{50}), and large particle fraction (LPF) affects the viscosity of alumina slurry when all of the dispersant was adsorbed on to the alumina surface. When non–adsorbing dispersant was present in the slurry, the changing manner of median size and log (d_{84}/d_{50}) cannot be directly related with the behavior of viscosity.

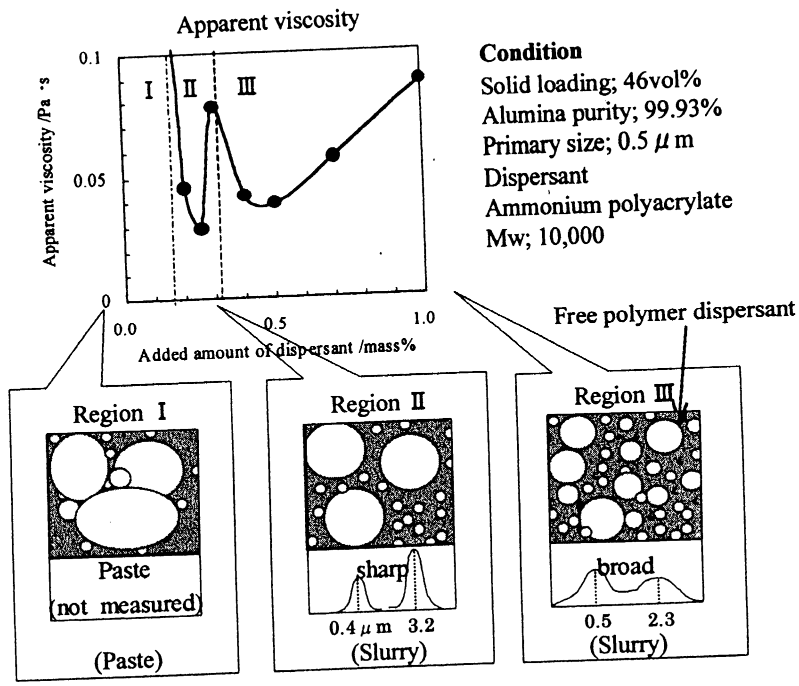

Fig.7 A correlation between apparent viscosity and particle size distribution of alumina slurry as a function of the added amount of dispersant.

ACKNOWLEDGEMENT

The present work was partly supported by Grand-in-Aid from The Ministry of Education, Science, Sport and Culture of Japan (No. 12650671).

REFERENCE

[1]G. Tari, J. M. F. Ferreira and O. Lyckfeldt, Influence of Magnesia on Colloidal Processing of Alumina, *Journal of the European Ceramic Society,* **17** 1341-1350 (1997).

[2]H. Kamiya, K. Isomura, G. Jimbo and J. Tsubaki, *Journal of the American Ceramics Society,* **78** 46-57 (1995).

[3]Andrei S. Dukhin, Philip J. Goetz, Vincent Hackley, Modified Log-normal Particle Size Distribution in Acoustic Spectroscopy, *Colloids and Surfaces; A,* **138** 1-9 (1998).

[4]A. S. Dukhin and P. J. Goetz, Acoustic Spectroscopy for Concentrated Polydisperse Colloids with Highly Density Contrast, *Langmuir,* **12** [21] 4987-4997 (1996).

[5]A. S. Dukhin and P. J. Goetz, Acoustic and Electroacoustic Spectroscopy, *Langmuir,* **12** [19] 4334-4344 (1996).

[6]V. A. Hackley, Colloidal Processing of Silicon Nitride with Poly(acrylic acid): II, Rheological Properties, *Journal of the American Ceramics Society,* **81** 2421-28 (1998).