A Model for Predicting the Size Distribution of Product from a Granulating Drum

By J. D. LITSTER,** A. G. WATERS** and S. K. NICOL**

Synopsis

Carefully controlled granulation experiments were carried out on a hematite ore with a wide size distribution (0~8 mm) over a range of moisture contents. At any one moisture content, the finest size fractions could be classified solely as layering particles whilst the largest size fractions acted only as nuclei particles. Between these two extremes, a proportion of particles in any one size fraction behaved as nuclei particles, the remainder being layering particles. Hence each particle size fraction could be characterised by a partition coefficient. The partitioning of an individual size range between nuclei and adhering particles was shown to be dependent on the moisture content. For each moisture content, the partition curve could be modelled using a log-normal function. The variation of adhering layer mass with granule size was investigated and it was found that adhering layer thickness was approximately proportional to nuclei diameter at lower moisture contents. At higher moisture contents there was an optimum nuclei particle size for which the ratio of granule mass to total nuclei mass was a maximum.

A population balance model to predict granule size distribution was developed and tested against experimental data. The important model parameters were those describing the particle partition curve. Excellent agreement was found between experimental and model predicted granule size distributions for the full range of moisture contents covered.

I. Introduction

Population balance type models have been used successfully to model the kinetics and the change in size distribution of pelletisation processes6-31 where the feed is very fine, typically 90 %, by mass less than 0.075 mm. The pellets that form have a fairly narrow size distribution the shape of which is conserved as the pellets grow.31 In contrast, the feed to the granulating process prior to iron ore sintering has a wide size distribution, typically 0~10 mm, and the size distribution changes significantly during the granulating process.

It is largely accepted that the principal mechanism in sinter feed granulation is the layering of fine particles onto larger nuclei particles. Microscopic studies4,5 have shown clearly that large granules or “ quasi-particles ” consist of a nucleus surrounded by a layer of fines. However, the approach to characterising the granulation process has generally been empirical in nature. Only Rankin and Rolle46 have attempted to predict granule size distribution based on the feed size distribution, but their model assumptions have not been fully tested. In this present work, carefully controlled laboratory experiments have been undertaken to study mechanisms involved in the granulation of iron ore with a wide size range. Based on these experiments, a preliminary model to predict granule size distribution given the feed size distribution is proposed and tested.

II. Experimental Equipment and Techniques

All experiments were carried out in a stainless steel batch granulating drum 0.285 m long and 0.310 m in diameter. The drum contained six wedge shaped lifter bars, each 4 mm high (after Kapur and Fuerstenau7). No scraper bar was used. The drum rotated on two rollers and the drum speed was controlled by a variable speed gear box. A measured mass of water was added to the granulation mix via a syringe and tube arrangement. The 6 mm diameter stainless steel tube, with 1 mm diameter holes drilled at a 5 mm spacing, ran the full length of the drum. The holes were aligned so that the water sprayed onto the material tumbling in the drum and not onto the drum itself. The sample mass (5 kg) and drum speed (20 rpm) used correspond to a space factor of 12 % and a Froude number of 0.034. These values correspond to cascade zone movement of the charge in the granulating drum which has been recommended for good granulation.9

For the experiments reported in this paper, a single component system consisting of a high grade haematite ore from Western Australia was used. The experimental procedure was as follows. The ore was carefully screened and the sized fractions were then dried in an oven at 105 °C for 12 h. The required mass of each of the size fractions was weighed and placed in the granulating drum. The drum was sealed and rotated for 2 min to mix the feed. While the drum continued to rotate, the required amount of water was added over a period of 0.5 to 1 min. The drum was allowed to rotate for a further 5 min and then stopped. The granulated material was transferred to a rotary sample divider where a 1 kg sample and two 0.5 kg samples were split out. The 1 kg sample was weighed before and after drying in an oven for 12 h at 105 °C to determine the total moisture content. One 0.5 kg sample was used for frozen granule size analysis and the other was kept as a spare.

Granule size analysis was performed by freezing the granules in liquid nitrogen then screening. The screened samples were dried for 12 h then weighed. This technique is fully described elsewhere.9 In some experiments a size analysis was performed on each granule size fraction obtained from the frozen size distribution screening. This was done by first wet screening the individual granule size fractions at 0.063 mm, then dry sizing the plus 0.063 mm material. All size analyses were carried out using a full 6 sieve series.

* Manuscript received on December 10, 1985; accepted in the final form on September 12, 1986. © 1986 ISIJ
III. Results and Discussion

1. Dependence of Layer Thickness on Nuclei Particle Size

The dependence of adhering layer thickness on nuclei particle size has received little attention in the literature but it is necessary to know the nature of this dependence to develop a population balance model for the prediction of granule size distribution. Rankin and Roller assumed that layer thickness was independent of nuclei particle size although this assumption was not fully tested. In the present work, three experiments were carried out to test this assumption. Feed to the drum consisted of three size fractions: minus 0.25 mm (layering particles), 1 to 2 mm and 4 to 5.6 mm (nuclei particles). The proportion of fines was held constant (25 %) and the mass ratio of large to small nuclei particles was varied. The mass of granules formed from each nuclei particle size range was measured and the mass of adhering particles was determined by difference.

Results are shown in Table 1. If the layer thickness was independent of particle size, the mass of adhering layer would be proportional to the total surface area of nuclei particles, i.e.,

\[
\text{Mass of adhering layer on } 4-5.6 \text{ mm particles} = \frac{\text{Mass of adhering layer on 1-2 mm particles}}{\text{Total surface area of 1-2 mm particles}}
\]

From Table 1, it is clear that this was not the case. The ratio of the mass of fines adhering to the large particles compared to that adhering to the smaller particles was 3 to 5 times greater than that predicted on this assumption. Rather, Table 1 shows that the mass of adhering fines was approximately proportional to the nuclei particle mass rather than surface area. On this basis, the proportion of layering fines on each nuclei size fraction mass was within ±30 % of the observed ratio. Given a constant ratio of adhering layer mass to nuclei particle mass, it follows that layer thickness is proportional to nuclei particle size. This assumption will be used in the model development outlined in Chap. IV. The dependence of adhering layer mass on nuclei particle mass suggests that during the granulation process, the ability of a particle to grow an adhering layer of fines depends on its momentum. Using tracer studies, Sastry and Fuerstenau showed that transfer of a small amount material from one pellet to another by abrasion during a collision was an important growth mechanism in pelletisation. In the granulation process, a collision between a large, high momentum particle and a small, low momentum particle could result in the transfer of some of the adhering layer from the small particle to the large particle. As a result of this mechanism, the mass of adhering layer that a particle could hold would be dependent on the particle mass.

2. Granulation of a Wide Size Range Feed

A series of experiments were carried out using a feed with a wide size range and varying the moisture content from 3.5 to 5.4 %. The feed size distribution used was similar to that of an operating sinter plant (see Table 2). Granule size distributions for the different moisture contents are shown in Fig. 1. Clearly, the extent of granulation increases with moisture. For each experiment, the size distribution of particles making up each granule size fraction was measured by the wet screening procedure described above. From this data, the partitioning of particles of a particular size into the various granule size fractions can also be determined. An example of the complete results from one experiment is given in Table 3.

From the particle partitioning data, it is possible to observe which particle size fractions act as nuclei and which act as adhering particles. Assuming all size enlargement is by layering and the extent of coalescence is minimal, particles which act as nuclei will appear in granules of the same sieve size fraction or the next larger sieve size fraction due to the adhering layer of fines increasing the particle size by a small amount. Particles which form part of the adhering layer around a larger nucleus particle will appear in all the larger granule size fractions. The partition coefficient \( a_i \) is therefore defined as the mass fraction of particles from size fraction \( i \) which act

Table 2. Feed size distribution.

<table>
<thead>
<tr>
<th>Sieve size (mm)</th>
<th>mass % passing</th>
</tr>
</thead>
<tbody>
<tr>
<td>8.0</td>
<td>100</td>
</tr>
<tr>
<td>5.6</td>
<td>95.1</td>
</tr>
<tr>
<td>4.0</td>
<td>78.1</td>
</tr>
<tr>
<td>2.8</td>
<td>60.4</td>
</tr>
<tr>
<td>2.0</td>
<td>49.7</td>
</tr>
<tr>
<td>1.4</td>
<td>42.2</td>
</tr>
<tr>
<td>1.0</td>
<td>36.0</td>
</tr>
<tr>
<td>0.71</td>
<td>31.1</td>
</tr>
<tr>
<td>0.50</td>
<td>26.2</td>
</tr>
<tr>
<td>0.355</td>
<td>22.3</td>
</tr>
<tr>
<td>0.25</td>
<td>18.8</td>
</tr>
<tr>
<td>0.18</td>
<td>17.0</td>
</tr>
<tr>
<td>0.125</td>
<td>15.5</td>
</tr>
<tr>
<td>0.090</td>
<td>14.2</td>
</tr>
<tr>
<td>0.063</td>
<td>12.8</td>
</tr>
</tbody>
</table>

Research Article
Fig. 1. Variation in granule size distribution with moisture content.

Table 3. Granule size distributions and particle partition distributions for 4.3% moisture.

(a) Granule size distributions
mass% of granule size $j$ made up from particle size $i$

<table>
<thead>
<tr>
<th>Particle size (mm)</th>
<th>Granule size (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>+8</td>
</tr>
<tr>
<td>+5.600</td>
<td>28.4</td>
</tr>
<tr>
<td>+4.000</td>
<td>7.5</td>
</tr>
<tr>
<td>+2.800</td>
<td>0.0</td>
</tr>
<tr>
<td>+2.000</td>
<td>3.0</td>
</tr>
<tr>
<td>+1.400</td>
<td>1.5</td>
</tr>
<tr>
<td>+1.000</td>
<td>1.5</td>
</tr>
<tr>
<td>+0.710</td>
<td>4.5</td>
</tr>
<tr>
<td>+0.500</td>
<td>4.5</td>
</tr>
<tr>
<td>+0.355</td>
<td>6.0</td>
</tr>
<tr>
<td>+0.250</td>
<td>7.5</td>
</tr>
<tr>
<td>+0.180</td>
<td>3.0</td>
</tr>
<tr>
<td>+0.125</td>
<td>3.0</td>
</tr>
<tr>
<td>+0.090</td>
<td>3.0</td>
</tr>
<tr>
<td>+0.063</td>
<td>3.0</td>
</tr>
<tr>
<td>-0.063</td>
<td>23.9</td>
</tr>
</tbody>
</table>

(b) Particle partition distributions
mass% of particle size $i$ reporting to granule size $j$

<table>
<thead>
<tr>
<th>Granule size (mm)</th>
<th>Particle size (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>+5.6</td>
</tr>
<tr>
<td>+8.0</td>
<td>7.4</td>
</tr>
<tr>
<td>+5.6</td>
<td>92.6</td>
</tr>
<tr>
<td>+4.0</td>
<td>79.8</td>
</tr>
<tr>
<td>+2.8</td>
<td>75.7</td>
</tr>
<tr>
<td>+2.0</td>
<td>80.4</td>
</tr>
<tr>
<td>+1.4</td>
<td>75.9</td>
</tr>
<tr>
<td>+1.0</td>
<td>57.8</td>
</tr>
<tr>
<td>+0.710</td>
<td>47.3</td>
</tr>
<tr>
<td>+0.500</td>
<td>37.6</td>
</tr>
<tr>
<td>+0.355</td>
<td>20.7</td>
</tr>
<tr>
<td>+0.250</td>
<td>12.0</td>
</tr>
<tr>
<td>-0.250</td>
<td>13.5</td>
</tr>
</tbody>
</table>

Research Article
as nuclei particles, and can be calculated from the experimental data as follows:

\[ a_i = \frac{m_{ij} + m_{ij+1}}{\sum_{j=1}^{n} m_{ij}} \] ............................(1)

where, \( m_{ij} \): the mass of particles of size fraction \( i \) which are found in granule size fraction \( j \).

Values of \( a_i = 1 \) and \( a_i = 0 \) correspond to nuclei and adhering size fractions, respectively. Figure 2 shows the values of \( a_i \) as a function of particle size for various moisture contents. It can be seen that there are a significant number of intermediate size fractions where some of the particles act as nuclei and some of the particles are adhering, i.e., \( a_i \) lies somewhere between 0 and 1. For example, at a moisture content of 5.0%, \( a_i = 0.1 \) corresponds to a particle size of 0.29 mm whereas \( a_i = 0.9 \) corresponds to 1.4 mm. Particles between these two sizes behave as nuclei and adhering particles. These observations differ from the conclusions of some previous workers. Furui et al. and Vidal et al. while acknowledging the presence of intermediate particles claimed they took no part in the granulation process and remained unchanged. It was claimed that all granules were made up of a large nucleus surrounded by a layer of ultrafines. Intermediate sized particles neither acted as nuclei or adhered to larger particles. From Fig. 2, however, it is clear that the intermediate particle size ranges do take part in the granulation process, some as nuclei and some as layering particles. These conclusions are supported by microscopic observation of green feed granules sampled from an operating sinter plant (see Photo. 1). The curves shown in Fig. 2 closely resemble a cumulative log normal distribution plot and the same data is plotted on log probability paper in Fig. 3. In all cases a straight line gives a good fit to the data. Thus, for a particular moisture content, the relationship between partition coefficient and particle size can be represented by the equation:

\[ a(x) = \frac{1}{\sigma \sqrt{2\pi}} \int_{-\infty}^{\ln(x)} \exp \left[ -\frac{(t - \ln(x_{0.5}))^2}{2\sigma^2} \right] dt \] ...........................(2)

where, \( x_{0.5} \): the particle size with a partition coefficient of 0.5

\( \sigma \): a measure of the spread of size range of intermediate particles.

The position of the partition curve moves towards the larger particle size fractions with increasing moisture content. Thus, particles that act as nuclei at low moisture contents may adhere at higher moisture contents. For example, at 3.5% moisture, 75% of the 0.5 mm particles are nuclei while at 5.4% moisture 85% are adhering (see Fig. 2). The variation of \( x_{0.5} \) and \( \sigma \) with moisture content is shown in Fig. 4. \( x_{0.5} \) increases with moisture. This is analogous to the increase in cut off size between adhering and nuclei particles with moisture found by Rankin and Roller. The value of \( \sigma \) decreased with increasing moisture content. In other words, the range of intermediate sizes became smaller as the moisture content increased. The values of these parameters at a given moisture content will be strongly dependent on the feed particle size distribution. At this stage, no attempt has been made to quantify the effect on \( x_{0.5} \) and \( \sigma \) of changing

Photo. 1. Section of green feed granules showing intermediate size particles (125-250 µm) acting as nuclei (A) and adhering particles (B).

Fig. 2. Variation in partition coefficient with particle size.

Fig. 3. Log-probability plot of partition coefficient vs. particle size.
the size distribution.

IV. Model Description

With information about particle partitioning, a population balance model can be developed to predict the final granule size distribution from the feed particle size distribution. At this stage, the model is for a single component feed only and is an equilibrium model taking no account of granulation kinetics. The major assumptions of the model are:

1. Granulation occurs solely by the mechanism of fine particles layering on larger nuclei particles.
2. For a particular size fraction, the proportion of particles acting as nuclei can be represented by a partition coefficient, \( a(x) \), which decreases from 1 and approaches 0 as the particle size decreases.
3. The thickness of the adhering layer of fines is proportional to the size of the nucleus particle.

The model is developed as follows. All masses are on a dry basis. Let \( y_1 \) and \( y_2 \) be the size distribution density functions of the feed particles and product granules respectively. Consider a particle of size \( x \) with a layer thickness of \( \epsilon(x) \).

Mass of particles size \( x \) to \( x+dx \) available as nuclei

\[
M_y(x)a(x)dx \quad \text{..................................(3)}
\]

No. of particles of size \( x \) to \( x+dx \) available as nuclei

\[
6M_y(x)a(x)dx \quad \text{..................................(4)}
\]

Mass of adhering layer on a particle of size \( x \) to \( x+dx \)

\[
\rho \epsilon(x)\pi x^2 \quad \text{..................................(5)}
\]

Therefore, Total mass of adhering layer on all particles of size \( x \) to \( x+dx \)

\[
\rho \epsilon(x)\pi x^2 J(x) \cdot \frac{6M_y(x)a(x)dx}{\rho \phi_x \pi x^2} \quad \text{..................................(6)}
\]

where \( \phi_x \) and \( \phi_o \) are the surface area and volume shape factors respectively defined by

\[
V_p = \frac{\phi_o \pi x^3}{6} \quad \text{and} \quad S_p = \phi_s \pi x^2
\]

Nuclei particles of size \( x \) to \( x+dx \) will form granules of size \( x+2\epsilon \) to \( x+dx+2\epsilon \). From Eqs. (3) and (7) the mass of granules of size \( x+2\epsilon \) to \( x+dx+2\epsilon \) is:

\[
M_{y_2}(x+2\epsilon)dx = M_y(x)a(x)dx + \frac{3\rho_l \rho_y}{\rho_o \rho_p} \cdot \frac{y_1(x)a(x)2J(x)Mdx}{x} \quad \text{..................................(8)}
\]

Rearranging

\[
y_2(x+2\epsilon) = y_1(x)a(x) \left[ 1 + \frac{K \cdot 2J(x)}{x} \right] \quad \text{..................................(9)}
\]

where \( K = \frac{3\rho_l \rho_y}{\rho_o \rho_p} \), which is a combination of material properties.

Since \( J \) is proportional to \( x \),

\[
2J(x) = Ax \quad \text{..................................(10)}
\]

Substituting in Eq. (9):

\[
y_2(x+Ax) = y_1(x)a(x)(1+KA) \quad \text{..................................(11)}
\]

Integrating Eq. (7), the total mass of adhering material on nuclei of all sizes

\[
MKA \int_0^\infty y_1(x)a(x)dx \quad \text{..................................(12)}
\]

The total mass of particles available for layering

\[
M \left( 1 - \int_0^\infty y_1(x)a(x)dx \right) \quad \text{..................................(13)}
\]

Equating Eqs. (12) and (13):

\[
A = \frac{1 - \int_0^\infty y_1(x)a(x)dx}{K \int_0^\infty y_1(x)a(x)dx} \quad \text{..................................(14)}
\]

\( a(x) \) is given by Eq. (2) above. Thus, given \( x_{0.5}, a \) and \( K \), Eqs. (2), (14) and (11) can be solved to give the granule size distribution. In a form to use a discrete size distribution, Eqs. (11) and (14) are:

\[
y_2(x) = y_1(x)(1+KA) \quad \text{..................................(11-a)}
\]

\[
A = \frac{1 - \sum_{i=1}^{n} y_1(x_i)a(x_i)dx}{K \sum_{i=1}^{n} y_1(x_i)a(x_i)dx} \quad \text{..................................(14-b)}
\]

and \( x_{0.5} = x_{0.5} + A\delta x \quad \text{..................................(15)}\)

The model has three parameters—\( K \), \( x_{0.5} \) and \( \sigma \). \( K \) is a lumped parameter which relates layer thickness to the mass of layering particles. The voidage of the adhering layer will be approximately 0.3 to 0.4. Therefore, on a dry basis, \( \rho_l/\rho_p \) will be in the range 0.6 to 0.7. Allen(11) gives values of \( \phi_x/\phi_o \) for a wide range of materials. All values lie in the range 1.5 to 2.7. Therefore, a realistic range for \( K \) is 2.7 to 5.6. Figure 5 shows the granule size distribution.
predicted by the model using the feed size distribution shown in Table 2 and values of \(x_{0.5}\) and \(\sigma\) of 0.45 mm and 0.737, respectively (4.3 % moisture). Values of \(K\) of 2.7, 4.0 and 5.6 are used. Clearly the granule size distribution is insensitive to changes in \(K\) over a realistic range of values. A value of \(K\) of 4 is therefore chosen for use in the model. Thus, the model becomes effectively a two parameter model. It should be noted, however, that the assumption that granule size distribution is insensitive to the value of \(K\) is only strictly valid when the shape factor ratio \(\phi_s/\phi_n\) is independent of particle size. This is a valid assumption for a one component system for particles larger than 0.125 mm.\(^6\) Although the particle shape factors appear explicitly in the model only in the lumped parameter \(K\), it is not implied that particle shape is unimportant in the granulation process. Particle shape is very important in determining the efficiency with which particles act as nuclei\(^5\) and hence the partition coefficient curve for a particular material can be expected to depend strongly on \(\phi_s\) and \(\phi_n\).

\[\text{Mass granules/Mass nuclei}_j = \sum m_j/m_{j-1} + m_j\]

\[\text{Fig. 5. Sensitivity of model to the value of } K. \]

\[x_{0.5}=0.45 \text{ mm, } \sigma=0.737.\]

\[\text{Fig. 6. Comparison of model and experimental size distributions at a moisture content of 5.4 %}.\]

V. Comparison of Model and Experiment

Comparisons of experimental and model predicted granule size distributions are shown in Figs. 6 to 10. In each case the values of \(x_{0.5}\) and \(\sigma\) used were those calculated from the experimental data (see above). It can be seen that for all moisture contents, there is excellent agreement between the experimental and model predicted size distributions. As the partition coefficients were taken from experimental data, these results confirm the model assumption that the adhering layer thickness is proportional to nuclei particle size is adequate.

The experimental distribution of the total mass of layering material with granule size can be determined from the particle size distribution of each granule size fraction and is shown in Fig. 11 as a function of moisture content. By the same argument used for the particle partitioning calculations, nuclei particles are defined as those from the same size fraction as the granules or one size fraction smaller, \(i.e.,\)

\[\text{(Mass granules/Mass nuclei)}_j = \sum m_j/m_{j-1} + m_j.\]

\[\text{Fig. 7. Comparison of model and experimental size distributions at a moisture content of 3.2 %}.\]
The preliminary experiments described in Sec. III.1 suggested that this ratio was independent of granule size. Figure 11 confirms that this is approximately true for a wide size range feed at the lower moisture contents. As the moisture content is increased, the ratio of granule mass to nuclei mass exhibits a maximum value. In other words, at higher moisture contents, there is an optimum nuclei particle size. This suggests that there are competing mechanisms which control the relationship between adhering layer

Fig. 7. Comparison of model and experimental size distributions at a moisture content of 5.0%.

Fig. 8. Comparison of model and experimental size distributions at a moisture content of 4.5%.

Fig. 9. Comparison of model and experimental size distributions at a moisture content of 4.0%.
thickness and particle size and that the process is more complex than the simple mechanism suggested in Sec. III.1. However, it is clear from a comparison of experimental and model predicted granule size distributions that the simplifying model assumption is justified.

The preliminary model developed in this paper shows great potential for use in the study of the granulation process and for comparing the granulating ability of different ores. There are several areas in which the model requires development and testing. Before the model can be applied to sinter feed, further work is necessary to:

1) Predict changes in model parameters with changes in feed size distribution,
2) Extend the model to cover multicomponent feeds.

VI. Conclusions

From this study the following conclusions are drawn:

(1) There is a smooth transition from completely adhering to completely nuclei particles as particle size increases. In the intermediate size ranges, for one particular size fraction, some particles act as nuclei while other particles of the same size are adhering.

(2) The distribution of particles between adhering and nuclei characteristics can be represented in terms of a partition curve. The shape and position of the partition curve are functions of the moisture content. The variation in partition coefficient with particle size can be adequately represented by a log-normal function.

(3) The thickness of the adhering layer of particles is approximately proportional to the nuclei particle size at lower moisture contents. At higher moisture contents, there is an optimum nuclei size for which the ratio of granule mass to nuclei mass is a maximum.

(4) The preliminary model presented in this paper shows excellent agreement with measured granule size distribution.

Nomenclature

\[ A: \] Proportionality constant between layer thickness and particle size

\[ K: \] Model parameter related to particle density and surface area characteristics

\[ M: \] Total mass of system (dry basis)
Acknowledgement

The authors wish to thank Mr P. W. Roller for useful comment and discussion.

REFERENCES