Coating of Seed Particles in Tumbling Fluidized Bed by Atomizing the Suspensions of Clayey Particles

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Abstract

Coating seed particles of 53 to 210 μm with clayey particles was carried out by means of a tumbling fluidized bed in order to enlarge and reuse the clayey particles wasted in producing a carrier for driftless dust formulation. The influence of operating factors on the coating efficiency η and on the growth rate of seed particles was investigated. η varied depending upon thermal operating factors but was not influenced much by mechanical ones. It was found that η was related roughly to $R_w$, defined as the ratio of the feed rate of water fed as slurry droplets to the maximum theoretical evaporation rate of water in a coating chamber. η increased with $R_w$. The relationship between η and $R_w$ varied with the additive ratio of binder PVA to clayey particles, $C_b$, and η became high with increasing $C_b$. The growth rate of seed particles was explained well by an equation derived on the basis of the mass balance of clayey particles in a coating chamber.

1. Introduction

Recently, the particle size of the dust among the various agricultural chemicals tends to be enlarged to reduce the public nuisances caused by the drift of fine particles generated on the spraying.

This agricultural chemicals called DL dust (particles size; 10 ~ 46 μm) contains few fine particles smaller than 10 μm. Therefore the fine particles smaller than 10 μm are generated as by-products in large quantities, when they produce carrier of DL dust, which leads to a possibility of causing problems in the disposition.

As one of the means of solving the problem, the authors have investigated the coating of the seed particles (about 100 μm) with the above-mentioned by-product particles using a tumbling fluidized bed coater1) to develop a new type carrier for the agricultural chemicals, F micro-granules (particle size 63 ~ 210 μm).

The tumbling fluidized bed coater has been devised as an apparatus that makes possible to coat particles less than 300 or 500 μm, which is a critical size for conventional fluidized bed coater. With regard to this apparatus, however, few studies have been reported so far and the instructions for optimum operating conditions to get desirable results have not been made clear yet at all.

This work elucidates the effect of the operating factors of the tumbling fluidized bed coater on the coating efficiency, indicates the instructions for the optimum operating conditions. The growth rate of seed particles is also discussed in this work.

2. Samples

As coating particles, Roseki (density; 2710

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† This report was originally printed in J. Soc. Powder Technology, Japan, 22, 278-287 (1985) in Japanese, before being translated into English with the permission of the editorial committee of the Soc. Powder Technology, Japan

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kg/m³, particle size; < 10 μm, mass median Stokes diameter; 3.6 μm) or Kunimine clay (density; 2710 kg/m³, particle size; < 20 μm, mass median Stokes diameter; 7 μm) and as seed particles, ground Toyoura sands (density; 2630 kg/m³, particle size; 53 ~ 210 μm) or calcite (density; 2700 kg/m³, particle size; 250 ~ 590 μm) were used in the experiments.

Roseki was a product from Kawatana factory of Goto Mining in Nagasaki pref. consisting mainly of quartz and agalmotalite. Kunimine clay was a product by Kunimine Kogyo Co., Ltd., which consisted mainly of quartz and feldspar and contained some clay minerals such as chlorite, sericite and talc.

3. Experimental apparatus and procedures

3.1 Tumbling fluidized bed coater

A schematic diagram of a tumbling fluidized bed coater (Okada Seiko Co., Ltd., type SP-25) is shown in Fig. 1.

The air from a blower through a heater and damper is introduced into the coating chamber (250 mm φ) through the gap (10 mm) between a rotating disk and the chamber wall. By the disc rotation, the tumbling seed particles in the chamber are forced to exist in the vicinity of the chamber wall, where they are fluidized by the hot air. The coating particles dispersed in water are sprayed to such violently moving seed particles from a nozzle (Fuso Seiki, Co., Ltd., type Rumina ST-5, external mixing type pneumatic nozzle, liquid outlet diameter; 1.0 mm φ) set above in the center of the disc. The spray of the slurry is directed to the point 9 cm away from the center on the disc face. The slurry droplets which adhered to the surface of the seed particle are dried by the hot air, so that the particles in the droplets coat the surface of the seed particle. A part of the coating particles which failed to adhere to the seed particles as a result of peeling etc. are discharged from the coating chamber and collected in a cyclone on the way to the air exhaust of this system.

The experimental procedures were as follows. The disc rotation speed and the flow rate of inlet air were adjusted after charging the coating chamber with a required amount of seed particles. When the temperature of the air reached a fixed value, the slurry was fed to the nozzle at a constant rate by a gear pump and began to spray under the constant air pressure (0.294 MPa). During the experiment, the air temperature after passing through the fluidized bed was measured with a thermocouple set near the nozzle. After a fixed coating time, the coated particles were taken out from the outlet, as soon as the feed of the slurry was stopped. The representative sample of 0.04 ~ 0.06 kg out of the product taken out from the chamber was sifted through the standard sieve (37 μm) after stirring in hot water (343 ~ 353 K, 400 cm³) for 4 hours. Having had confirmed that the particles of Roseki and the Kunimine clay passed the standard sieve (37 μm) entirely, the undersize was regarded as the coating particles. The mass of the undersize per total mass of the seed particles supplied to the coating chamber was adopted as the total mass of the coating particles adhering to the seed particles.

In this article, the coating efficiency \( \eta \) is defined as the ratio of the total mass of the coating particle adhering to the seed particles to the total mass of the coating particles fed into the coating chamber during the coating operation.

As for the operating factors, there are the mechanical operating factors such as the disc rotation speed \( N \) [s⁻¹] and the mass of seed particles \( M_s \) [kg] etc., and the thermal operating factors such as the feed rate of the coating particles \( F_c \) [kg/h], the slurry concentration (mass ratio of coating particles to slurry) \( s \) [kg/kg], the flow rate of inlet air \( Q \) [m³/h], the humidity of inlet air \( H \) [kg H₂O/kg dry air] and the temperature \( t_i \) [K] etc. The experiments were carried out under the wide range of conditions shown in Table 1 to make clear the
relationship between those operating factors and the coating efficiency. In the experiments, ground Toyoura sands and Roseki were used as the seed particles and as the coating particles respectively.

Additionally, PVA (Nippon Gosei Kagaku Kogyo Co., Ltd., Gohsenol GL-05S) was used as a binder to improve the coating efficiency. The influence of the additive ratio of binder $C_b$ on the coating efficiency was also examined.

The concentration of PVA in the water solution is 0.5 ~ 1.2 wt% under the condition of $C_b = 0.02$, $s = 0.2 \sim 0.37$, and 0.2 ~ 1.7 wt% in the case of $s = 0.3$, $C_b = 0.005 \sim 0.04$.

According to Yamada et al., Roseki slurry with PVA indicates the flow characteristic of thixotropy, newtonian or dilatancy depending upon the additive ratio of PVA and the viscosity increases with increasing slurry concentration.

Roseki slurry in this work showed dilatancy when $s = 0.3$, $C_b = 0.02$, and the apparent viscosity was $2.8 \times 10^{-1} \sim 4 \times 10^{-3}$ Pa·s at the shear rate of $81 \sim 401$ s$^{-1}$ and the temperature of 307 K.

In addition to the above-mentioned coating experiments, the coating experiments (using Kunimine clay as coating particles and calcite as seed particles) were carried out and compared with the case of coating ground Toyoura sands with Roseki, in order to examine whether the relation between the operating factors and coating efficiencies depends on the physical properties of the seed and coating particles or not. The coating time was 0.5 hr in all the experiments except the one conducted to examine the growth rate of the seed particles.

The nozzle height is another mechanical operating factor in addition to the disc rotation speed and the mass of the seed particles. Harada et al. have made clear already that the coating efficiency of a conventional fluidized bed coater is decreased as the nozzle height increases. Therefore, it becomes a problem how near the distance between the nozzle and the disc face can be fixed for the troubleless operation.

In preliminary experiments under the condition of $M_s = 0.6$ kg and $N = 1.5$ s$^{-1}$ when the nozzle height varied from 22, 16, 13 down to 10 cm, the seed particles on the disc face were eliminated by the air flow from the nozzle and the coating particles adhered to the disc face directly in case of the nozzle height less than 13 cm. On the other hand, no difference in the coating efficiency between the nozzle heights 16 and 22 cm was noticed. Therefore, all the following experiments were conducted with the nozzle height of 16 cm or 22 cm.

### 3.2 Measurement of the porosity of the coating layer

To consider the growth rate of the seed particles, the value of the porosity of the coating layer on the surface of particles must be known. The porosity was measured with two methods, a method of measuring the packed volume of the seed particle before and after the coating and the mercury porosimetry.5)

Assuming that the particles have the same shape before and after the coating and that the porosity $\epsilon_s$ in case of packing non-coated particles equals the one of interstices of particles in the case of packing coated particles, the porosity of the coating layer $\epsilon$ is given by,

$$\epsilon = 1 - \frac{m_e/\rho_e}{V(1-\epsilon_s) - m_s/\rho_s}$$

where $m_s$ [kg] is the mass of the seed particles packed in a vessel, $m_e$ [kg] is the mass of the particles coating the seed particles having the mass of $m_s$, $V$ [m$^3$] is the packed volume of the particles having the mass of $m_s + m_e$, $\rho_e$, $\rho_s$ [kg/m$^3$] is the density of the coating particles and the seed particles respectively. $V$ was measured by tapping a vessel of 28 mm diameter under the condition of the stroke 1 cm.
and counts of tapping 200.

On the other hand, POROSIMETO SERIES 1500 (CALRO ERBA STRUMENTAZIONE) was used as the instrument of the mercury porosimetry.

3.3 Measurement of volume shape factor of the seed particles before and after coating

To consider the growth rate of the seed particles, the value of volume shape factor is required in Eq. (11) that will be described later in this article.

On the basis of Eq. (2), the volume shape factor \( \phi_v \) was obtained by counting the number \( n_p \) of the particles having the size \( D \) and the mass \( M_p \) enlarged with a projector.

\[
\phi_v = \frac{M_p}{n_pD^3\rho_p}
\]

where \( D \) is the arithmetic mean of sieve aperture, \( \rho_p \) is the average particle density calculated from Eq. (3) using the porosity \( \epsilon \) of the coating layer.

\[
\rho_p = \frac{m_g + m_c}{m_g/\rho_g + m_c/(1 - \epsilon)\rho_c}
\]

4. Experimental results and Discussions

4.1 Droplets size

The results of the photograpical size analysis of more than 1000 droplets collected by the immersion liquid method \(^6\) are shown in Fig. 2. 

It's seen that the droplets having the mass median diameter of 17 ~ 28 \( \mu \)m are formed under the following conditions; the additive ratio of binder \( C_b = 0.02 \), the slurry concentration \( s = 0.3 \), the ratio of mass feed rate of slurry to that of air from the nozzle 0.35 ~ 1.0.

The droplet size distributions obeyed log-normal distributions as reported by Yamada et al.\(^2\), and the geometrical standard deviations were about 1.5. Under the operating conditions shown in Table 1, the ratio of mass feed rate of slurry to that of air from the nozzle was within the range of 0.33 to 0.69.

4.2 Influence of thermal operating factors on the coating efficiency

(1) Evaporation of water in fluidized bed

Some examples of the relationship between the thermal operating factors and the coating efficiencies are shown in Fig. 3.

As seen from the figure, the coating efficiency \( \eta \) increases as the feed rate of water \( W_E \) increases when other conditions are kept constant. Conversely, \( \eta \) decreases with increasing the difference between the adiabatic saturation humidity and the humidity \( H_s - H \), the temperature \( t_i \), or the volume flow rate of the inlet air \( Q \). These experimental results lead to the following consideration.

In Fig. 3 (a), the evaporation rate of water in the fluidized bed must have a maximum value because the operating factors except \( W_E \) are kept constant. If only \( W_E \) increases under the condition, a certain amount of water exceeding the evaporation rate is accumulated in the fluidized bed.

Similarly in Figs. 3 (b), (c) and (d) the decrease of \( H_s - H \), \( t_i \) or \( Q \) makes the accumulation rate of water increase, if the other factors are constant. Above-mentioned considerations suggest that the coating efficiency \( \eta \) might be determined by the accumulation rate of water in the fluidized bed and \( \eta \) becomes higher as this rate increases. The evaporation of water in the tumbling fluidized bed will be discussed in the following.

If the tumbling fluidized bed coater is regarded as an adiabatic humidifier in terms of the inlet air, the variation of air temperature and humidity through the apparatus ought to be illustrated as Fig. 4.

The air of temperature \( t_i \) and humidity \( H \) from the blower is heated by the heater until the temperature reaches \( t_i \). This heated air
gives the latent heat of vaporization to the slurry droplets in the fluidized bed. As a result, the temperature of the air decreases along the adiabatic cooling line through the point \((t_1, H)\) and the humidity increases.

If the contact time between the air and the seed particles is assured long enough, the maximum theoretical evaporation rate of water in the coating chamber \(W_T\) [kg H₂O/h] is expressed by

\[
W_T = W_G (H_s - H) = \left( \frac{Q}{V} \right) \times (H_s - H)
\]

where

- \(W_G\) [kg dry air/h]; mass flow rate of inlet air
- \(H\) [kg H₂O/kg dry air]; humidity of inlet air

**Fig. 3** Relationship between coating efficiency and operating factors
Fig. 4 Temperature and humidity variation of air through a coating apparatus

\( H_s \) [kg H\(_2\)O/kg dry air];
adiabatic saturation humidity of inlet air
\( v \) [m\(^3\)/kg dry air];
humid volume of the inlet air.

On the other hand, the feed rate of water \( W_E \) [kg H\(_2\)O/h] is given by

\[ W_E = F_c \left( \frac{1 - s}{s} - C_b \right) \]  

(5)

where

\( F_c \) [kg/h]; feed rate of coating particles
\( s \) [kg/kg]; slurry concentration
\( C_b \) [kg PVA/kg solid]; additive ratio of PVA

It is presumable that the air temperature becomes the adiabatic saturation temperature \( t_s \) in case of \( W_E > W_T \), while it reaches the temperature \( t' \) in case of \( W_E < W_T \), where \( t' \) is the temperature at the humidity of \( H + W_E/W_D \) on the adiabatic cooling line of the inlet air.

Figure 5 shows the comparison of the measured values of the air temperature in the fluidized bed obtained from all the coating experiments in this work with the theoretical values from a humidity chart on the basis of the above-mentioned procedure. The difference between the measured values and the theoretical ones is at most within \( \pm 2 \) K as seen from the figure so that it is considered that adiabatic humidification takes place actually in the tumbling fluidized bed coater.

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(2) Effects of moisture on the coating efficiency

As is already mentioned, adiabatic humidification takes place practically in the tumbling fluidized bed coater and the coating efficiency tends to become higher as the accumulation rate of water in the fluidized bed increases.

Since the accumulation rate of water in the fluidized bed is determined by the synthetic effect of the thermal operating factors, we use \( R_w \) defined by Eq. (6) as an index to evaluate the synthetic effect of the thermal operating factors and examine whether the coating efficiency can be definitely related to \( R_w \).

\[ R_w = \frac{W_E}{W_T} \]  

(6)

Figure 6 shows the relation between the coating efficiency \( \eta \) and \( R_w \) under the condition of \( C_b = 0.02 \), \( N = 1.5 \) s\(^{-1}\), and \( M_s = 0.6 \sim 0.69 \) kg.

The figure indicates that as far as the thermal operating factors vary in the range shown in Table 1, \( \eta \) is strongly correlated with the index \( R_w \). The relationship between \( \eta \) and \( R_w \) at slurry concentration of \( s = 0.5 \), however, shifted to high efficiency in the figure. The reason has not been clear and is the subject to be investigated in the future. Therefore, it is not to affirm that \( \eta \) should be determined by \( R_w \).
4.3 Influence of mechanical operating factors on the coating efficiency

Figure 7 shows the relationship between $\eta$ and $R_w$ in the case of the disc rotation speed $N = 2.1 \text{ s}^{-1}$ and $3.3 \text{ s}^{-1}$ under the condition of the slurry concentration $s = 0.3$, the additive ratio of PVA $C_b = 0.02$, the mass of the seed particles $M_s = 0.6 \sim 0.63 \text{ kg}$.

The broken line in the figure indicates the relationship between $\eta$ and $R_w$ in case of $N = 1.5 \text{ s}^{-1}$ shown in Fig. 6. Though a slight difference is noticed between the experimental results in case of $N = 2.1 \text{ s}^{-1}$, $3.3 \text{ s}^{-1}$ and $N = 1.5 \text{ s}^{-1}$ in the figure, the coating efficiency is hardly influenced by the disc rotation speed as far as it varies from $1.5 \text{ s}^{-1}$ to $3.3 \text{ s}^{-1}$.

The relation between the mass of the seed particles $M_s$ and coating efficiency $\eta$ is shown in Fig. 8. The figure makes clear that $\eta$ scarcely varies with $M_s$. Therefore, the mass of the coating particles per unit mass of the seed particles $M_c/M_s$ naturally decreases in inverse proportion to the mass of the seed particles as seen in the figure, when the feed rate of the coating particles is kept constant.

4.4 Influence of the additive ratio of binder on the coating efficiency

Figure 9 shows the relation between $\eta$ and $R_w$ obtained under the condition of the additive ratio of binder $C_b$ ranging from 0 to 0.04. The operating conditions of other factors are shown in Table 1. In the figure, the data under the condition $C_b = 0.02$ are the ones already shown in Figs. 6, 7 and 8.

As clearly seen from the figure, the coating efficiency $\eta$ is correlated well with $R_w$ by in-
Introducing $C_b$ as a parameter and $\eta$ increases with increasing $C_b$ at the same value of $R_w$. $\eta$ without addition of binder is so low that the addition of binder like PVA etc. is indispensable to carry out the coating with fine particles.

Although $\eta$ increases with increasing $R_w$, there exist critical values of $R_w$ depending upon $C_b$, beyond which particles are inclined to agglomerate and it is no longer possible to continue coating operation.

The solid line in Fig. 9 indicates the critical values determined roughly by the observation. The critical value of $R_w$ varies slightly with $C_b$, and it gets larger a little with increasing $C_b$ in the range $C_b < 0.02$. In the case of $C_b = 0.04$, however, 30 per cent of the whole particles become agglomerates having the size of about 3.0 mm at $R_w = 1.25$ so that the critical value of $R_w$ is rather smaller than that in the case of $C_b = 0.02$.

As is clear from the figure, the coating efficiency of 90 per cent is able to be achieved in the case of coating Toyoura sands with Roseki under the operating conditions of $C_b = 0.02$ and $R_w \approx 1.2$.

The reason why the coating efficiency $\eta$ becomes high with increasing $R_w$ and a parameter $C_b$ will be discussed in the following.

The coating operation in the tumbling fluidized bed coater may be regarded as a process where the sprayed slurry droplets being dried by the hot air collide with and adhere to the seed particles being tumbled or fluidized and the coating particles in the droplets coat the surface of the seed particles by further drying.

Regarding the slurry droplets as water droplets to simplify the phenomena, the water droplets need longer time with increasing $R_w$ for disappearance by the evaporation in the range of $R_w < 1$. On the other hand, when $R_w > 1$, only a fraction of $W_e$ in the fed water is evaporated and the water droplets equivalent to $W_e - W_t$ still remain. Moreover the higher $R_w$ is, the larger are the diameters of the remained droplets. Therefore the probability of the collision between the droplets and the seed particles which would dominate the coating efficiency becomes higher with increasing $R_w$, whether $R_w$ is smaller than 1 or not. Consequently, the coating efficiency will become higher with increasing $R_w$.

According to the above-mentioned considerations, the increase of $C_b$ is surmised to cause more droplets to collide with and to adhere to the seed particles and to give rise to the higher coating efficiency in consequence, because the drying rate of the droplets of PVA solution is small in comparison with that of the water alone.

4. Variation of the coating efficiency with the difference in physical properties of the seed particles or the coating particles

Figure 10 shows the experimental results in the case of coating calcite having the size of 297 ~ 590 $\mu$m or ground Toyoura sands as the seed particle with Roseki and the case of coating calcite with Roseki or Kunimine clay. The experimental conditions were $s = 0.5$ and $C_b = 0.02$ in the both cases.

The coating efficiency $\eta$ is correlated with $R_w$ as well as in the case of Fig. 9.

If the seed particles differ in material such as calcite or Toyoura sands as well as in particle size, the relation $\eta$ vs $R_w$ is unvaried as far as the coating particles are the same. Conversely, the relation $\eta$ vs $R_w$ is different obviously depending upon the physical properties of
coating particles if the same seed particles are used. For example, the coating efficiency of Kunimine clay is lower than that of Roseki at the same value of $R_w$.

This seems to be due to the fact the tensile strength of the wet powder bed reduces as the particle size increases. It would be also attributed to the trend that the sprayed droplets of larger solid particles contain less fraction of liquid as confirmed by Yamada et al. In this way, the particles of Kunimine clay are less adhesive to the seed particles than Roseki and are more likely to come off from them even having adhered, since the median diameter of the former is twice as large as that of the latter.

To examine the coating efficiency more minutely and quantitatively, hereafter it is necessary to analyze the drying process of the slurry droplets in the tumbling fluidized bed and that in the coating layer of fine particles, particularly the case with addition of high molecular binder such as PVA.

4.6 Growth rate of the seed particles

Figure 11 shows an example of the variation of the size distribution of the seed particles with time. The reason for the slight differences in the values of $\eta$ and $R_w$ at each coating time noticed in the figure is attributed to the fact that the experiments were carried out individually in case of $\theta = 0.5, 1.0$ and $2.0$. The figure clearly shows the steady growth of the seed particles with coating time.

As the relationships between $\eta$ and $R_w$ at each coating time agree well with the result obtained under the condition of $s = 0.3$ and $C_b = 0.02$ in Fig. 9, it is obvious that the coating efficiency is not affected by the coating time.

Harada et al. derived the growth rate equation of uniform sized spherical seed particles in the case of coating with an eductional solution (sucrose) in a conventional fluidized bed.

In the present work, the authors deal with the coating of the seed particles having size distribution with fine particles. Then the variation of the mean diameter of the seed particles with time in the coating with fine particles is discussed below.

From the material balance of the coating particles, the following equation is obtained

$$\sum_{0}^{\infty} n_f(D, \theta) \phi_s D^3 dD - \sum_{0}^{\infty} n_f(D, \theta) \phi_{s0} D^3 dD
$$

$$= \frac{M_e/(1 - e) \rho_c}{M_s/\rho_s}$$

where

- $n_f(D, \theta)$: total number of seed particles
- $f_s(D, \theta)$: frequency number size distribution of seed particles at $\theta = 0$
\( f(D, \theta) \); frequency number size distribution of seed particles at \( \theta = \theta \)

\( \phi_v \); volume shape factor of particles of size \( D \) at \( \theta = \theta \)

\( \phi_{\text{vo}} \); volume shape factor of particles of size \( D \) at \( \theta = 0 \)

Substituting \( M_c = \eta F_c \theta \) into Eq. (7) yields,

\[
\int_0^\infty f(D, \theta)\phi_v D^3 dD \left[ \frac{\eta F_c \theta \rho_s}{M_s(1 - \epsilon) \rho_c} \right] - 1 = \frac{\eta F_c \theta \rho_s}{M_s(1 - \epsilon) \rho_c} \int_0^\infty f_0(D, \theta)\phi_{\text{vo}} D^3 dD (8)
\]

When the volume mean diameters of the seed particles at \( \theta = \theta, \theta = 0 \) and the average volume shape factors at \( \theta = \theta, \theta = 0 \) are denoted by \( D, D_0 \) and \( \phi_v, \phi_{\text{vo}} \) respectively, the numerator and denominator of the left side of Eq. (8) are rewritten based on the definition of the volume mean diameter as follows.

\[
\int_0^\infty f(D, \theta)\phi_v D^3 dD = \phi_v D^3 \quad (9)
\]

\[
\int_0^\infty f_0(D, \theta)\phi_{\text{vo}} D^3 dD = \phi_{\text{vo}} D_0^3 \quad (10)
\]

Substituting Eqs. (9), (10) into Eq. (8), yields the growth rate equation of the seed particles in the coating process with fine particles.

\[
\left( \frac{D}{D_0} \right)^2 = \left[ \frac{\eta F_c \theta \rho_s}{M_s(1 - \epsilon) \rho_c} + 1 \right] \frac{\phi_{\text{vo}}}{\phi_v} \quad (11)
\]

In Eq. (11), \( \epsilon \) equals 0 for the coating with eductional solutions, and \( \phi_v/\phi_{\text{vo}} \) equals 1.0, unless the shape of the particles varies during the coating. In this case, Eq. (11) is reduced to the equation proposed by Harada et al.

Experimental values of the volume mean diameter at each coating time are compared with the calculated ones from Eq. (11) in the following. To make use of the equation, the volume shape factor and porosity of the coating layer must be known.

**Table 2** shows the measured values of the volume shape factor of the particles sifted in the ranges of 125 \( \sim \) 149 \( \mu \)m and 177 \( \sim \) 210 \( \mu \)m. It is clear from the table that though there exists a difference in the volume shape factors between the two groups, they increase with increasing \( \theta \), which means the particles get spherical gradually with time. This tendency was also confirmed visually on a projector.

| \( \theta \) | 0 | 0.5 | 1 | 2 |
|---|---|---|---|---|
| 125 \( \sim \) 149 \( \mu \)m | 0.68 | 0.70 | 0.81 | 0.92 |
| 177 \( \sim \) 210 \( \mu \)m | 0.63 | 0.68 | 0.72 | 0.76 |

**Table 3** shows the measured values of the porosity of coating layer \( \epsilon \) by the packing method. The value of the porosity of the non-coated seed particles \( \epsilon_s \) was 0.37. The measured values of \( \epsilon \) were 0.55 regardless of the coating ratio \( m_c/m_s \).

The relation between the pore radius and the specific pore volume measured by the mercury porosimetry is shown in Fig. 12.

The pore volume consists of both the volume of interstices of coated particles and the pore volume of coating layer, as generally seen on the penetration of mercury into a layer of fine granules. The median of the pore radius of coating layer is about 0.4 \( \mu \)m. The specific pore volume of the coating layer gives the porosity of 0.54, which is nearly equal to the value obtained by the packing method.

The assumption at the time when Eq. (1) was derived is not valid in the strict sense because the particle shape varies with coating time as shown in **Table 2**. However, the values of the coating ratio demonstrated in **Table 3** are not so large that the difference of the volume shape factor before and after the coating is small. Hence, it appears that the values of porosity obtained by both methods were almost the same.

**Table 4** shows the experimental values of

| \( m_c \times 10^3 \) | \( m_s \times 10^3 \) | \( m_c/m_s \) | \( V \times 10^6 \) | \( \epsilon \) |
|---|---|---|---|---|
| 49.8 | 0 | 0 | 30.2 | 0.37 (=\( \epsilon_s \)) |
| 40.1 | 5.63 | 0.14 | 31.5 | 0.55 |
| 35.9 | 6.84 | 0.19 | 30.5 | 0.55 |
| 32.4 | 8.03 | 0.25 | 30.0 | 0.55 |

**Table 4** Comparison of experimental values of volume mean diameter of coated particles with calculated ones

| \( \theta \) | 0 | 0.5 | 1 | 2 |
|---|---|---|---|---|
| \( m_c/m_s \) | 0 | 0.29 | 0.64 | 1.46 |
| \( D \times 10^6 \) | 103 | 118 | 126 | 146 |
| \( D_{\text{cat}} \times 10^6 \) | – | 119 | 130 | 152 |
| \( D_{\text{cat}} \times 10^6 \) | – | 121 | 137 | 165 |

Table 2 Measured values of volume shape factor

Table 3 Measured values of porosity of coating layer

Table 4 Comparison of experimental values of volume mean diameter of coated particles with calculated ones
the volume mean diameter $\bar{D}$ at each coating time and the calculated values $\bar{D}_{cal}$, $\bar{D}_{cal}'$ obtained from Eq. (11) regarding as $\varepsilon = 0.54$, the variation of the particle shape being taken into account and not respectively.

$\bar{D}$ was calculated from Eq. (12) on the basis of the particle size distribution shown in Fig. 11, assuming the volume shape factor was independent of particle size.

\begin{equation}
\bar{D} = \left[ \frac{\sum w}{\sum (w/D^4)} \right]^{\frac{1}{3}}
\end{equation}

In calculation of Eq. (11), the arithmetic mean values of volume shape factor of two groups shown in Table 2 were used for $\bar{\phi}_e$, $\bar{\phi}_ro$.

As seen from the table, $\bar{D}_{cal}$ calculated taking the variation of shape factor with time into account agrees better with the measured values than $\bar{D}_{cal}'$ obtained assuming $\bar{\phi}_ro/\bar{\phi}_e = 1$.

5. Conclusions

Coating of seed particles in the tumbling fluidized bed was carried out by atomizing the suspensions of clayey particles. Investigating the influence of operating factors on the coating efficiency and the growth of seed particles, the following conclusions have been obtained.

(1) The coating efficiency hardly depends on the mechanical operating factors such as the mass of the seed particles, the disk rotation speed and the coating time.

(2) Introducing the additive ratio of binder, $C_b$, as a parameter, the coating efficiency $\eta$ is roughly related to and increased with $R_w$, which is an index to evaluate the synthetic effect of the thermal operating factors and the ratio of the feed rate of water as slurry droplets to the maximum theoretical evaporation rate of water in the tumbling fluidized bed coater.

$\eta$ becomes high with increasing $C_b$ under the same value of $R_w$. There exist critical values of $R_w$ depending upon $C_b$ beyond which it is no longer possible to continue the coating operation.

The instructions for the optimum operating condition of the tumbling fluidized bed coater can be obtained according to the relationship between $\eta$ and $R_w$.

(3) The growth of the seed particles can be known as the variation of the volume mean diameter with coating time calculated from the growth rate equation proposed in this article on the basis of the material balance of the coating particles.
Nomenclature

\[ C_b \] : additive ratio of binder to coating particle [kg/kg]
\[ D \] : particle size [m]
\[ D_0 \] : volume mean diameter of seed particle at \( \theta = 0 \) [m]
\[ D_{cal} \] : \( D \) calculated from Eq. (11) taking account of volume shape factor [m]
\[ D_{cal}' \] : \( D \) calculated from Eq. (11) regarding as \( \bar{\phi}_{e_0}/\bar{\phi}_p = 1 \) [m]
\[ F_e \] : feed rate of coating particle [kg/h]
\[ f(D, \theta) \] : frequency number size distribution of seed particle at \( \theta = \theta \) [-]
\[ f_0(D, \theta) \] : frequency number size distribution of seed particle at \( \theta = 0 \) [-]
\[ H \] : humidity of inlet air [kg H\(_2\)O/kg dry air]
\[ H_s \] : adiabatic saturation humidity of inlet air [kg H\(_2\)O/kg dry air]
\[ H' \] : humidity of outlet air [kg H\(_2\)O/kg dry air]
\[ M_c \] : total mass of coating particle adhering to seed particle of mass \( M_s \) [kg]
\[ M_s \] : total mass of seed particle in coating chamber [kg]
\[ M_p \] : mass of coated particle of size \( D \) and number \( n_p \) [kg]
\[ m_c \] : mass of coating particle adhering to seed particle of mass \( m_s \) [kg]
\[ m_s \] : mass of seed particle [kg]
\[ N \] : rotation speed of disk \([s^{-1}]\)
\[ n \] : total number of seed particle in coating chamber [-]
\[ n_p \] : number of particle of size \( D \) and mass \( M_p \) [-]
\[ Q \] : volume flow rate of inlet air \([m^3/h]\)
\[ R_w \] : index defined by \( W_E/W_T \) [-]
\[ s \] : slurry concentration [kg/kg]
\[ t_i \] : temperature of inlet air [K]
\[ t_r \] : room temperature [K]
\[ t_s \] : adiabatic saturation temperature [K]
\[ t \] : temperature at \( H' \) on adiabatic cooling line [K]
\[ V \] : packing volume of particle \([m^3]\)
\[ \nu \] : humid volume of particle on adiabatic [K]
\[ W_E \] : feed rate of water as slurry droplet [kg/h]
\[ W_O \] : mass flow rate of inlet air [kg/h]
\[ W_T \] : maximum theoretical evaporation rate of water in coating chamber [kg/h]
\[ w \] : mass fraction of particle of size \( D \) [-]
\[ \epsilon \] : porosity of coating layer [-]
\[ \epsilon_x \] : porosity of packed bed of seed particle [-]
\[ \eta \] : coating efficiency \((= M_c/F_e \theta)\) [-]
\[ \theta \] : coating time [h]
\[ \rho_c \] : density of coating particle [kg/m\(^3\)]
\[ \rho_s \] : density of seed particle [kg/m\(^3\)]
\[ \rho_p \] : density of coated particle [kg/m\(^3\)]
\[ \phi_v \] : volume shape factor of particle of size \( D \) at \( \theta = \theta \) [-]
\[ \phi_{e_0} \] : volume shape factor of particle of size \( D \) at \( \theta = 0 \) [-]
\[ \bar{\phi}_v \] : average volume shape factor at \( \theta = \theta \) [-]
\[ \bar{\phi}_{e_0} \] : average volume shape factor at \( \theta = 0 \) [-]

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