Slip Casting of Al_2O_3 Slurries Prepared by Wet Jet Milling

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湿式ジェットミル処理したアルミナスラリーの鋳込み成形

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Green body fabricated by slip casting using wet jet milled slurry, which had low viscosity and low re-flocculation property, had very high relative density and showed very small shrinkage during sintering. The square of the thickness of green bodies was proportional to casting time indicating slip casting was implemented well. The relative density of the green body prepared by wet jet milled slurry was 67%, markedly higher than that of ball milled slurry (58%). After sintering, the compacts prepared by wet jet milled and ball milled slurry had a similar relative density of 99%. The linear shrinkages of the sintered bodies prepared by wet jet milled and ball milled slurry were 11.5 and 16.8%, respectively.

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1. Introduction

Slip casting^{1)–11)} is one of the most useful forming methods and is widely applied for the commercial production of ceramics because it is able to form compacts with large size and complex shape easily. In this process, the dispersion of ceramic powder in an aqueous medium is required. These aqueous slurries for slip casting must satisfy the following conditions to produce high dense green bodies: (1) low viscosity, (2) high solid content, (3) good dispersion and (4) optimum particle size distribution.^{3),4),12),13)}

In general, ball mill process is widely used to prepare the ceramic slurries.^{13)–15)} However, it is known that ball milling requires 24 h or more to produce good slurry.¹⁶⁾ In addition, ceramic particles prepared by ball milling have tendency to re-flocculate.^{17),18)} The re-flocculation causes a viscosity increase and thixotropic property.¹⁹⁾ The flocculated particles lead inhomogeneous and loose structure of green bodies, i.e., broad pore size distribution and low relative density.^{1),10)} Inhomogeneous and low dense green body warps and cracks during the drying and sintering process. Our previous work²⁰⁾ showed that 30 vol% Al_2O_3 slurries prepared by wet jet milling had very low viscosity of 6 mPa·s, low re-flocculation and low thixotropic properties. This process can pulverize raw materials till primary particle size and prepare good slurry within a short time.

The aim of the present work is to produce high dense green bodies by slip casting using low viscosity and low thixotropic slurry prepared by wet jet milling, and to investigate the casting and sintering properties. We will demonstrate that the green body with high relative density can be fabricated using wet jet milled slurry by slip casting.

2. Experimental procedures

A commercially available high purity $\alpha\text{-Al}_2\text{O}_3$ (AKP-20, Sumitomo Chemical Co., Ltd.) having a mean particle size of 0.5 μm was used. A commercially available NH_4^+ salt of poly (acrylic acid) (Aron A-6114; Toagosei Co., Ltd.) was used as dispersant. A suspension was prepared by dispersing Al_2O_3 in 70 vol% of distilled water with 0.12 mass% dispersant. The suspension was stirred for 2 h before wet jet milling. A labora-

tory scale wet jet mill system (PRE03–20–SP; Genus, Japan) was used for this study. The slurry was pumped into collision-unit once at 200 MPa, in which the particles in the slurry collide mutually. For the purpose of comparison, the same Al_2O_3 slurry was also prepared by ball milling. The ball milling was performed for 24 h using high grade Al_2O_3 balls with a diameter of 3 mm.

The rheological characteristic of the slurries was measured using Sine-wave vibro viscometer (SV-10; A&D, Japan) at 20°C.

Slip casting was carried out with a gypsum mold, and green bodies of 20 mm in a diameter were formed. The green bodies were dried in air after slip casting and removed from the mold. Relative density of the green bodies were estimated using Archimedes method after calcination of 800°C. The green bodies were sintered at 1600°C for 2 h in air. Both the heating and cooling rates were 100°C/h, respectively. The relative density of the sintered bodies was determined by Archimedes method. Fracture surface of green bodies was observed by scanning electron microscopy (S-4300; Hitachi Ltd.).

3. Results and discussion

Figure 1 shows the change in apparent viscosity of slurries with time. The apparent viscosity of wet jet milled slurry is very lower than that of ball milled slurry. The viscosity of ball milled slurry increases rapidly with time. In general, it is known that aggregated and flocculated slurry causes a increase of viscosity.^{1),19)} Hence, this rapid increase of viscosity indicates that the Al_2O_3 particles in the ball milled slurry aggregate and flocculate. On the other hand, the viscosity of wet jet milled slurry is constant indicating non-flocculation and low thixotropic property.

Figure 2 shows the relationship between square of the thickness of green body and casting time. The square of the thickness of green body is proportional to casting time. The gradient of the straight line of wet jet milled slurry is very lower than that of ball milled slurry. This means that casting rate of wet jet milled slurry is lower than that of ball milled slurry.

Generally, the casting behavior can be quantified as a filtration process on the base of Darcy's law,^{21),22)}

Fig. 1. Apparent viscosity of slurries prepared by wet jet milling and ball milling.

Fig. 2. Relationship between square of the thickness of green body and casting time.

$$Q = \frac{dV}{Adt} = \frac{k\Delta P}{L} \quad (1)$$

where Q is the fluid flux (m^3/s), V is the filtrate volume (m^3), A is the filter area (m^2), k is the permeability of the filter ($\text{m}^2/\text{Pa}\cdot\text{s}$), ΔP is the pressure difference across the filter (Pa) and L is the thickness of the filter (m).

From Eq. (1),

$$L^2 = (k\Delta P)t \quad (2)$$

where t is the slip casting time.

Equation (2) indicates that the square of the thickness of green body formed by slip casting is proportional to the casting time. As shown in Fig. 2, the square of the thickness of green bodies produced by slip casting with wet jet milled and ball milled slurries is proportional to casting time. This result indicates that the slip casting is implemented well with wet jet milled and ball milled slurry. The gradient of the straight line can be expressed by permeability of green body (k) and capillary pressure of gypsum mold (ΔP). The permeability of green body is represented as follows²³⁾

$$k = \frac{2E^3}{5S^2\rho\eta(y-1)(1-E)^2} \quad (3)$$

$$y = \frac{X}{V} \quad (4)$$

where E is the porosity of green body ($-$), S is the specific

Table 1. Relative Density of Green Body

Method	Wet jet milling	Ball milling
Relative density (%)	67.2	57.7

surface area of raw material (m^2/g), ρ is the density of raw material (g/m^3), η is the apparent viscosity of solvent ($\text{Pa}\cdot\text{s}$) and X is the volume of slurry containing volume V of solids ($-$), i.e., y is the reciprocal of slurry concentration.

In this work, slip casting was carried out on the same condition (gypsum mold, Al_2O_3 powder, concentration of dispersant, slurry concentration). Thus, all parameters are constant in the present works except the porosity of green body (E). The porosity of green body only influences on the gradient and casting rate. Some parameters in the present work are given below:

Specific surface area of Al_2O_3 particle (S) : $4.0 \text{ m}^2/\text{g}$
 Density of Al_2O_3 (ρ) : $3.98 \times 10^6 \text{ g}/\text{m}^3$
 Viscosity of solvent (η) : $1.1 \times 10^{-3} \text{ Pa}\cdot\text{s}$
 Reciprocal of slurry concentration (y) : 3.33
 Capillary pressure of gypsum mold (ΔP) : $1.5 \times 10^5 \text{ Pa}$

The capillary pressure of gypsum mold (ΔP) is calculated using Eq. (3) from the relative density of green body produced by stirred Al_2O_3 slurry. In this study, permeability of green bodies (k) produced by wet jet milled and ball milled slurry can be calculated using Eq. (2) from the gradient of the straight line in Fig. 2, and is 3.78×10^{-14} and 1.76×10^{-13} ($\text{m}^2/\text{Pa}\cdot\text{s}$), respectively. By substituting these parameters for Eq. (3), the porosity of green bodies prepared by wet jet milled and ball milled slurry can be estimated and is 0.315 and 0.452, respectively. **Table 1** shows relative density of green bodies measured by Archimedes method. The relative density of green Al_2O_3 bodies prepared by wet jet milled and ball milled slurry is 67.2 and 57.7%, respectively. Thus, the porosity of green bodies is 0.328 and 0.423, respectively. These values correspond well with the calculation values of porosity. **Figure 3** shows the fracture surface of green bodies prepared by wet jet milled and ball milled slurry. It is clearly seen that Al_2O_3 particles prepared by ball milling form aggregates. On the other hand, each Al_2O_3 particles in the compact prepared by wet jet milled slurry disperse and pack well.

The difference in the relative density and morphology of the fracture surface between the green bodies prepared by wet jet milled and ball milled slurry is attributed to viscosity and flocculation property of slurry. It is known that high packing density of green body is achieved by rearrangement of particles.²⁴⁾ The wet jet milled slurry has low viscosity, low flocculation and low thixotropic properties (Fig. 1). These properties of slurry facilitate moving the particles to stable sites. Hence, Al_2O_3 particles prepared by the wet jet milling can easily move and rearrange, and very high relative density of about 67% is achieved. On the other hand, the ball milled slurry has high viscosity, high flocculation and high thixotropic properties. Hence, Al_2O_3 particles aggregate during slip casting and pack loosely indicating low relative density.

Figure 4 shows appearance photographs of the compacts sintered at 1600°C for 2 h in air. The circles drawn in broken line express the size of green body prepared by wet jet milled and ball milled slurries. Both green bodies shrink during the sintering. The shrinkage of sintered body produced by wet jet milled slurry is smaller than that produced by ball milled slurry. **Table 2** shows the relative density and linear shrinkage

Fig. 3. Fracture surface of green bodies prepared by (a) wet jet milled slurry and (b) ball milled slurry.

Fig. 4. Appearance photographs of the sintered bodies prepared by (a) wet jet milled slurry and (b) ball milled slurry. The circle of broken line shows the size of green body.

Table 2. Relative Density and Linear Shrinkage Along the Diametrical Direction of Sintered Body

Method	Wet jet milling	Ball milling
Relative density (%)	99.1	99.0
Linear shrinkage (%)	11.5	16.8

along the diametrical direction of sintered bodies. The sintered bodies prepared by wet jet milled and ball milled slurry has a similar density of 99%. Linear shrinkage of sintered bodies prepared by wet jet milled and ball milled slurry is 11.5 and 16.8%, respectively. Green bodies which particles are well-packed show very small shrinkage after sintering. Thus, the wet jet milled slurries have an advantage of forming design, e.g., near net shaping, than ball milled slurries.

4. Conclusion

Al_2O_3 slurry prepared by wet jet milling had very low viscosity, low flocculation and low thixotropic properties. Hence, the dispersed Al_2O_3 particles, which were prepared by wet jet milling, packed well. The green body produced by wet jet milled slurry had high relative density. This high relative density of green bodies led small shrinkage during sintering. On the other hands, Al_2O_3 slurry prepared by ball milling had

high viscosity, high flocculation and high thixotropic properties. Hence, Al_2O_3 particles aggregated during slip casting, and relative density of green body was about 10% lower than that produced by wet jet milled slurry. This low relative density of green bodies caused large shrinkage.

The wet jet milled slurry is useful for the dimensional accuracy of the ceramic products.

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