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Observation of Settling Behavior of Particles in Slurry under Centrifugal Force

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Abstract

Powder-using processing such as ceramic processing and powder metallurgy is one of the powerful tools to provide advanced materials or parts difficult to fabricate by other processing routes. One of key technologies for powder-using process lies on compacting method, since packing microstructure determines performance and reliability of finished products. Settling powders under high centrifugal force can provide dense, homogeneous and defect-free powder compacts. In the present paper, we observe settling behavior of particles in slurry under centrifugal force, varying initial condition of slurries. The settling phenomenon is basically depicted by Kynch plot, showing that the compacting speed under centrifugal force is basically insensitive not only to initial concentration but also to viscosity of the slurries. This is great advantage for practical application of the HCP to powder-using processing.

1. Introduction

Ceramic processing has casual image of traditional processing method for producing table wares, tiles etc., but actually it is a powerful tool to create new materials and provide key parts for advanced products. In ceramic processing, starting materials are prepared as powders, then, densely compacted into desired shape, and heated up to just below the melting temperature (sintering) to get strong bodies. That is, there are no needs of melting, deforming or cutting of materials throughout the process. Therefore, it is suitable to process hard and brittle materials, namely, difficult-to-processing materials into complicated shapes.

Same kind of powder-using process is also applied to metallic powders, called powder metallurgy (P/M). The powder-using process spans not only mechanical products but also electrical and chemical devices. Their importance is still increasing since the development of products is lead by, at least partly, the development of materials performance. That is, more difficult-to-processing materials are required for advanced products.

There is a general weak point, however, in powder-using process. The products by powder-using process tend to contain defects in the microstructures. Just one remaining 10 μm pore may spoil the whole properties of the product. Such defects are introduced, in many cases, during powder compaction. Tiny powders show somehow sticky nature, loosing fluidity for dense and homogeneous packing into powder compacts.

In such circumstance, an idea of utilizing centrifugal force for compaction of fine powders was emerged in the late 80th. Around the same time, our laboratory also had started to develop centrifugal compaction[1)]. We named the process "High-speed Centrifugal Compaction Process, HCP" since we revealed that the use of high centrifugal acceleration of about 10,000 was the key for providing dense, homogeneous, and defect-free compacts of tiny, namely, micron to nano powders[2,3)]. The compacts processed by the HCP were easy to be sintered, resulted in strong products[4-7)].

Through the research on the HCP, we also found that the HCP had unique compacting mechanism different from other conventional ones[8-12)]. The aim of present paper is to analyze compacting mechanism of the HCP in detail, focusing especially on the effect of slurry concentration and viscosity.

2. High-speed Centrifugal Compaction Process

High-speed Centrifugal Compaction Process (HCP) is a variation of colloidal processing. Prior to the compaction, starting powders must be dispersed in dispersing media to form slurry. In slurry, friction between particles is dramatically reduced and they get higher fluidity. In centrifugal compaction, the slurries is poured in a die, set in a centrifuge, and rotated at certain speed for certain period of time (**Fig. 1**). During such process, the particles flow in the slurry downward to form densely packed sediment on the bottom of the die, and this sediment is a compact.

Brief summary of compacting mechanism of the HCP is as follows[8-11)]. **Figure 2** is a schematic of density profile of slurry during compaction. It consists of, from the top, fully transparent supernatant with powder concentration ϕ =0 (clear zone), intermediate flowing slurry ($\phi \fallingdotseq \phi$ initial, falling zone), and

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solid like sediment (compacted zone) at the bottom. Formation of these separate regions was previously studied by Kynch[13], and it was applicable to our previous case. He focused on the flux of the powder, namely amount powder passing through unit area in a unit time, rather than falling speed of particles in a slurry, and summarized it as a function of powder load (volumetric) of slurry (Kynch plot). **Figure 3** is empirically derived Kynch plot of our alumina slurry[8,10,11]. Kynch revealed that elevating speed of a boundary in a slurry was shown as gradient of Kynch plot at certain concentration. In our case, the slurry-sediment boundary was formed where powder load in slurry of about 50 vol%[9,10]. The calculated compacting speed from gradient of Kynch plot at ϕ =50% was good accordance to measured result[10]. We shall show the details of deduction of Kynch plot in the later part of this paper.

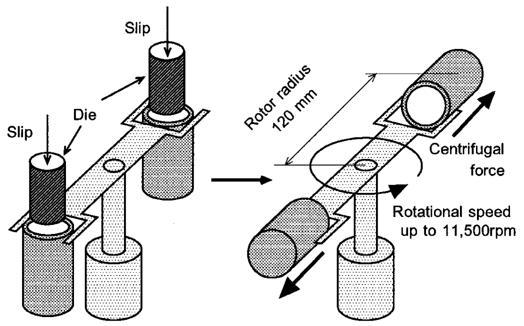


Fig. 1 Schematic illustration of the HCP

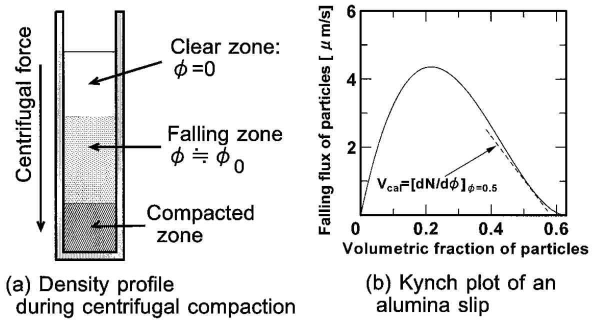


Fig. 2 Schematic illustration of density Profile during the HCP and falling flux of particles (Kynch plot)

3. Experimental Procedure

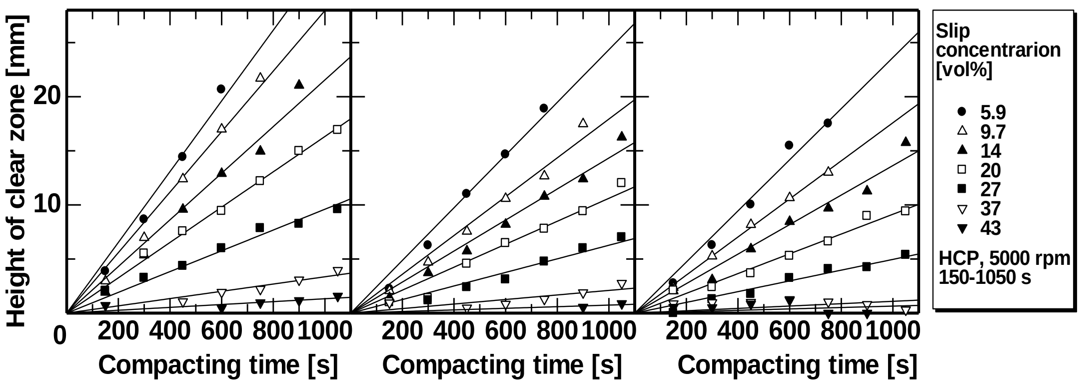
Alumina powder of 0.2 microns (TM-DAR, Taimei Chemical Co., Japan) is used. Powder is dispersed in ion-exchanged water with volumetric concentrations of 5.9－43 % and ball-milled for 48 hours. Dispersing agent of 0.6, 1.2, 1.8, 3.0 are added to vary the viscosity of the slurries. Since the agent of 0.6 % is appropriate concentration to obtain well dispersed state, the excessive addition of the agent makes viscosity of the slurry increase. Although the viscosity value is quite important factor, we don't measure it in this paper since it varies by several factors of concentration of slurries, share speed, etc. There may be thixotropic natures as well. As we will show in results and discussion, actual movement of particles in slurries (several tens μm/s) is much slower than share speed in viscosity measurement.

Prepared slurry is then compacted by the HCP. Slurry is poured into a couple of cylinder-shaped glass tubes, and they are loaded on the centrifuge with rotor radius of 120 mm, then rotated at 5,000 rpm for 150－1050 s. Then, the heights of clear and compacted zones are measured by height gauge.

4. Results

4.1 Falling Speed of Particles in Slurries

Figure 4 shows the height changes of clear zone during the



(a) agent of 0.6 mass% (b) agent of 1.2 mass% (c) agent of 3.0 mass%

Fig. 3 Width change of clear zone during the HCP, varying initial concentration of slurry and dispersing agent content.

HCP, using slurries of different initial concentrations and dispersing agent content. Formation speed of the clear zone is obviously affected by the slurry concentration, whereas the speed is almost unchanged by excess addition of agent (1.2, 3.0 %), namely, by viscosity increment.

The forming speed of clear zone (slope of each plots in **Fig. 4**) can be taken as falling speed of particles in the slurries of respective initial concentrations. Curiously enough, the particles in dilute slurry can fall more rapidly than thick ones.

The falling speeds of particles of different slurry conditions are summarized in **Fig. 5**. Regardless of the agent content, the falling speed decreases dramatically as the concentration increases. There is only slight decrease is remarked, on the other hand, with excess addition of the agent.

4.2 Compacting Speed

Compacted zone height change during the HCP is shown in **Fig. 5**. A clear contrast is remarked between formation speeds of clear and compacted zones. Formation speed of the clear zone (**Fig. 3**) is obviously affected by the slurry concentration, whereas that of the compacted zone is almost indifferent to slurry concentration. The same tendency is seen with viscous slurries. Compacting speed, namely, the slope of the plot, is slightly reduced as slurry viscosity increases.

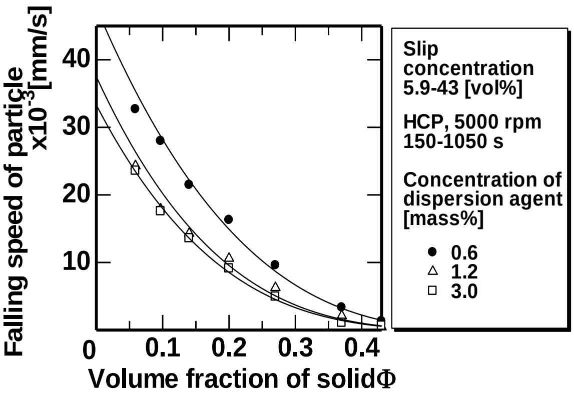


Fig. 4 Falling speed of particles in slurry during the HCP.

5. Discussion

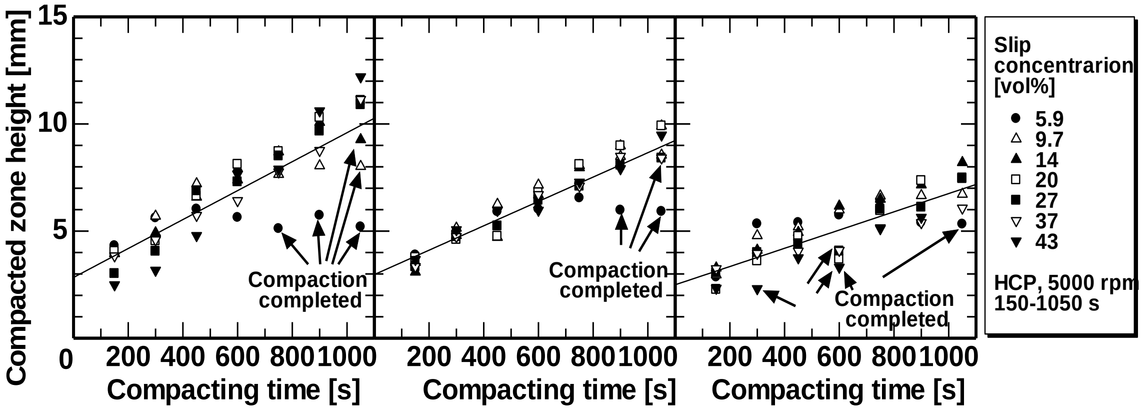
5.1 Falling Speed of Particles in Slurries

Higher falling speed of particles in dilute slurries than thick one (**Fig. 4**) is somehow contradictory to our intuitive image. This phenomenon can be explained by taking into account the counter flow of dispersing medium (**Fig. 6**). As the slurry concentration becomes higher, a larger amount of particles goes downward pushing aside the medium around the particles, which results in a faster counter flow (upward flow) of the medium. This upward flow of the medium reduces relative falling speed of particles from macroscopic and stationary view point. The relationship between falling speed of particles Vp and slurry concentration ϕ in this theory is expressed simply as follows.

$$Vp = V_0 (1 - \phi) \qquad (1)$$

V_0 is falling speed of particles in respect to the medium, which practically coincides with falling speed of particles in dilute slurry. This equation, however, only partly coincides with empirical data, since there are further factors affecting the Vp such as interference between particles, agglomerate formation, etc.

In previous studies, we found that the falling speed of particles under the HCP was well depicted by the Buscall's equation (2), which was a modification of above simple



(a) agent of 0.6 mass% (b) agent of 1.2 mass% (c) agent of 3.0 mass%

Fig.5 Height change of compacted zone during the HCP, varying initial concentration of slurry and dispersing agent content.

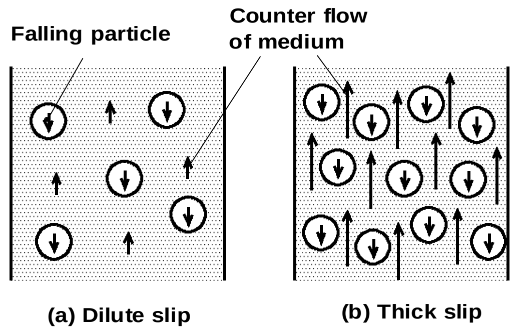


Fig. 6 Counter flows effect on apparent falling speed of particles from stationary view point.

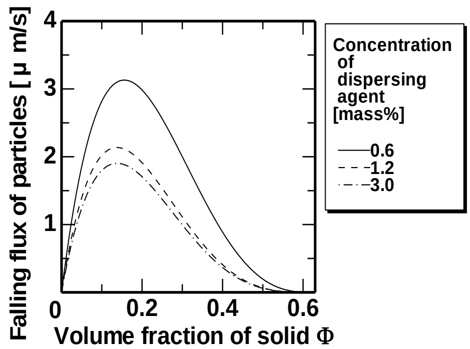


Fig. 7 Falling flux of particles in slurry during the HCP, varying dispersing agent content.

Table 1 Empirically derived constants k and V_0 in Buscall's equation (2), which describes falling speed of particles during the HCP.

Concentration of dispersion agent [mass%]	k	V_0 [×10^{-3}mm/s]
0.6	4.83	47.7
1.2	5.68	37.5
3.0	5.66	33.3

equation[14].

$$Vp = V_0 (1 - \phi / \phi_{max})^{\phi max \times k} \qquad (2)$$

Where ϕ max is the maximum packing density of particles, and k is the empirical constant. The ϕ max of this slurry is about 0.63, as shown in another study[15] .

Using the data in **Fig. 4**, the values of V_0 and k are calculated (**Table 1**). Actually, the curved lines superimposed in each plot in the **Fig. 4** are made from these data. They are well coincided. That is, falling speed of particles in viscous slurries (with excess dispersing agent) under the HCP also can be adopted the Buscall's equation.

5.2 Effect of Viscosity Increment on Falling Speed of Particles

The k and V_0 in the **Table 1** remark distinct trends. That is, the k values is slightly increased with dispersing agent content, whereas the V_0 is reduced as the concentration of dispersing agent increases. The increment of k suggests that the excessive agent may affect, in some manner, interference between particles.

The meaning of V_0 in equation (1) is that it indicates the falling speed of particles in very dilute slurries without counter flow. The reduction of V_0 with the agent suggests, therefore, that the excessive agent in the slurries hinders straightforwardly the falling movement of the particles in the slurries.

In spite that the excessive addition of agent remarks k and V_0 change in Buscall's equation, reduction of falling speed is not so large. A very slow movement of particles (several tens μm/s) may mask the viscosity effect which we see in macroscopic sense. In total, particle falling movement in slurries under the HCP is relatively insensitive to apparent viscosity of the slurry.

5.3 Conversion of Falling Speed of Particles into Flux of Particles

In the last section, we argued falling speed of particles in slurry. But the falling speed is not the critical factor which determines compacting speed. The critical factor is the flux N of the particles (Kynch's theory[13] that we introduced former part of this paper). The flux is defined as the volume of particles conveyed across a unit area in a unit time. Actually, it is deduced just multiplying Vp and volumetric concentration of slurry ϕ .

$$N = Vp \cdot \phi \qquad (3)$$

The three curves in **Fig. 4** are, then, converted to the figure of flux N vs. concentration (Kynch plot, **Fig. 7**). Mountain peaks at around the concentration ϕ of 0.2 indicate that a more amount of particles can be conveyed in dilute region. Therefore, dilute slurry is thickened in relatively short time, (which is observed macroscopically as quick reduction of falling zone depth in **Fig. 2**), keeping almost constant compacting speed regardless of initial concentration of slurries.

Note that the effect of excessive agent addition corresponds to decrease of mountain height in Kynch plot. From this view point, the excessive agent addition has definitely hindering effect on falling movement of the particles. Nonetheless, it is not direct relationship onto compacting speed as we shall see in the next section.

5.4 Predicting Compacting Speed by HCP from Kynch Plot

As we mentioned above, compacting speed of each slurry is calculated from the slope of the Kynch plot. Same as previous studies[9,10], we assume the concentration at the falling and compacted zones boundary as ϕ = 0.5. The calculated

Table 2 Comparison of compacting speeds calculated from Kynch plot and by measurement.

Concentration of dispersion agent [mass%]	Calculated forming velocity of collapse zone [×10^{-3}mm/s]	Measured forming velocity of collapse zone [×10^{-3}mm/s]
0.6	5.4	10.9
1.2	2.2	6.5
3.0	2.0	3.9

compacted speeds are compared with measured ones in **Table 2**. The calculated values are about a half of measurement data, but they show similar tendencies with measured ones. The result indicates, still they are roughly coincided, that the Kynch plot is also applicable to depict compacting phenomenon under the HCP with viscous slurry systems.

One more noteworthy result shown in the **Table 2** is that the degree of compacting speeds with viscous slurries. Although the dramatic viscosity increment is observed such agent rich slurries, the reduction of compacting speed from 0.6 % agent to 3.0% agent is just a half. That is, compacting phenomenon under the HCP is almost insensitive to viscosity of the slurries.

The argument thus far shows that the compacting phenomenon under the HCP is basically insensitive to not only initial slurry concentrations but also viscosity of the slurries. Such characteristics of the HCP become great advantages over other slurry-using compacting methods in practical use. Usually, compacting velocitity of conventional slurry casting is dramatically affected by initial concentration and viscosity of the slurries. And in many cases, dilute concentration and high viscosity reduce compacting speed in a great deal, and may degrade the packing microstructures of the compact.

6. Summary

Compacting phenomenon of particles in slurries under High-Speed Centrifugal Compaction Process (HCP) is observed varying initial concentration and viscosity of the slurries.

Falling speed of particles is strongly affected by initial concentration. Quicker speeds are observed in thinner slurries. In contrast, compacting speeds are almost constant regardless of initial concentration of the slurries. Viscosity increment has only slight effect on falling speeds and compacting speed. The compacting phenomenon under the HCP is basically insensitive to not only initial concentration and viscosity of the slurries. This is a great advantage for practical application of the HCP to powder-using processing.

The falling speed of particles in slurries of various initial conditions is well depicted by Buscall's equation, and the compacting speeds can be calculated from Kynch plot.

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