CHAPTER 3

A REVIEW OF GRANULATION PROCESS SIMULATION MODELS

In this thesis, a good starting point is to review appropriate mathematical models which can be used to simulate the fertilizer granulation process shown in figure 1-2. The drum granulator is the key to particle size change and then the screen and the crusher will be added to complete the simplest granulation circuit. The suitable mathematical models for drum granulator, screen and crusher must be tested to validate their suitability.

3.1 A Review of Drum Granulator Models

A review of the existing models will be carried out. The justification for the suitability and limitation of the models will also be carried out. Several of the granulation techniques found in the literature are applied to fluidized granulator, pan granulator and spray granulator. The rotating drum granulator is the one of interest here.

3.1.1 The drum granulator model

Lister's Model and limitation

Lister et al. (1986), Lister and Waters (1988, 1990) and Water et al (1989) have developed a model for predicting the size distribution of product from a drum granulator using data obtained from the batch granulation of a wide initial size

distribution of iron ore sinter feed particles. Assuming that granule growth is mainly via pseudo-layering (preferential coalescence), the iron ore pellets are partitioned into layering and nuclei particles thereby neglecting the effect of random coalescence. During granulation, a layer of fines is assumed to have attached itself onto the nuclei particle to produce a granule of larger size.

There are some of the shortcomings associated with Lister's model to limit the model performance. Lister's model gives no information as to the kinetics of the granule size distribution. This is very important in fertilizer drum granulator modeling. The model is based on the assumption that granule growth is mainly via pseudo layering, thereby neglecting the effect of random coalescence which may be important in the first stage of granulation.

In view of these shortcomings, it is concluded that Lister's model is not so suitable for use in modeling the fertilizer drum granulator.

Sherrington's Model

Sherrington (1968) has developed a model that is capable of predicting the kinetics of the median granule diameter. The model correlates the average granule diameter with the solution phase ratio.

Sherrington (1968) postulates that for a well packed mass of particles, the solution phase ratio, y, is related to the average granule diameter.

In developing the model equation to describe the granule size diameter, it is assumed that ;

1). the resulting granules are spherical and uniformly sized,

2). the granule is composed of uniformly sized particles of average radius and

3). the outer surface of the granule is surface dry, i.e. the liquid is withdrawn by capillary suction, from the granule's outer surface, to an average depth into its interstices.

Limitations of the Sherrington's Model

Adetayo (1993) explained that due to the complexities of the granulation process the parameters in the modified Sherrington's model cannot be predicted but must be determined from experimental data by a graphical method.

i) Solubility, other binder properties such as viscosity, surface tension, deformability etc. also have a significant effect on the granulation process and can not be neglected. These properties are not taken into account by the Sherrington model. This results in the establishment of different empirical correlations for each material.

ii) Sherrington's model assumes negligible effect of kinetics. It gives no information as to the dynamics of the average granule diameter. The correlation has to be validated for every case otherwise it only gives information on the median granule diameter at steady state.

iii) Sherrington's approach only gives information about the average granule diameter. It says nothing of the size distribution of the granules as a whole.

iv) The development of the model is based on the assumption of a uniformly distributed initial feed material. For a wide initial granule size distribution the choice of the "average diameter" is not clear.

In short, Sherrington's model is not suitable for modeling the fertilizer drum granulator which encounters the effect of wide size distribution of fertilizer particles.

Adetayo's Model

Adetayo et al. (1993) have developed a mathematical model by applying the population balance model of Hounslow (1990) to the rotating drum granulator at the laboratory scale.

The population balance model is applied to describe the drum granulation of feed with bound initial size distribution (e.g. recycle fertilizer granules). The model utilizes a sequential two-stage granulation kernel to describe the distribution of output particles from the drum granulator. The model accurately predicts the shapes of the granule size distributions over the full range of data.

In the population balance model, it is assumed that

1). Particles are present in sufficiently large concentration for the population density to be treated as a continuous function.

2). Particles are present in sufficiently low concentration to ensure all collisions are binary in nature.

3). The total volume of two agglomerating particles in a system is conserved.

Adetayo (1993) has shown that the model is suitable for use in the dynamic simulation of the granulation circuit where both moisture content and recycle size distribution may vary significantly with time.

From their experimental results the model could accurately predict the shape of the granule size distribution and also takes account of the effect of kinetics on the equilibrium size distribution. Thus Adetayo's model has been adopted for this work.

The details of Adetayo's model will be given in the next section of this chapter.

3.1.2 The Screen Model

Figure 1.2 illustrates the complete circuit of granulation in an actual fertilizer plant which requires other unit operations to complete the recycle function of the granulated fertilizer stream.

Due to the close interactions between the unit operations in the granulation circuit, development and validation of each unit operation model is an essential prerequisite to developing a reliable simulation of the granulation circuit .

Adetayo et al, (1993) mentions that the dynamics of the granulation circuit are controlled by the kinetics of the drier and the drum granulator. The kinetics of the screen and crusher are relatively fast and therefore has insignificant effect on the circuit dynamics. A steady state model of the screen and crusher is sufficient.

The screen model of Whiten (1972) is adopted by Adetayo (1993). The model is based on the probability that a particle will pass through the aperture of the screen. The present work has adopted the same model. For the sake of brevity, a summary of the screen model is presented below.

3.1.2.1 Screen model as applied to the granulation process

(Adetayo et al, (1993))

Consider the size distribution of particles going through a screen of aperture h_s and mesh diameter w_s. The mass flowrate of particles in the ith size interval coming out as oversize materials, O_i is given by:

$$O_i = F_i y_{di} \tag{3.1}$$

Where F_i = mass flowrate of the particles in the ith size interval entering the screen as feed materials.

O_i = mass flowrate of the oversize material

y_{di} = probability that a particle in the ith size interval will not pass through the screen.

Here

$$y_{di} = \begin{cases} 1 & d_{i-1} \geq h_s \\ ar + \left[\dfrac{0.89 - g(y_{i-1})}{r_s(h_s - d_{i-1})}\right][1 - ar] & d_{i-1} < h_s \text{ and } d_i > h_s \\ \dfrac{1}{r_s(d_i - d_{i-1})}[g(y_i) - g(y_{i-1})] & d_i \leq h_s \end{cases} \tag{3.2}$$

where d_i is the top size of particles in the ith size interval, and

$$y_i = r_s(h_s - d_i) \tag{3.3}$$

$$r_s = \frac{\sqrt{m}}{h_s + w_s} \tag{3.4}$$

$$g(y_i) = \int_{y_i}^{\infty} e^{-y^2} dy = \frac{0.124734}{y^3 - 0.4378805y^2 + 0.266982y + 0.138475} \tag{3.5}$$

m is an adjustable constant, and:

$$ar = \frac{[d_{(i)} - h_s]}{[d_{(i)} - d_{(i-1)}]} \tag{3.6}$$

Here we define m_o = parameter for the oversize screen

m_p = parameter for the product screen

The screening unit is composed of two sets of screens; the oversize and the product screen. The feed to the product screen is taken as the undersize of the oversize screen.

3.1.3 The Crusher Model

Crusher and grinding mill models in the literature can be classified by the modeling approaches into three groups: matrix model, kinetic model and perfect mixing model.

Detailed analysis of each modeling approach is given elsewhere (Lynch, 1977). The crusher model adopted for use in this work is a matrix model.

The hammer crusher model is composed of the breakage and the classification zones.

In the breakage zone, particles selected for breakage are broken by the hammers. The exit size distribution from the breakage zone enters the classification zone where a certain fraction of it is retained in the crusher to mix with incoming feed and thus re-enter the breakage zone. The rest comes out as the crusher product. The proposed breakage event is given by Lynch (1997) as follows :

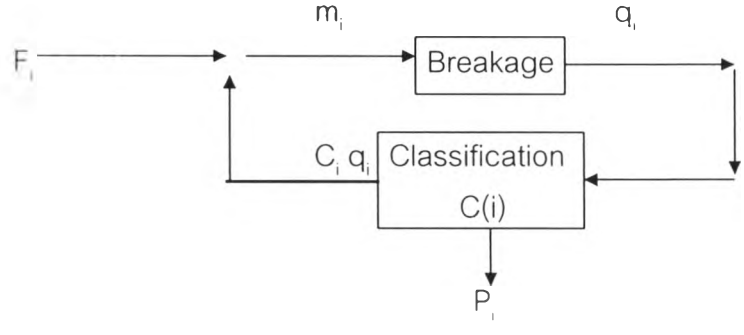

Figure 3.1 Schematic diagram of the crusher model

In the hammer crusher model, the following assumptions are made ;

1). Single fracture breakage occurs.

2). The breakage function is assumed to be time-invariant within a size interval irrespective of whether the particle is selected for breakage early or later in the grinding process.

3). The system is assumed to be perfectly mixed.

An overall mass balance of the crusher over size interval i for a steady state breakage process is as follows :

$$OUTLET = INLET + BIRTH - DEATH \tag{3.7}$$

Output is the broken particles which should pass the classified zone to become output.

Input means the feed of oversize particles to undergo breaking in the breakage zone.

Birth in the ith size interval, B_i^c occurs as a result of particles in the j^{th} size interval (j = 1, to i - 1) being broken into particles in the i^{th} size interval.

$$B_i^c = \sum_{j=1}^{i-1} B_{ij} m_j S_j \tag{3.8}$$

Breakage Function B_{ij} : The probability of particles in the j^{th} size fraction being broken into particles less than the top size in the i^{th} size interval.

Death in size interval i, D_i^c, occurs as a result of particles in that size function being broken to particles less than the i^{th} size interval

$$D_i^c = m_i S_i \tag{3.9}$$

3.1.3.1 Crusher model as applied to the granulation process

In the general model for the hammer crusher, the flow rate of particles in the ith size fraction exiting the crusher is given by

$$P_i(1 + \frac{C_i S_i}{1 - C_i}) = (1 - S_i) f_i + \sum_{j=1}^{i-1} B_{ij} S_j (f_j + \frac{C_j P_j}{1 - C_j}) \tag{3.10}$$

where f_i = feed flowrate of particles in the i^{th} size fraction entering the crusher (kg/hr)

Breakage Function

B_{ij} = Breakage function which defines the probability of a particle in the j^{th} size fraction being broken into particles less than the top size in the i^{th} size interval.

For fertilizer particles, the breakage function, should be a function that emphasises the coarse size region of the distribution (Garside and Wildsmith, 1969). The Gaudin-Meloy function (Gaudin and Meloy, 1962) has been found to give a good fit to the data. This is consistent with the observation of Garside and Wildsmith (1969). The Gaudin-Meloy equation is given by,

$$B_{ij} = 1 - \left(1 - \frac{d_i}{d_j}\right)^{N_b} \qquad (3.11)$$

where N_b is the fitting parameter for the breakage function

Selection Function

S_i = Selection function representing the probability that a particle in the i^{th} size interval is selected for breakage.

The selection function assumes that particles below a certain size, d^s_{low}, are not selected for breakage. For these particles, the selection function is taken as zero. Above a bigger size, d^s_{upp}, all particles are assumed to be selected for breakage, I.e. $S_i = 1$

$$S_i = \begin{cases} 1 & \overline{d_i} \geq d^s_{upp} \\ 1-\left[\dfrac{d^s_{upp}-\overline{d_i}}{d^s_{upp}-d^s_{low}}\right]^{Ns} & \\ 0 & \overline{d_i} \leq d^s_{low} \end{cases} \qquad (3.12)$$

where, $\overline{d_i}$ is the average size of particles in the i[th] size interval.

d^s_{upp} = particles bigger than the limiting size selected for breakage (mm).

d^s_{low} = particles smaller than the limiting size not selected for breakage (mm).

N_i is an empirical parameter in the selection function.

Classification function

C_i = Classification function representing the fraction of size (i) in the product from the breakage zone. The classification zone assumed that some of the broken particles might have to pass through the breakage zone more than once. This is because these particles are too big to exit the crusher due to material segregation in the crusher.

$$C_i = \begin{cases} 0.999 & \overline{d_i} \geq d^c_{upp} \\ 1-\left[\dfrac{d^c_{upp}-\overline{d_i}}{d^c_{upp}-d^c_{low}}\right]^{Nc} & \\ 0 & \overline{d_i} \leq d^c_{low} \end{cases} \qquad (3.13)$$

where d^c_{upp} = upper critical size above which bigger particles are returned back to the breakage zone.

d^c_{low} = lower critical size below which smaller particles are not returned for re-breakage.

N_c = an empirical parameter for the classification function.

To avoid numerical problem, $0 \leq C_i \leq 0.999$

3.2 Population Balance Model for Drum Granulator

The laboratory scale experiments were carried out by Adetayo(1993) to understand the fundamental mechanisms of fertilizer granule formation. He then carried out a review of the most commonly used modeling techniques for the granulating system in order to establish a suitable model for use in modeling the drum granulator.

He concludes that the population balance modeling technique is the most suitable of all the modeling techniques reviewed. The population balance model has the ability of following the kinetics of the granulation process. After a systematic comparison of the various numerical approximations to the corresponding known analytical solutions of the population balance equation, the Hounslow's sectional mid-point model (Hounslow et al.,1988) is found to be adequate for use in the development of a suitable model for the fertilizer drum granulator.

3.2.1 The Population Balance Equation

For a particulate system, Hounslow (1990) defines the population balance as a statement of continuity that describes how the particle-size distribution may change with time and position. A review of the various application areas of the population balance is given by Ramkrisahna (1985).

A general population balance equation (GPBE) is developed by carrying out a balance over the population density in a fixed subregion of the particle-phase space.

Accumulation = Input- Output + Birth + Death (3.17)

Randolph and Larson (1988) present the general form of the PBE as;

$$\frac{\partial n}{\partial t} + \nabla.(un) - B + D = 0 \quad (3.18)$$

where n is the number density function , u is the velocity vector of the particulates phase, B and D are the birth and death terms, respectively.

The population balance for a well mixed system undergoing coalescence alone and using the internal coordinate is given by

$$\frac{\partial n(v,t)}{\partial t} = \frac{1}{N_t^{\alpha}} \int_0^{\infty} \beta(v,u) n(u,t) n(v,t) du + \frac{1}{2N_t^{\alpha}} \int_0^{v} \beta(v-u,u) n(u,t) n(v,t) du \quad (3.19)$$

where

$n_{(v,t)}$ = number density function given as the number of particles at time t per unit volume of system.

β = coalescence rate kernel

N_t = total number of particle at time t

α = 0, where for free-in-space system (aerosol)

= 1, where for restricted-in-space system (granulation process)

u, v = suitable internal coordinates

Advantage of the population balance model.

Due to the dependence of the coalescence rate constants on the process variables, the population balance model could be effectively used to account for the effect of these variables on the process. The population balance model also gives information on the dynamics of the system.

The solution to this integro-differential equation is not a trivial matter. Known analytical solutions are only available for special forms of the coalescence kernel with an assumed initial number density distribution (Ramkrishna., 1985). Numerical solutions to this equation have been obtained by various methods : moment (Hulburt and Katz.,1964), discrete (Landgrebe and Pratsinis, 1989,1990), sectional (Gelbard, Tambour and Seinfield.,1980) and sectional-midpoint (Hounslow .,1988) method.

3.2.2 Hounslow Model

Hounslow et al. (1988) develop a population balance for batch aggregation of particulate suspensions that is recast in a form that may be solved simply and accurately. The transformed equation is deduced with the introduction of only one additional parameter, which is found to be a constant for all case. The transformed equation is tested by comparision with some analytical solutions, with which it is found to be in excellent agreement. In particular, the equation is shown to predict correctly the rate of change of the total particle number and volume. Compatible descriptions of linear growth and nucleation have been developed with similar success.

Hounslow et.al.(1988) using volume as the internal coordinate discretize the system domain of interest into intervals in geometric series.

The following 4 mechanisms which affect the number of granules in the ith size interval are considered.

1). Birth of a particle in the i^{th} interval occurs as a result of a particle in the $(i-1)^{th}$ interval coalescing with another particle in one of the first to $(i-1)^{th}$ intervals only when the resultant granule is larger than the lower size limit of the i^{th} interval.

2). Birth in the i^{th} size fraction is due to coalescence of two particles in the $(i-1)^{th}$ size fraction.

3). Death of a particle in the i^{th} size fraction is due to its coalescence with another particle sufficiently large enough for the resultant granule to be larger than the upper size limit of the ith interval.

4). Death of a particle in the i^{th} size fraction is due to coalescence between that particle with another particle from that or a higher size interval.

The total rate of change of the number of particles in the i^{th} size interval is given by;

$$dN_i / dt = R_i^{[1]} + R_i^{[2]} - R_i^{[3]} - R_i^{[4]} \qquad (3.20)$$

where $R_i^{[l]}$ = rate of change of particles in the ith interval due to the $[l]th$ mechanism

N_i = number of particles in the ith size interval

From the various rates, Hounslow showed that, for a restricted in space system (drum granulator) ,

$$\frac{dN_i}{dt} = \frac{1}{N_T^{\alpha}}\left(Ni-1 \sum_{j=1}^{i-2} 2^{j-i+1} \beta_{i-1,j} N_j + \frac{1}{2}\beta_{i-1,i-1} N_{i-1}^2 - N_i \sum_{j=1}^{i-1} 2^{j-i} \beta_{i,j} N_j - N_i \sum_{j=i}^{\infty} \beta_{i,j} N_j \right) \quad (3.21)$$

where $\beta_{i,j}$ = collision rate function referred to here, as the coalescence kernel.

N_i = number of particle in the ith size interval

Houslow's model accurately predicts the total number and conserves the total volume (Hounslow, 1990)

3.3 Solution to the population balance equation

Adetayo (1993) has made a systematic comparison of the various numerical solutions (i.e., moment method, discrete method, sectional method and sectional-midpoint method) to known analytical solutions of the general population balance equation. Houslow's sectional model solution was found to be adequate for modeling the fertilizer granulation process.

3.3.1 The Coalescence Kernel

The coalescence kernel $\beta(a,b)$ plays an important role in the population balance model. It is a measure of the frequency of successful collision between two particles of volume a and b to form a particles of volume $a+b$. Sastry (1975) postulates that

The coalescence kernel is symmetrical,

$$\beta(a,b) = \beta(b,a)$$

The coalescence kernel is homogeneous.

$$\beta(a,b) = b^{\theta}\beta(a/b,b)$$

where : θ = degree of homo geneity of the kernel (order of the kernel)

The coalescence kernel can be divided into two parts

$$\beta(a,b) = \beta_o.\beta^*(a,b)$$

β_o = coalescence rate constant that for determines the rate of granulation (In other words, it controls the rate of change of the mean of the granule size distribution)

$\beta^*(a,b)$ = coalescence kernel that determines the shape of the granule size distribution

The population balance model is developed to describe the drum granulation of a feed with a broad size distribution. The solution to the population balance equation with coalescence kernel should predict the two-stage mechanism which is found in lab-scale fertilizer granulation.

3.3.1.1 The two-stage coalescence kernel

Since two stages of granulation have been identified by Adetayo et al.(1993), it is expected that a two-stage granulation kernel will be necessary to model adequately the granule size distributions over a wide range of conditions.

The first stage coalescence kernel.

In the first stage or non-inertial regime of granulation, the probability of successful coalescence following a collision is independent of the particle size and collision velocity. When the rate of collisions is assumed to be independent of particle size, the first stage mechanism becomes a random process with a size independent coalescence kernel

$$\beta_{i,j}^{(1)} = k_1 \qquad (3.22)$$

The second stage coalescence kernel

During the second stage or inertial regime of granulation, the granule size distribution begins to widen. Particle deformation is important and, therefore, collisions involving large granules are favoured due to their increased inertia upon impact. A size-dependent kernel is necessary to treat this stage of granulation.

A first-order kernel is selected to predict the second stage regime as follows (Adetayo, 1993):

$$\beta_{ij}^{(2)} = k_2 (v_i + v_j) \qquad (3.23)$$

This equation is developed by Golovin (1968), where k_2 describes the rate constant for the Golovin kernel.

The first stage of granulation (non-inertial regime) is fast, relative to the duration of Adetayo's experiments. For experiments in which the second stage of granulation does not occur, an equilibrium size distribution is quickly reached. For the fertilizers studied by Adetayo, the second stage of granulation (inertial regime),

which is slow, only occurs after coalescence in the non-inertial regime is essentially complete. Due to the differing time scales in the two growth mechanisms, he proposes a sequential kernel for both stages of granulation:

$$\beta_{i,j} = \begin{cases} \beta_{i,j}^{(1)} & t \le t_1 \\ \beta_{i,j}^{(2)} & t > t_1 \end{cases} \qquad (3.24)$$

where $\beta_{i,j}^{(1)}$ and $\beta_{i,j}^{(2)}$ are given by Eq.3.22 and Eq.3.23 , respectively, Here t_1 represents the time required to reach the final equilibrium size distribution of the first non-inertial stage of granulation. When the second stage of granulation does not occur, the population balance at $t = t_1$ gives the equilibrium granule size distribution for coalescence in the non-inertial regime as the final result for $t > t_1$.

In summary the two-stage granulation mechanism is given by:

$$\beta_{i,j} = \begin{cases} k_1 & t \le t_1 \\ k_2 (v_i + v_j) & t > t_1 \end{cases} \qquad (3.25)$$

3.3.1.2 Estimation of the coalescence rate constants

The values of k_1 and k_2 must be estimated from available experimental data, as Adetayo, (1993) has done . In the present work, these estimates of k_1 and k_2 will be used in comparing the resulting simulation results with his experimental results in order to confirm the suitability of the coded computer model.

When additional experimental data are available, for example, from National Fertilizer Company, the suitable parameteric values should be re-estimated for the case of NFC.