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The effect of packing structure of powder particles on warping during sintering

Napat Chantaramee*, Satoshi Tanaka, Keizo Uematsu

Department of Materials Science and Technology, Nagaoka University of Technology, 1603-1 Kamitomioka, Nagaoka, Niigata 940-2188, Japan

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Abstract

In this study, aqueous alumina suspension consisting of constant solid loading was chosen as a model system in order to observe the effect of particle packing structure in green tape on sintering deformation. The specific particle packing structure in consolidated green tape was intentionally formed during tape casting, for instance, a non-uniform particle packing structure (Type-I) and a relatively uniform particle packing structure (Type-II). The development of deformation in sintered specimens was explainable from anisotropic packing structure. Alumina tapes with Type-I structure particularly showed warping which was transverse to the casting direction. Tapes with Type-II structure appeared with irregular distortion which was expected as a result of the localized non-uniform particle packing structure.

Keywords: Packing structure; Warping; Tape casting; Al_2O_3; Sintering

1. Introduction

Casting affects the characteristics of green tape markedly in the doctor blade process. The origin of this strong effect is ascribed to the specific structure of consolidated tape, which is developed during the casting process. The flow of slurry during and immediately after casting should be responsible for the development of structure in the tape. It is very important to understand the evolution of structure in the green tape and its relevance to the shrinkage behavior in sintering.

The arrangement of particles into a specific direction under shear flow is often noted in studies with extremely anisotropic particle shapes.[1–5] The long-axes of particles are highly oriented in the flow direction (*x*-direction) in these studies. Simulation study also showed that even particles with relatively low aspect ratio should align under shear flow in the same manner.[6] Therefore, particle alignment is frequently mentioned as the cause of anisotropic shrinkage in the casting and transverse directions. However, there have been no detailed investigations on the correlation between the particle arrangement and the distortion which is governed by non-uniform shrinkage in the cast tape.

Our previous study showed non-uniform structure within alumina green tapes prepared by the doctor blade process.[7] The processing parameters affect the packing structure of particles markedly. The long-axes of alumina particles aligned along the casting direction from the top surface (air-side) to near the lower surface (carrier-side), then gradually deviated off direction as closely approaching the lower surface. This non-uniform particle packing structure may lead to tape distortion during sintering. Therefore, in this study, the objective is to show the significance of particle packing structure on sintering deformation, with a particular focus on the effect of localized non-uniform structure.

2. Experimental procedure

Casting slurries were prepared by mixing alumina powders (AL160SG1, Showa Denko, Japan) in distilled water containing dispersant (Ammonium polyacrylate-type, D305, Chukyoyushi, Japan). The mixture was milled in a PE bottle with alumina balls for 24 h and de-aired for several minutes. After de-airing, glycerol (Kanto Chemical, Japan) and polyvinyl alcohol (JL-05E, Japan Vam & Poval Co., Ltd., Japan) were added to the slurry, and gently mixed by a magnetic stirrer. Rheological

* Corresponding author. Tel.: +81 258 47 9317; fax: +81 258 47 9337.
E-mail address: chantaramee_napat@mst.nagaokaut.ac.jp (N. Chantaramee).

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behavior was determined using a viscometer (VT550, HAAKE, Germany) equipped with a coaxial cylinder sensor system (ISO3219/DIN53019). Details of slurry compositions, preparation procedure and rheological measurement were described in a previous study.[7]

Tape casting was performed by a laboratory tape casting bench with a stationary double blade system (DP-150, Sayama Riken, Japan). The gap height from the carrier to the blade was controlled by gauge micrometers with a precision of 0.01 mm. The gap height parameter was kept constant for all tape casting experiments. The first blade, with the exit length of 1.1 mm, was set to 0.3 mm in height. The second blade gap was set to 1 mm. Casting speed was adjusted in order to induce the desired structure.[7] The first type of structure (Type-I) is as schematically illustrated in Fig. 1(a). The long-axes (*a*- or *b*-axis) of alumina particles embedded in the upper region align in the casting direction (*x*-axis), but those in the lower region gradually deviate from the casting direction. In the second (Type-II) structure which is composed of basically uniform particle packing as shown in Fig. 1(b), the long-axes of alumina particles are aligned in the casting direction except some local fluctuation in the direction of orientation. To confirm particle packing structures in the green tape were as mentioned in Fig. 1, the particle packing structure was examined using a polarized light microscope (OPTIPHOT2-POL, Nikon, Japan).[7–9] At least 10 specimens were prepared for structure examination. A sensitive tint plate ($n = 530$ nm) was used to enhanced the colouration which directly exhibited the direction of particle alignment. The main observation was noted on the *x*–*z* plane, where *x*- and *z*-axis signify the casting direction and the tape thickness direction, respectively. The packing structure view on the *y*–*z* plane and the *x*–*y* plane is excluded since it was correlated to the structure observed in the *x*–*z* plane.[7]

The dried green tape with uniform thickness was punched into circular specimens with approximately 21 mm diameter, in order to avoid the effect of non-symmetrical shape on densification. For each specimen, a mark was introduced to trace the casting direction. After placed on an alumina substrate, the specimen was heated in an electric furnace (heating rate 10 °C/min) and

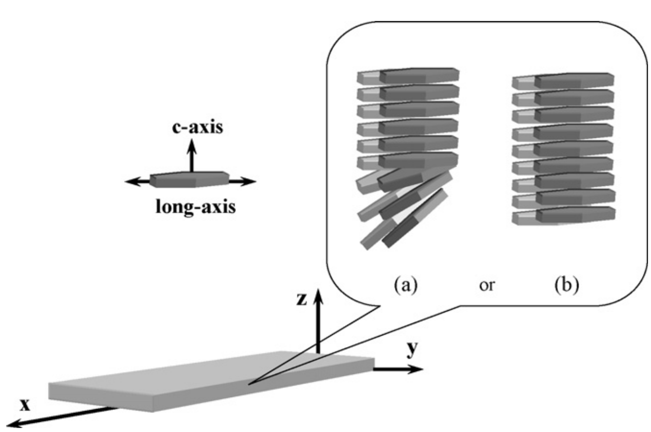

Fig. 1. Diagram illustrating alumina particle packing structure in *x*–*z* plane (*x*: casting direction; *y*: cross-casting direction; *z*: thickness direction). (a) Type-I structure; (b) Type-II structure.

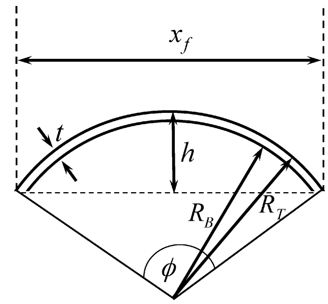

Fig. 2. Schematic of geometry used for calculating the linear shrinkage at the top plane and the bottom plane of the cast tape.

held at 1200, 1300, 1400, 1500 or 1600 °C for 2 h. For each run, at least 10 specimens were used; half the specimens were placed with the surface of the carrier side down and the other half up. After sintering, the warping height was measured for all specimens by a micrometer with the precision of 0.01 mm. The warping direction in reference to the casting direction was also noted.

Linear sintering shrinkage in the plane of the top surface and bottom surface of the cast tape were calculated bases on the warping height (h) data and chord (x_f), which is measured directly using a traveling microscope with an accuracy of 0.01 mm. Fig. 2 shows schematic of geometry used for the calculation. The radius for the top surface of the sample (R_T) is obtained from the relation given in the following equation:

$$R_T = \frac{h^2 + (x_f^2/4)}{2h} \tag{1}$$

The radius for the bottom plane (R_B) is given by the following equation:

$$R_B = R_T - t \tag{2}$$

where t is the thickness of the tape after sintering. The central angle of an arc (ϕ) was measured from the circular sector. The arc length of the top plane (S_T) and bottom plane (S_B) were calculated from their circular radius multiplies by the central angle of an arc ($S = R\phi$). Then, the arc length S_T and S_B were further used for linear sintering shrinkage evaluation.

3. Results

Fig. 3 shows a cross polarized light image of Type-I structure taken on the *x*–*z* plane. The photographs were taken with a sensitive tint plate. The long-axes (*a*–*b* plane of crystal) of alumina particle tended to align along the casting direction in the blue region of the micrograph (Fig. 3(a)). The variation of blue color (Fig. 3(a)–(c)) indicates a gradual change in the direction of long-axes. Hence, it confirmed the packing structure which was mentioned in Fig. 1(a).

Fig. 4 shows cross polarized light image of Type-II structure taken on the *x*–*z* plane. The blue zone (Fig. 4(a) and (b)) also indicates the aligned long-axes in casting direction. Though, the long-axes are aligned predominantly in the casting direction, however the streaky area (Fig. 4(b)) indicates the existence of localized non-uniform alignment across the tape

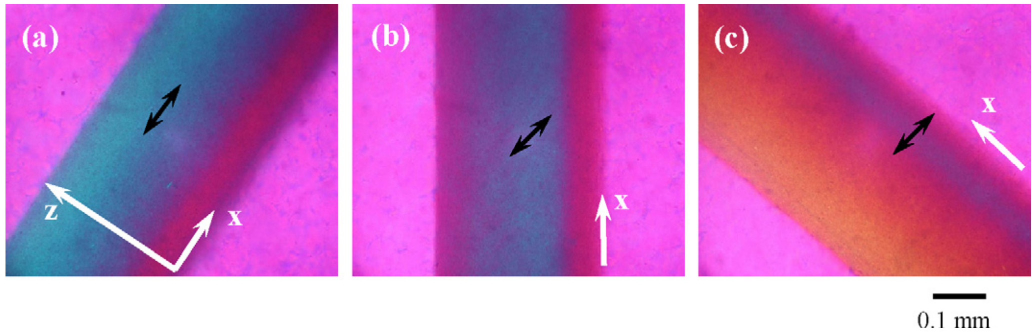

Fig. 3. Cross polarized light image of Type-I structure taken on the x–z plane. Photographs were taken at 45° from maximum extinction. Blue color represent for (a) long-axis aligned in x-axis, (b) long-axis gradually turned and (c) long-axis inclined to carrier contacted surface (black arrow is noted for long-axes of alumina particles).

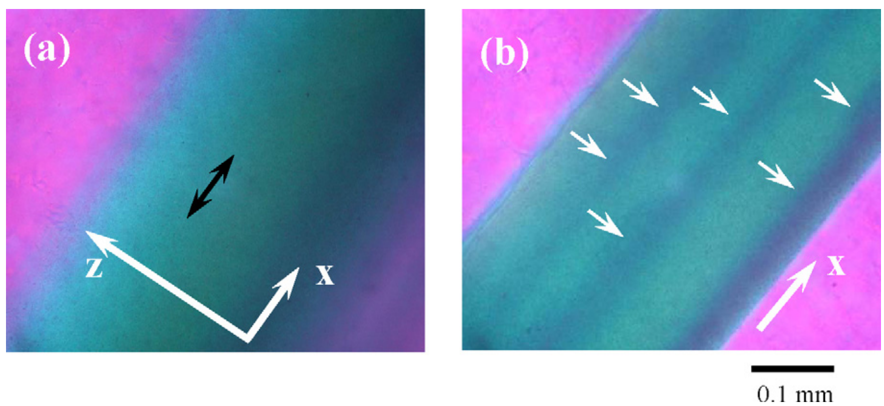

Fig. 4. Cross polarized light image of Type-II structure taken on the x–z plane. Photographs were taken at 45° from maximum extinction. (a) uniform long-axes alignment across tape thickness, blue color represent for long-axes aligned in x-axis, (b) non-uniform long-axes alignment across tape thickness, non-uniform region are indicated by white arrows.

thickness. Thus, Type-II structure represents a uniform particle packing structure consisting of some localized non-uniform structure.

Fig. 5 shows the warping characteristic of sintered specimens at various temperatures from 1200 to 1600 °C. The photographs were taken from the x–z plane of cast tape specimen. There is the obvious warping pattern of Type-I structure. All specimens (Fig. 5(a)) showed the same warping direction which was transverse to the tape casting direction and toward the top surface. The specimens which were sintered at lower temperature (1200 °C) showed less warping height compared to others. It was evidently a slightly distorted specimen at 1600 °C. Type-II structure, in contrast, sintered specimens presented the irregular distorted shape (Fig. 5(b)). A relatively flat shape was noted only when sintered at 1300 °C. No distortion trend could be noted for the warping distortion of Type-II structure specimens.

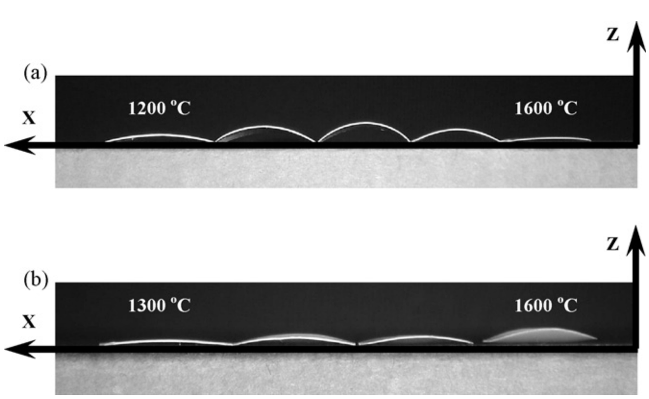

Fig. 5. Distortion characteristic of the round-shape specimen (x–z plane view) at various temperature. (a) sintering deformation of Type-I structure specimen; (b) sintering deformation of Type-II structure specimen.

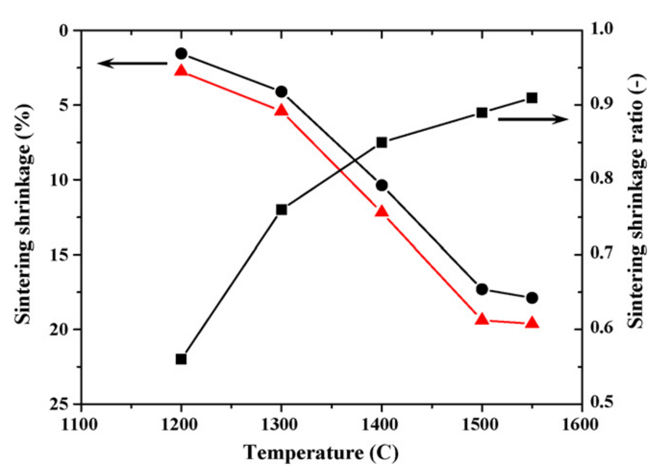

Fig. 6. The change of sintering shrinkage (measured in x-direction) with temperature for Type-I structure specimens (●: top surface; ▲: bottom surface).

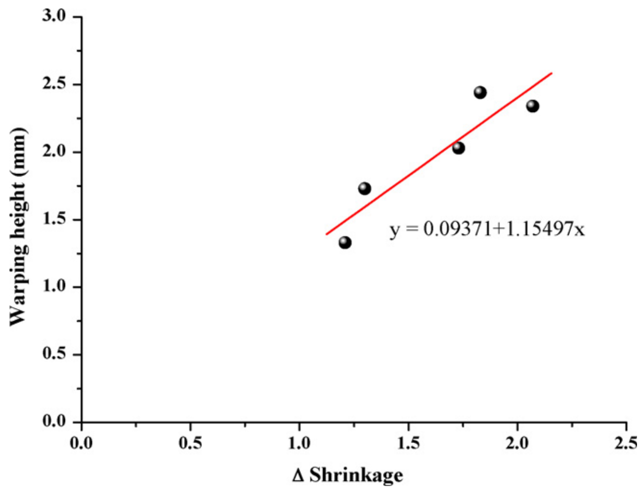

Fig. 7. The change of warping height with shrinkage difference between top and bottom surface of Type-I structure specimens.

Fig. 6 shows the change of sintering shrinkage measured in the casting direction plotted with temperature. The shrinkage that occurred in the plane of the top surface is less than that occurring in the plane of the bottom surface for all temperatures. This observed anisotropic shrinkage is attributed to the difference of particle packing structure between top and bottom surface (Type-I structure). The sintering shrinkage ratio which is defined as the ratio of shrinkage calculated from top surface relative to that of bottom surface is also included in Fig. 6. The anisotropic shrinkage between surfaces is very obvious at the beginning of the sintering (1200 °C) where the sintering shrinkage ratio is of 0.56 and it becomes less anisotropic as sintering temperature increases to 1550 °C where the ratio is of 0.91.

Fig. 7 shows the change of warping height with shrinkage difference between top and bottom surface of Type-I specimens. The warping height correlates with the difference in shrinkage between top and bottom surface as the linear relationship. This distortion height is nearly zero where the shrinkage difference becomes infinitesimally small.

4. Discussion

The sensitive examination tool used in this study can provide details of packing structure of powder particles. By means of long-axes alignment across the tape thickness, two types of particle packing structure are confirmed and referred to as Type-I and Type-II structure. After heat treatment, particular distortion obviously existed in tape specimens with Type-I structure. In this case, specimens from various sintering temperatures showed warping which curved toward the air-side surface. This warping pattern arises from tape structure, as the top surface of tape (air-side) consisted of aligned long-axes in the casting direction, while the bottom surface (carrier-side) composed of deviated long-axes from the casting direction. The presence of warping direction is generated because of the shrinkage difference between surfaces (Fig. 7), i.e. bottom surface induced higher linear shrinkage than top surface (Fig. 6). This result of alignment-driven shrinkage agrees to either the simulation of sintering of oriented ellipses which suggested higher shrinkage in the direction perpendicular to the long-axes of elliptical particle[10] or the experimentally determined origin of anisotropic sintering shrinkage which was ascribed to the particle orientation.[11–13] In cases of Type-II structure, instead of no distortion, the sintered specimens apparently showed irregular distorted shape for all sintering temperatures. A possible reason for this could be the randomly remaining localized non-uniform structure. Thus, no occurrence of warping and unpredictable distorted shape found in these sintered specimens (Fig. 5(b)).

In summary, particle packing structure, in terms of direction of alignment within the consolidated green tape, significantly induced deformation in sintered specimens. In this study, the relevance of particle alignment structure on deformation pattern is understandable.

5. Conclusions

Experiment showed the deformation of sintered tape specimens is influenced by particle alignment. By means of non-uniform aligned long-axes between the upper and lower surfaces of alumina tape, the pattern of particle alignment significantly induced warping transverse to the casting direction. Moreover, the consolidated green tape with basically uniform aligned long-axes in the casting direction showed irregular distorted shape specimens which are governed by randomly localized non-uniform structure.

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