Changes in Size Distributions of Lime Sorbents during Fluidization

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Abstract—An unsteady state population model is represented to predict the changes in size distribution of bed materials during fluidization. The unsteady state population model is verified experimentally. Change of lime size at two different velocities is examined with the model. The solid size distributions predicted by the model agree well with the corresponding experimental size distributions. In addition, the computational results showed that the decreasing rate of the mass mean diameter at the lower velocity (2 m/s) is greater than that at the higher velocity (4 m/s). The model may be applicable for the batch and continuous operations of fluidized beds in which the solid size reduction is predominantly resulted from attrition and elutriation. Such significance of the mechanical attrition and elutriation is frequently seen in a fast fluidized bed as well as in a circulating fluidized bed.

Key Words: Lime, SO₂, Particle, Attrition, Flue Gas Desulfurization (FGD), Fluidization

INTRODUCTION

Since one of the primary mechanisms in poor sorbent utilization of the dry sorbent injection FGD process is the fact that the products of the SO₂-sorbent reaction have such large molar volumes that they plug the pores necessary for SO₂ diffusion into the unreacted core and the thin product layer of impervious material prevents the fresh portion of the solid sorbent from being exposed to SO₂. The removals of the product layer by attrition would result in greater utilization of the sorbent. Therefore, identifying of attrition mechanisms and the changes in solid size distribution in Circulating Fluidized Bed Absorber (CFBA) play an important role in increasing sorbent utilization of the dry sorbent FGD processes. Moreover, understanding of the extent of attrition would allow the proper design of the particulate collection equipments used for product recycle and wastage.

The changes in size distributions of bed materials as well as the attrition mechanism should be recognized to fulfill the optimal design and operation of a fluidized bed, because performance variables such as minimum fluidization velocity, terminal velocity, elutriation rate, chemical reaction rate, etc. are strongly dependent upon the size of solids. The size changes during fluidization may be predicted by the population balance model which has been developed and applied by others.

Kunii and Levenspiel developed a population model in terms of mass balance to relate the particle size distributions, with feeding rate and outflow in a fluidized bed. The model took into account the size distribution of feed solids, arbitrary particle growth or shrinkage within bed, and the effect of elutriation on the properties of both outflow and carryover under the steady state continuous fluidization, and is applicable for the continuous operations of circulation systems where solids grow in one unit and shrink in the other.

Grimmett presented a general mathematical model which describe the particle dynamics in fluidized bed processes. The model included the various rates of particle growth, particle attrition, elutriation of particles...
from the surface of the bed, and particle feedings to the bed, on the basis of the fluidized bed calcination process. The partial differential equation describing particle size distribution in the fluidized bed calcination process as a function of size and time was numerically solved, and compared with the particle size distributions in the actual calciner runs. He suggested that an elutriation model consistent with actual observations and an expression for the rate of attrition as a function of particle size and the various operating parameters should be developed for the more accurate prediction of the particle size distribution in a fluidized bed process. He particularly emphasized that the lack of information on the attrition rate was so far evident, and that the study of the attrition provided the valuable information to the field of fluidized bed technology.

Chen and Saxena\textsuperscript{3} developed a mechanistic model applicable for the noncatalytic gas-solid reaction in a bubbling fluidized bed. The model described a general steady state population balance in which both solid size and density vary as a result of chemical reaction while the previous models considered only the size changes. However, the model considered the decreased size of solids as a result of elutriation and shrinking chemical reaction, neglecting the attrition effect.

Steconi\textsuperscript{3} considered the particle attrition besides a chemical reaction of unreacted-core by modifying the model developed by Chen and Saxena\textsuperscript{3}. The solid mass balance in his model was carried assuming that the attritted solids are fine and immediately elutriated, and considering that an unreacted core type of chemical reaction occurs together with superficial abrasion and the solid feed has a two dimensional distribution as a function of size and fractional conversion. The model may be extended to reactor-generator systems which are typical of many catalytic reactions.

Overturf and Reklaitis\textsuperscript{10} also developed a particle balance model which accommodated the particle distributions dependent on both size and density as well as populations consisting of multiple solids. The particle balance was imbedded within a nonisothermal Davidson-Harrison bubbling bed model which includes both grid and freeboard region compartments. Comparison of the model was made to the experimental results obtained from the fluidized bed combustor of Babcock and Wilcox, and its results suggested that the satisfactory modelling of the combustors required a more reliable prediction of single-particle elutriation rates.

Ray and Jiang\textsuperscript{3} represented a steady state population model, considering the effect of the fines generated in the particle mixtures of fluidized bed, while Steconi\textsuperscript{3} assumed the fines are immediately blown out. In the model, the evolution of the attritted fines was more realistically assessed so that, the possible effects of fines on the attrition and reaction were considered. They introduced a surface-reaction model under many assumptions in order to assess the effect of fines on attrition rates of a multicomponent mixtures, and stated that the attrition rate for each component is proportional to its surface area, and is also a function of the interactions between different materials. The population model combined with the surface reaction model which describes the effect of fines on the attrition and reaction, however, was not verified experimentally.

In the present study an unsteady state population model is represented to predict the changes in size distribution of bed materials. The unsteady state population model was verified with the experimental data. It may be applicable for the batch and continuous operations of fluidized beds in which the solids size reduction is predominantly resulted from attrition and elutriation. Such significance of the mechanical attrition and elutriation is frequently seen in a fast fluidized bed as well as in a circulating fluidized bed.

**EXPERIMENTAL METHODS**

The attrition tests have been conducted in a bench scale of circulating fluidized bed absorber (CFBA) unit which is schematically shown in Fig. 1, and described in details at others.\textsuperscript{30} The bench scale CFBA unit consists
of a bed reactor of 0.076 cm in diameter and 2.5 m in length, two high efficiency cyclones for gas/solid separation, sorbent injection and recycling system, water injection system, gas heating system, and gas flow, concentration, temperature and pressure monitoring system.

Fig. 1. Scheme of bench scale CFBA unit

The verifying test was carried out in the CFBA with two discrete ranges of Dravo limes. The sizes of the narrowly sieved limes are 903 MICRON for lime-1 sample and 1,764 MICRON for lime-2 sample in mass mean diameter (MMD). Physical and chemical characteristics of the lime samples are summarized in Table 1.

For an attrition test, 0.5 kg of limes were injected so that the initial pressure drop in a bed reactor reach about 15 cm H.O. During the test, the recirculating valve was closed to prevent the attritted fines captured by the first cyclone from entering the bed. At regular time intervals (from 30 minutes to 5 hours) the fluidization was turned off, and the samples from the bed, the first cyclone and second cyclone were immediately taken out. The collected samples were weighted and thus the extent of attrition was obtained. The size distributions of bed materials were also measured by a sieving method. For the measurement of size distribution, the lime samples ranging from 0.2 to 0.25 kg were placed onto the sievers and the sieving time was fixed at 2 minutes to prevent the size reduction of samples in the process of sieving. The lime samples were coarse enough to be separated within 2 minutes.

### RESULTS AND DISCUSSION

#### Population model

A population model has been considered as an useful tool in determining the particle generation rate and the continuous feeding rate as well as particle size distributions for continuous operations in a fluidized bed. The particle population models have mainly been developed and applied under the assumptions of steady-state by many people, while an unsteady-state population model is essential in predicting the changes of size distributions of solids for a batch operation of the fluidized bed.

The unsteady-state population balance is illustrated in Fig. 2. Consider the particles lying the interval \( d_0 \) and \( d_0 + \Delta d_0 \). An unsteady state material balance around the particle size interval may be stated as follows:

\[
\begin{align*}
\text{solids feeding into bed} & + \text{solids recirculating into bed} - \text{solids elutriated from bed} + \\
\end{align*}
\]
Eq. (1) represents the solid population balance. The terms in the equation represent the changes in the number of particles in each size interval due to feeding, elutriation, and recirculation, respectively. Dividing by $\Delta d_p$ in both side of the above equation and taking limits as $\Delta d_p \to 0$, the following equation is obtained:

$$ F_o(t) P_o(d_{p'}, t) + F_r(t) P_r(d_{p'}, t) - F_e(t) P_e(d_{p'}, t) - \frac{d}{dt} \left[ \frac{[\mathcal{R}(d_{p'}) P_b(d_{p'}, t)]}{d_{p'}} \right]_{d_{p'}} - \frac{3 W(t) P_b(d_{p'}, t)}{d_{p'}} \mathcal{R}(d_{p'}) = 0 $$

where $\mathcal{R}(d_{p'}) = \frac{d d_{p'}}{dt}$ is the size-based attrition rate (cm/sec) because the size reduction of lime sorbents occurs due to attrition during fluidization in CFBA. The recirculating solids flow rate, $F_r(t)$, are obtained from the elutriation rate of solids, $F_e(t)$ and the cyclone efficiency, $\eta_c$. The elutriation rate, $F_e(t)$ is a strong function of particle size and for any particular size is represent by the elutriation constant, $\kappa(d_{p'})$ as follows:

$$ F_e(t) P_e(d_{p'}, t) = \kappa(d_{p'}) W(t) P_b(d_{p'}, t) $$

Thus, the recirculating rate of solids is written as follows:

$$ F_r(t) P_r(d_{p'}, t) = \eta_c F_e(t) P_e(d_{p'}, t) = \eta_c \kappa(d_{p'}) W(t) P_b(d_{p'}, t) $$

Substituting Equation (4) and (5) into Equation (3) yields a general unsteady-state partial differential equation to describe the changes of solid size distribution in CFBA system as followings:

$$ F_o(t) P_o(d_{p'}, t) - [1 - \eta_c \kappa(d_{p'}) W(t)] P_b(d_{p'}, t) - \frac{d}{dt} \left[ \frac{[\mathcal{R}(d_{p'}) P_b(d_{p'}, t)]}{d_{p'}} \right]_{d_{p'}} - \frac{3 W(t) P_b(d_{p'}, t)}{d_{p'}} \mathcal{R}(d_{p'}) = 0 $$

where $F_o(t)$ is the solid feeding rate (g/sec), $F_e(t)$, the recirculating rate of solids (g/sec), and $F_e(t)$, the elutriation rate (g/sec) at time $t$. In addition, $W(t)$ is the total weight of solids in a bed (g), and $P_o(d_{p'}, t)$, $P_r(d_{p'}, t)$, $P_e(d_{p'}, t)$, and $P_b(d_{p'}, t)$ are the fractional size distributions (cm⁻¹) of the solid feeding, bed, elutriation, and recirculation, respectively.
For the batch operation of CFBA, the feeding rate \( F_s(t) \) is eliminated, and thus, the following equation is obtained. The equation may be numerically solved to determine the size changes of solid sorbents for a batch operation of CFBA, based on the discrete size distribution of solids, if the elutriation rate, \( \kappa(d_p) \), attrition rate, \( \beta(d_p) \), and cyclone efficiency, \( \eta_c \), are known.

\[
\frac{\partial[W(t)P_b(d_p,t)]}{\partial t} = -(1 - \eta_c)\kappa(d_p)W(t)P_b(d_p,t) - W(t)\frac{d[\beta(d_p)P_b(d_p,t)]}{dd_p} + \frac{3W(t)P_b(d_p,t)}{d_p}\beta(d_p)
\]

(7)

An empirical expression of the elutriation rate for high velocity and large particles is given by Geldart\(^{23} \) as followings:

\[
\kappa(d_p) = 23.7 \left( \frac{\rho_g U_a A_b}{W} \right) \exp \left[ -5.4 \left( \frac{U(d_p)}{U_a} \right)^2 \right]
\]

(8)

where \( \kappa(d_p) \) is the elutriation rate constant (cm\(^3\)), and \( \rho_g \) and \( W \) are gas density (g/cm\(^3\)) and weight of solids (g), respectively. And \( A_b \) is a cross-sectional area of bed (cm\(^2\)), and \( U_a \) and \( U(d_p) \) are gas velocity (cm/s) and terminal velocity of solids (cm/s). The terminal velocity is given as:

\[
U(d_p) = \left[ \frac{4 d_p (\rho_s - \rho_g) g}{3 \rho_a C_D} \right]^{1/2}
\]

(9)

where \( C_D \) is a drag coefficient (dimensionless), and \( \rho_s \) and \( g \) are the density of solids (g/cm\(^3\)) and gravitational constant (cm/sec\(^2\)), respectively. The terminal velocity may be determined by trial and error depending on Reynolds number, \( R_e = d_p \rho_a U(d_p)/\mu \), where \( \mu \) is viscosity. If the size-based attrition rate is substituted into Equation (7), the following equation is obtained:

\[
\frac{\partial[W(t)P_b(d_p,t)]}{\partial t} = -(1 - \eta_c)\kappa(d_p)W(t)P_b(d_p,t) - \frac{2k_a}{3}W(t)P_b(d_p,t) + \frac{5k_a W_{\min p,MMD}}{3W_{o}d_p^3}W(t)P_b(d_p,t)
\]

(10)

For the numerical solution of Equation (10), a discrete size distribution should be employed. The fractional weight distributions in a bed, \( W_b(d_p,t) \), based on the discrete size, \( d_p \), is defined as follows:

\[
W_b(d_p,t) = W(t)P_b(d_p,t) \Delta d_p
\]

(11)

\[
\sum_{i=1}^{n} \frac{W_b(d_p,t)}{W(t)} = \sum_{i=1}^{n} P_b(d_p,t) \Delta d_p = 1
\]

(12)

With substitution of Equation (11) and (12) into Equation (10), the population equation based on the discrete sizes is written as followings:

\[
\frac{\partial[W_b(d_p,t)]}{\partial t} = \left[ (1 - \eta_c)\kappa(d_p) + \frac{2k_a}{3} - \frac{5k_a W_{\min p,MMD}}{3W_{o}d_p^3} \right] W_b(d_p,t)
\]

(13)

\[
+ \frac{k_a}{3} \left( \frac{d_p - W_{\min p,MMD}}{W_{o}d_p^2} \right) \frac{d[W_b(d_p,t)]}{dd_p}
\]

Where the initial condition, \( W_b(d_p, t) = W_b(d_p) \) at \( t = 0 \) is obtained by measuring the size distribution of the lime sorbents before fluidization. The boundary conditions are given by:

for \( t > 0 \),
\[
W_b(d_{pl},t) = 0, \text{ at } d_{pl} = 0
\]
\[
W_b(d_{pl},t) = 0, \text{ at } d_{pl} = d_{pl,max} + \Delta d_p
\]

An implicit method based on the backward Euler method is used for the numerical computation of Equation (13). Thus, the following finite difference equation is written:
\[
\left( \frac{1 + \alpha}{\beta} \right) W_{ij+1} - \left( \frac{\alpha}{\beta} \right) W_{i,j+1} = W_{ij} \quad (14)
\]

Where \( d_{w} \) and \( t \) are replaced with \( i \) and \( j \), respectively, and \( \alpha \) and \( \beta \) are:
\[
\alpha = \frac{k^a}{3} \left( d_{pl}^3 - \frac{W_{\min}^3 d_{pl, MMD}^3}{W_o d_{pl}^2} \right) \left( \frac{\Delta t}{\Delta d_p} \right)
\]
\[
\beta = 1 - \left( 1 - \eta_r \right) \frac{k^a}{3} - \frac{5 k W_o d_{pl}^3}{3 W_o d_{pl}^3} \Delta t
\]

The Equation (14) may be written in a diagonal matrix form, and thus, the unknowns, \( W_{i+1,j}, W_{2j+1}, \ldots, W_{n+1,j} \) are obtained by iterations, together with the initial and boundary conditions. The input data necessary for the computations are shown in Table 2.

In a computer program the fractional weights measured at discrete sizes are divided by the difference between the lower and upper ranges and multiplied by the size increment to give the initial fractional weight, \( W_o \) at time, \( t=0 \) and each size, \( d_{w} \). In other words the initial fractional weight at each size within the lower and upper ranges is assumed as the same. The terminal velocities for the computations of the elutriation rate at each size and time interval are obtained by trial and error, depending on the Reynolds number. The size and time interval may be smaller to give more accurate

Table 2. Batch operating conditions of CFBA

<table>
<thead>
<tr>
<th>Diameter ranges (( \mu m ))</th>
<th>( W_o(d_{w}0) ) (%)</th>
<th>Diameter ranges (( \mu m ))</th>
<th>( W_o(d_{w}0) ) (%)</th>
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<tbody>
<tr>
<td>100 - 297</td>
<td>1.5</td>
<td>300 - 595</td>
<td>0.9</td>
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<tr>
<td>237 - 420</td>
<td>0.1</td>
<td>595 - 841</td>
<td>0.1</td>
</tr>
<tr>
<td>420 - 595</td>
<td>0.4</td>
<td>841 - 1008</td>
<td>0.1</td>
</tr>
<tr>
<td>595 - 841</td>
<td>31.3</td>
<td>1008 - 1190</td>
<td>4.0</td>
</tr>
<tr>
<td>841 - 1000</td>
<td>31.9</td>
<td>1190 - 2000</td>
<td>61.3</td>
</tr>
<tr>
<td>1000 - 1190</td>
<td>34.7</td>
<td>2000 - 2380</td>
<td>33.6</td>
</tr>
<tr>
<td>Total</td>
<td>100</td>
<td>Total</td>
<td>100</td>
</tr>
</tbody>
</table>

**Operating conditions**

- Density of Particle: 2.1 (g/cm³)
- Density of Gas: 1.02E-03 (g/cm³)
- Viscosity: 1.78E-04 (g/cm/sec)
- Gravitational Constant: 980.66 (cm/sec)
- Bed Diameter: 7.52 (cm)
- Bed Height: 250 (cm)
- Gas Velocity: 200 and 400 (cm/sec)
- Attrition Rate Constant: 4.82E-5 & 9.09E-5 (sec⁻¹)
- Size Interval: 10 (\( \mu m \))
- Time Interval: 10 (sec)
Fig. 3. Size distribution of lime sorbents after 30 minutes of fluidization ($u = 2m/s$ for lime 1 and $u = 4m/s$ for lime 2)

Lime 1
MMD measured = 856 microns
MMD predicted = 697 microns

Lime 2
MMD measured = 1584 microns
MMD predicted = 1724 microns

Fig. 4. Size distribution of lime sorbents after 1 hour of fluidization ($u = 2m/s$ for lime 1 and $u = 4m/s$ for lime 2)

Lime 1
MMD measured = 823 microns
MMD predicted = 863 microns

Lime 2
MMD measured = 1509 microns
MMD predicted = 1672 microns

results, depending on the computer capacity since the size interval greater than 40 MICRON and the time increment greater than 20 sec may cause an unacceptable computational result.

Model verification and application for a batch operation
The fractional size distribution curves obtained from the computations and corresponding measured size distributions after fluidization of 30 minutes, 1 hour, 2 hours, 3 hours, and 5 hours are shown in Fig. 3 - Fig. 7. As illustrated in the figures, the size distributions computed by the unsteady state population model agree...
Changes in Size Distributions of Lime Sorbents during Fluidization

Fig. 7. Size distribution of lime sorbents after 5 hours of fluidization ($u = 2\text{m/s}$ for lime 1 and $u = 4\text{m/s}$ for lime 2)

Fig. 8. Changes of Solid size distribution at the velocity of 4m/s

Fig. 9. Changes of Solid size distribution at the velocity of 4m/s

well with the experimental data. However, slightly lower estimation occurs at the lower size ranges because elutriation of solids are considered in the model.

The effect of gas velocity on the size distribution of solids is computed and illustrated in Fig. 8 and Fig. 9. The results was obtained under the same operating conditions and with an arbitrary initial size distribution at the different velocities. As shown in Fig. 8 and Fig. 9, the fractional weight distribution of initial solids was chosen as normal gaussian distribution for the purpose of illustration. The Fig. 8 and Fig. 9 show that the decreasing rate of the mass mean diameter at the lower velocity (2 m/s) is greater than that at the higher velocity (4 m/s), while the size range at 2 m/s...
becomes much wider than 4 m/s. The results suggest that the solids becoming rapidly smaller at the higher velocity are easily elutriated and only relatively coarse solids remains in a bed as the fluidization time increases. As a result, size reduction of the remaining parent solids may be relatively slower at higher velocity than lower velocity.

CONCLUSIONS

An unsteady state population model is represented to predict the changes in size distribution of bed materials during fluidization. The unsteady state population model is verified experimentally. The results showed that the solid size distribution predicted by the model agreed well with the corresponding experimental size distributions. Therefore, the model may be applicable for the batch and continuous operations of fluidized beds in which the solids size reduction is predominantly resulted from attrition and elutriation. Such significance of the mechanical attrition and elutriation is frequently seen in a fast fluidized bed as well as in a circulating fluidized bed.

Change of lime size at two different velocities is examined with the model. The computational results showed that the decreasing rate of the mass mean diameter at the lower velocity (2 m/s) is greater than that at the higher velocity (4 m/s).

NOTATION

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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<tbody>
<tr>
<td>(A_s)</td>
<td>cross-sectional area of bed reactor, m²</td>
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<tr>
<td>(C_b)</td>
<td>drag coefficient</td>
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<tr>
<td>(d_p)</td>
<td>diameter of parent solid, (\mu m)</td>
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<tr>
<td>(d_{MMMD})</td>
<td>mass mean diameter of solid, (\mu m)</td>
</tr>
<tr>
<td>(F_e(t))</td>
<td>elutriation rate of solids, kg/s</td>
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<tr>
<td>(F_f(t))</td>
<td>feeding rate of solids, kg/s</td>
</tr>
<tr>
<td>(F_r(t))</td>
<td>recirculating rate of solids, kg/s</td>
</tr>
<tr>
<td>(g)</td>
<td>gravitational constant</td>
</tr>
<tr>
<td>(k_e)</td>
<td>attrition rate constant, s⁻¹</td>
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<tr>
<td>(P_e(d_{*}, t))</td>
<td>fractional size distributions of elutriating solids, cm⁻¹</td>
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<tr>
<td>(P_f(d_{*}, t))</td>
<td>fractional size distributions of feeding solids, cm⁻¹</td>
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<td>(P_r(d_{*}, t))</td>
<td>fractional size distributions of recirculating solids, cm⁻¹</td>
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<td>(t)</td>
<td>time, s</td>
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<tr>
<td>(U_s)</td>
<td>superficial gas velocity, m/s</td>
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<td>(U_t)</td>
<td>terminal velocity, m/s</td>
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<tr>
<td>(W)</td>
<td>weight of solids, kg</td>
</tr>
<tr>
<td>(W_0)</td>
<td>fractional weight distribution in a bed</td>
</tr>
<tr>
<td>(W_{min})</td>
<td>minimum weight of parent solids in a bed, kg</td>
</tr>
<tr>
<td>(W_i)</td>
<td>initial weight of solids, kg</td>
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Greek Letters

<table>
<thead>
<tr>
<th>Symbol</th>
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<tr>
<td>(\eta)</td>
<td>cyclone efficiency</td>
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<tr>
<td>(\kappa(d_p))</td>
<td>elutriation rate constant, m¹</td>
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<tr>
<td>(\mu)</td>
<td>viscosity, kg/m/s</td>
</tr>
<tr>
<td>(\rho_g)</td>
<td>density of gas, kg/m³</td>
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<tr>
<td>(\rho_s)</td>
<td>density of solid, kg/m³</td>
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<tr>
<td>(\mathcal{R}(d))</td>
<td>size-based attrition rate, m/s</td>
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REFERENCES

Technology, 2, 87-96 (1968).


