Chapter 6

The use of the population balance to describe growth and breakage of pellets in a high-shear mixer

Abstract

In this study, three adaptations of the population balance have been made. First of all, breakage of pellets was included in the population balance. Second the population balance was presented in mass distributions instead of a number distribution. Finally, the pellet size distribution as well as a tracer component distribution was modelled with the population balance. Population balance modelling on the growth and breakage of pellets in a high-shear mixer was performed. Two types of high-shear mixers (a coffee grinder and a Collette Gral 10) were used for the preparation of pellets. The kinetics of pellet growth was measured using tracer experiments. Population balance modelling of the continuous growth and breakage of pellets during an equilibrium stage of growth was performed. The results showed that the most important breakage mechanisms in the coffee grinder were both fragmentation and abrasion. For the Gral high-shear mixer, no distinction could be made on the basis of the chi-square test. The three modelled breakage mechanisms, fragmentation, shattering and abrasion, were supposed to occur simultaneously during pelletisation. Whereas each breakage mechanism occurs at a different location within the mixing bowl. It is most likely that abrasion occurs due to pellet-pellet and pellet-wall collisions, and takes place inside the torus and at the wall, respectively. Fragmentation and shattering are likely to occur at the impeller and chopper site of the bowl due to pellet-impeller and pellet-chopper collisions. The population balance showed to be a useful tool for describing the pellet size distribution and the tracer concentration distribution during the high-shear pelletisation process. A physical interpretation of the fitted parameters was made, based on the total power input during processing and a collision frequency of the pellets.
6.1. Introduction

6.1.1. Mechanisms of granule growth

The mechanisms of growth and breakage of pellets or granules have been thoroughly investigated by several authors\textsuperscript{1-3}. Focussing on the pelletisation process, the mechanisms of growth can be described as a nucleation stage, followed by a simultaneously acting combination of coalescence, layering and breakage. 

\textit{Nucleation} is the process where fine wet particles stick together, or where a droplet engulfs a relative large amount of fine particles. \textit{Coalescence} occurs when two or more granules collide and form one new larger granule. During \textit{layering}, fine materials (primary particles or small granules) adhere to the surface of larger granules.

In fact, coalescence and layering are based on similar principles. The only difference between these two mechanisms is the size of the growing particles. If the particles are almost of the same size, the mechanism is called coalescence. If primary particles or small granules stick to the surface of much larger granules, the mechanism is called layering.

The growth and break-up of pellets, made in a high-shear mixer, was experimentally investigated as described in chapter 2 and 3 of this thesis. In this chapter, the population balance modelling is used to describe the growth and breakage of the pellets in a high-shear mixer.

6.1.2. Mechanisms of granule breakage

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure6.1.png}
\caption{The three selected breakage mechanisms and break-up distributions of the high-shear pelletisation process. Here the situation for breakage towards two size classes is given ($x = 2$).}
\end{figure}
Several authors have also described breakage of granules. Kapur described the transition region of pellet growth in a rotating drum, where small particles are crushed and layered on larger particles. This crushing and layering results in a size dependent growth of the pellets. After this transition region the ball growth region is entered, in which abrasion was one of the most important net growth mechanisms. Kapur also mentioned a steady-state situation in which the mean particle size of the agglomerates remains constant because the processes of crushing and layering are balanced. During this steady-state situation, crushing and layering are supposed to occur only when the agglomerates are brittle and relatively weak.

Newitt et al. and Sastry et al. used tracer experiments to investigate the mechanisms of pellet growth and breakage in a rotating drum. The most important breakage mechanism was described as the crushing action of the larger granules, resulting in breakage of the small granules, which were layered on the surfaces of the larger ones.

Holm et al. described growth of pellets made from a mixture of lactose and microcrystalline cellulose in a roto-granulator. Growth of pellets occurred until an equilibrium was reached, where the disc speed and the control of the moisture content were recognised as the most important process parameters.

All descriptions of granule growth and breakage are related to different processes (e.g. rotating drum or Lödige) and products (pharmaceutics, sand, glass ballotini or salts) and therefore can not be compared with each other. The differences in processes and product used have a considerable effect on the mechanisms of growth. Comparing the rotating drums with the high-shear mixers (like Gral, Diosna and Fielder), the shear forces in the rotating drum are low compared to the high-shear mixer. This results in relatively more growth than breakage in the rotating drum. To model net pellet growth one has to include growth as well as breakage, in particular in high-shear mixers.

Figure 6.2. Schematic representation of the breakage mechanisms from the point of view of size class $i$. Key: (—) fragmentation, (---) shattering, and (···) abrasion.
The most commonly found breakage mechanisms in literature were used in our population balance modelling. The three different breakage mechanisms are fragmentation, shattering and abrasion (see figure 6.1 and figure 6.2). For the description of the breakage mechanisms, the particle size distribution of a batch of pellets is divided into \( n \) size classes. The smallest pellets belong to size class 1, the largest to size class \( n \). Growth and breakage will be described from a point of view of a particle from size class \( i \) \( (1 \leq i \leq n) \).

**Fragmentation** is the process in which the original pellet (or granule) breaks into relatively large fragments. In other words, breakage by fragmentation occurs from one pellet of size class \( i \) breaks into several particles of size classes \( i-1, i-2, \ldots i-x \).

**Shattering** is the process of total breakage of the original pellet into small particles without leaving any large fragments, which represent the crushing of small pellets as described by Newitt et al.\(^2\) and Sastry et al.\(^3\) The size of the particles resulting from shattering are much smaller compared to the fragmentation process: no large fragments are obtained, only numerous very little particles. Theoretically, shattering means that the original pellets breaks into primary particles all belonging to the smallest size class\(^3\). In practise, it is hard to imagine that all liquid bridges between the primary particles are broken during shattering. It should be expected that the newly formed particles belong to more than one size class. The number of size classes to which breakage occurs is called the breakage interval \( (x) \). Thus breakage by shattering occurs from one pellet of size class \( i \) towards size classes \( x, x-1, \ldots 2, 1 \).

**Abrasion** (also called attrition) is the process in which the outer shell of the pellet wears at the surface. This results in one large pellet and a number of very little particles (of the first few size classes). Abrasion of one pellet from size class \( i \) results in one pellet of size class \( i-1 \) and some very little particles of size class 1, 2, \ldots 1.

6.1.3. Previous experiments

The growth and break-up of pellets, made in a high-shear mixer, was experimentally investigated as described in chapter 2 and 3. Tracer experiments, performed in a Gral 10 high-shear mixer and a coffee grinder, were used to investigate the growth and breakage behaviour of pellets during an equilibrium stage of pellet growth. By adding a small amount of coloured pellets (= tracer) in a specific size class, the distribution of the colour over the different size classes could be measured as a function of time. It was found that within a short time-period, a homogenous distribution of the colour is obtained, and that growth and breakage occur simultaneously during the equilibrium stage of growth in a high-shear mixer.

6.1.4. Aim

The aim of the present study was to find an application of the population balance to the earlier results obtained with the tracer experiments. In the present study population balance
modelling is used to describe the growth and breakage of pellets in a high-shear mixer. Further an attempt is made to find a physical interpretation of the fitting parameters.

6.2. The Population Balance

Although a substantial amount of population balance modelling has been published in literature\(^5^\)\(^-^\)\(^9^\), none of these publications include breakage of granules during granulation. This is rather remarkable, especially for the modelling of granule growth in high-shear mixers, where the relatively large impact forces of the impeller arms and choppers cause breakage of granules. It was therefore decided to adapt the population balance of Hounslow\(^1^0^\) and extend it by including the breakage of granules. This population balance model intends to describe the following aspects:

- dynamics of the particle size distribution, including growth and breakage of particles;
- dynamics of the distribution of a tracer component between the particles;
- presentation of the data in mass fraction.

6.2.1. Definitions

6.2.1.1. Definition of growth, breakage, birth, and death

Four different terms are often used with regard to population balances: growth, breakage, birth, and death. Because the model includes growth mechanisms and break-up mechanisms, the above mentioned four terms may be confusing. Therefore, the definitions of these terms are given next (see figure 6.3).

![Figure 6.3. Schematic representation of growth, breakage, birth and death for size class i.](image)

*Growth* describes all processes resulting in an increase of the particle size. As mentioned before, the mechanisms involved are coalescence and layering. *Breakage* describes all
processes resulting in a decrease of the particle size. The mechanisms involved are fragmentation, shattering, and abrasion. Birth describes all processes resulting in an increase of the amount (mass fraction) of material in a certain size class. Death describes all processes resulting in a decrease of the amount of material in a certain size class.

### 6.2.1.2. Definition of the size classes

Growth plays an important role in the description of the birth and death of a size class. Growth occurs due to layering and coalescence of pellets. Although the mechanisms are physically different, layering and coalescence can mathematically be described in the same way. Both mechanisms can be seen as the combining of two pellets of size classes \( i \) and \( j \) (with masses of \( m_i \) and \( m_j \), and for \( i \geq j \)), resulting in a new pellet of total mass \( (m_i + m_j) \). Because a discrete description of the pellet size distribution is used instead of a continuous description, the newly formed pellet will have a mass that does not fit exactly within a certain size class. A part of the mass of the newly formed pellet will be donated to size class \( i \), and another part will be donated to size class \( i+1 \). To minimise the amount of size classes receiving the newly formed pellet, the following limitation has been applied to the model. The ratio of the pellet masses of two adjacent size classes has been set at two:

\[
\frac{m_{i+1}}{m_i} = 2
\]  

(6.1)

The mass of a pellet from size class \( i \) is \( 2^i m_0 \) (where \( m_0 \) is the mass of one pellet from a fictive size class 0, which is introduced in order to normalise the masses of the pellets). Coalescence of two pellets of size classes \( i \) and \( j \) (for \( i \geq j \)) results in a pellet of size class \( i+1 \).

### 6.2.2. Population balance

In the population balance, we both include the pellet size distribution as well as the tracer concentration. Population balances are normally based on a number distribution, where the number of pellets in the smallest size classes has a great influence in the development of the growth. The population balance according to Hounslow et al.\(^{10}\) is given by:

\[
\frac{dN_i}{dt} = \sum_{j=1}^{i-1} 2^{i-j} \beta_{i-1,j} N_j N_{i-1} + \frac{1}{2} \beta_{i-1,i-1} N_{i-1}^2 - \sum_{j=1}^{i} 2^{i-j} \beta_{i,j} N_j N_i - \sum_{j=i}^{\infty} \beta_{i,j} N_j N_i
\]  

(6.2)

Where \( N_i \) represent the number of particles in size class \( i \), \( dN_i/dt \) is the change of the number of particles from size class \( i \) as a function of time, and \( \beta \) is the coalescence kernel.

Because the masses of the pellets from the smallest size classes are relatively low, their influence on the growth and breakage of granules is not as large as expected with the number distribution. To overcome this problem the data are presented - as experimentally measured - in mass densities, which requires a different representation. Therefore, we use a population
balance that is principally based on numbers. But, for the sake of convenience, the numbers are represented by the total mass per size class, divided by the average mass of the particles in that size class. In this way, the problem of the zeroth moment of the balance is avoided, and the convenience of the mass distributions (as experimentally measured) can be used.

The density function of pellets in a specific size class is defined as the number of pellets in a size class \( N_i \) divided by the volume of one pellet from this specific size class \( v_i \). The volume of one pellet is equal to \( 2^i v_0 \) (where \( v_0 \) stands for the volume of one pellet from a fictive size class 0, which is introduced in order to normalise the pellet volume).

\[
\rho_i = \frac{N_i}{v_i} = \frac{N_i}{2^i v_0}
\]

(6.3)

The mean volume of pellets from size class \( i \) can be calculated with:

\[
V_i = \frac{\int 2^i v_0 n(v) dv}{N_i} = v_0 \frac{2^{2i+2} - 2^{2i}}{2} = \frac{3}{2} 2^i v_0
\]

(6.4)

Here, \( V_i \) is the total volume of pellets from size class \( i \). The undersize of the size class is indicated with \( i \), and the mass ratio of two adjacent size classes is equal to 2 (eq. 6.1). For the estimation of the mean volume of a pellet of size class \( i \), one has to introduce the factor 3/2.

The mass fraction of size class \( i \) (\( \omega_i \)) depends on the mass of one pellet in the size class \( (2^i m_0) \), the number of pellets in the size class \( (N_i) \), the total mass \( (m_{tot}) \), and the density of the pellets \( (\rho_p) \). The fractional mass of size class \( i \) is given by:

\[
\omega_i = \frac{V_i \cdot \rho_p \cdot N_i}{m_{tot}} = \frac{3}{2} \frac{2^i v_0 \rho_p N_i}{2 m_{tot}} = \frac{3}{2} \frac{2^i m_0 N_i}{m_{tot}}
\]

(6.5)

In terms of mass fractions, using eq. 6.5, Hounslow’s population balance can be rewritten as:

\[
\frac{d\omega_i}{dt} = \sum_{j=0}^{i} \frac{4}{3} \frac{m_{tot}}{2^{i-j} m_0} \beta_{i-j} \omega_j \omega_{i-j} + \frac{2}{3} \frac{m_{tot}}{2^{i-j} m_0} \beta_{i-j} \omega_j \omega_{i-j}^2
\]

\[
- \sum_{j=0}^{i} \frac{2}{3} \frac{m_{tot}}{2^{i-j} m_0} \beta_{i-j} \omega_j \omega_i - \sum_{j=0}^{i} \frac{2}{3} \frac{m_{tot}}{2^{i-j} m_0} \beta_{i-j} \omega_j \omega_i \omega_j
\]

(6.6)

The colour concentration distribution can be written similar to the mass distribution (eq. 6.6), but now the amount of colour in a size class \( (C_i \omega_i) \) must be used instead of the fractional mass of the size class \( (\omega_i) \):

\[
\frac{dC_i \omega_i}{dt} = \sum_{j=0}^{i} \frac{4}{3} \frac{m_{tot}}{2^{i-j} m_0} \beta_{i-j} C_j \omega_j C_i \omega_{i-j} + \frac{2}{3} \frac{m_{tot}}{2^{i-j} m_0} \beta_{i-j} C_j \omega_j C_i \omega_{i-j}^2
\]

\[
- \sum_{j=0}^{i} \frac{2}{3} \frac{m_{tot}}{2^{i-j} m_0} \beta_{i-j} C_j \omega_j C_i \omega_i - \sum_{j=0}^{i} \frac{2}{3} \frac{m_{tot}}{2^{i-j} m_0} \beta_{i-j} C_j \omega_j C_i \omega_i \omega_j
\]

(6.7)
6.2.2.1. Description of breakage

Breakage of a pellet of size class \( i \) is described as the transport of mass from size class \( i \) towards one or more smaller size classes. The changes of mass and volume from size class \( i \) towards size class \( j \) are defined as \( \Delta m_{i,j} \) and \( \Delta V_{i,j} \) respectively. Here, the donor size class is given before the comma, and the receiver size class after the comma.

Breakage generally occurs towards more than one size class, therefore the number of size classes to which breakage occurs is given by \( x \). Breakage of a pellet from size class \( i \) is modelled according to an equal density distribution towards the receiving size classes as schematically given in figure 6.4.

Figure 6.4. Equal density distribution for the breakage from size class \( i \) towards size classes \( j \) and \( j+1 \).

Take for example a pellet from size class \( i \) breaking into fragments with sizes of size classes \( j \), \( j+1 \), and \( j+2 \). The particle density distribution has a constant value in this interval. The volume of a pellet from size class \( j \) ranges between the undersize of the size class (volume \( 2^j v_0 \)) to the upper size of the size class (volume \( 2^{j+1} v_0 \)). The volume of the broken particles coming into size classes \( j \) to \( j+x \) (\( \Delta V_{i,j} \)) is given by:

\[
\Delta V_{i,j} = \int_{2^j v_0}^{2^{j+x} v_0} n(v) dv = \frac{1}{2} v_0^2 n(v) \left(2^{2j+x} - 2^{2j}\right) = \frac{1}{2} v_0^2 n(v) \cdot 2^{2j} \left(2^x - 1\right) \tag{6.8}
\]

Using the discretised scheme of Hounslow, the volume of the broken particles from the donor size class \( i \) (\( \Delta V_i \)) can be obtained from eq. 6.4 and is given by:

\[
\Delta V_i = \frac{3}{2} 2^j v_0 N_i \tag{6.9}
\]

Because \( \Delta V_{i,j} = \Delta V_i \), the change of the density function due to breakage can be estimated using eq. 6.8 and 6.9:

\[
n(v) = \frac{3 \cdot 2^j v_0}{2^{2j} \cdot (2^x - 1)} \cdot \frac{N_i}{v_0} \tag{6.10}
\]
The number of pellets of size class \( j+k \), after breakage of a pellet from size class \( i \) into size class \( j+k \) \((0 \leq k < x)\), is given by:

\[
N_{j+k} = 2^{j+k} v_0 n(v) = \frac{3 \cdot 2^i \cdot 2^{2k}}{2^{j+k} \cdot (2^{2i} - 1)} \cdot N_i
\]  
(6.11)

Here \( j \) is the first size class to which breakage occurs, \( x \) is the total number of size classes to which breakage occurs.

The following estimation of the breakage distribution can be used in case of fragmentation (breakage to size classes \( i-1, i-2 \ldots i-x \)), and in case of shattering (breakage to size classes 1, 2 \ldots \( x \)). The breakage distribution of abrasion can be determined similarly.

In general, the volume of particles transferring from size class \( i \) to size class \( j \) (which can be a function of \( x \)) is given by:

\[
\Delta V_{i,j(x)} = \frac{3}{2} \sum_{k=0}^{x-1} 2^{j+k} v_0 \cdot N_{j+k} = \frac{3}{2} 2^{2j} n(v) \cdot v_0^2 \cdot \sum_{k=0}^{x-1} 2^{2k}
\]  
(6.12)

In case of fragmentation, \( j \) is equal to \( i-x \), and in case of shattering, \( j \) is equal to 1.

The volume of particles transferring from size class \( i \) to a specific size class \( j+k \), after substitution of eq. 6.10, is given by:

\[
\Delta V_{i,j+k} = \frac{3}{2} 2^{2j} 2^{2k} n(v) \cdot v_0^2 = \frac{3}{2} \cdot \frac{2^i 2^{2k}}{2^{2i} - 1} v_0 N_i
\]  
(6.13)

The increase of the mass fraction of a specific size class \( j+k \) due to breakage of a pellet from size class \( i \) can be written as a function of \( \omega_i \) using eq. 6.5 and eq. 6.13:

\[
\omega_{j+k} = \frac{3 \cdot 2^{2k}}{2^{2i} - 1} \cdot \omega_i
\]  
(6.14)

The rate of change of mass from this particulate size class \((j+k)\) is given by:

\[
\frac{d\omega_{i,j+k}}{dt} = \alpha_i \cdot \frac{3 \cdot 2^{2k}}{2^{2i} - 1} \cdot \omega_i = \alpha_i \cdot f(i, j) \cdot \omega_j
\]  
(6.15)

Here, \( \alpha_i \) is the breakage kernel, which is independent of the pellet size as explained in further detail in the appendix. The break-up distribution function towards \( x \) size classes is represented by \( f(i, j)\) and given by:

\[
f(i, j) = \sum_{k=0}^{x-1} \frac{3 \cdot 2^{2k}}{2^{2i} - 1}
\]  
(6.16)

Many different break-up distributions can be used to describe breakage in population balance modelling. Fragmentation, shattering and abrasion are the three most commonly found break-up mechanisms in literature. Figure 6.1 gives a schematic representation of these three mechanisms. The break-up mechanisms and the corresponding break-up distribution functions are given in table 6.1.
Table 6.1. The three breakage distribution function.

<table>
<thead>
<tr>
<th>breakage mechanism</th>
<th>birth into size class</th>
<th>breakage distribution function</th>
</tr>
</thead>
<tbody>
<tr>
<td>fragmentation</td>
<td>i-1, i-2, ..., i-x</td>
<td>( f(i,i-x+k) = \sum_{k=0}^{i-1} \frac{2^{2k}}{2^{2x+1}} )</td>
</tr>
<tr>
<td>shattering</td>
<td>1, 2, ..., x</td>
<td>( f(i+1+k) = \sum_{k=0}^{x-1} \frac{2^{2k}}{2^{2x+1}} )</td>
</tr>
<tr>
<td>abrasion</td>
<td>1, 2, ..., x &amp; i-1</td>
<td>( f(i+1+k &amp; i-1) = \sum_{k=0}^{x-1} \frac{2^{2(k+1)}}{2^{2x+1}} + \frac{2^{2x+1}}{2^{2x+1}} )</td>
</tr>
</tbody>
</table>

6.2.2.2. Overall population balance

The breakage function as given in eq. 6.15 can be introduced in the population balance of Hounslow et al. The overall population balance including the breakage of pellets is given by:

\[
\frac{d\omega_i}{dt} = \sum_{j=1}^{i} \frac{4}{2^{i-1}} \frac{m_{tot}}{m_0} \beta_{i-1,j} \omega_j \omega_{i-1} + \frac{2}{3} \frac{m_{tot}}{2^{i-1} m_0} \beta_{i-1,i-1} \omega_{i-1}^2 \\
- \sum_{j=1}^{i-1} \frac{2}{3} \frac{m_{tot}}{2^i m_0} \beta_{i,j} \omega_j \omega_i - \sum_{j=1}^{i} \frac{2}{3} \frac{m_{tot}}{2^i m_0} \beta_{i,j} \omega_j \omega_i - \alpha_i \omega_i f(i,j)
\]  

(6.17)

This equation is used in our population balance modelling. Different breakage mechanisms, as given in table 6.1, were used during modelling. The function \( f(i,j) \) of equation 6.17 can be replaced by one of the breakage distribution functions as given in table 6.1. During modelling, optimisations of the over all coalescence kernel (\( \beta_0 = \beta_{i-1,j} = \beta_{i-1,i-1} = \beta_{i,i} \)) as well as the breakage kernel (\( \alpha_0 \)) were made.

The colour concentration distribution can be written similar to the mass distribution (eq. 6.17). The amount of colour in a size class (= \( C_i \omega_i \)) must be used instead of the fractional mass of the size class (\( \omega_i \)):

\[
\frac{dC_i \omega_i}{dt} = \sum_{j=1}^{i} \frac{4}{2^{i-1}} \frac{m_{tot}}{m_0} \beta_{i-1,j} C_j \omega_j C_i \omega_{i-1} + \frac{2}{3} \frac{m_{tot}}{2^{i-1} m_0} \beta_{i-1,i-1} C_i \omega_i^2 \\
- \sum_{j=1}^{i-1} \frac{2}{3} \frac{m_{tot}}{2^i m_0} \beta_{i,j} C_j \omega_j C_i \omega_i - \sum_{j=1}^{i} \frac{2}{3} \frac{m_{tot}}{2^i m_0} \beta_{i,j} C_j \omega_j C_i \omega_i - \alpha_i C_i \omega_i f(i,j)
\]  

(6.18)

6.3. Experimental set-up

Pellets were made from an equal mixture of microcrystalline cellulose (Pharmacel PH101, DMV Veghel, The Netherlands) and \( \alpha \)-lactose monohydrate (Pharmatose 200 mesh, DMV Veghel, The Netherlands). Pellets were prepared in a coffee grinder at two different impeller speeds (Moulinex 980, Ireland), and in a Gral 10 high-shear mixer at one impeller speed, with the chopper turned-off (Machines Collette, Wommelgem, Belgium). Demineralised water was
used as the binder liquid. The equilibrium stage of growth was reached after about 1 minute mixing in the coffee grinder, and 15 minutes of mixing in the Gral. During the equilibrium stage of growth, tracer-experiments with amaranth as colouring agent were performed. A part of the (uncoloured) batch of wet pellets was replaced by a known sieve fraction of coloured pellets of the same composition (apart from the tracer component). The distribution of the tracer among all sieve fractions was measured spectrophotometrically at varying mixing times. A detailed description of the experiments is given in a chapter 3. The diameter of the bowl and the impeller tip speed \( v_{\text{tip}} = \pi ND \) are given in table 6.2.

<table>
<thead>
<tr>
<th>process &amp; impeller speed</th>
<th>diameter impeller (cm)</th>
<th>tip speed impeller (m/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>gral impeller 6.7 rps</td>
<td>24.5</td>
<td>5</td>
</tr>
<tr>
<td>coffee grinder 82 rps</td>
<td>8.0</td>
<td>19</td>
</tr>
<tr>
<td>coffee grinder 187 rps</td>
<td>8.0</td>
<td>44</td>
</tr>
</tbody>
</table>

Porosity changes during processing are not included in our model. Equal porosities of all pellets in all size classes are assumed in the population balance. It was experimentally shown in chapter 2 that the porosity of the pellets is very low during the equilibrium stage of growth (about 10%), and that minor densification of the already dense pellets occurs during the equilibrium stage of growth.

6.3.1. Population balance modelling

Only the equilibrium stage of pellet growth was modelled, with a non-changing pellet size distribution and fast changing colour concentrations. The number of size classes (18) was chosen on the basis of the experimental sieving results. The experimental results (sieving results and amount of colour in a size class) are adapted in order to obtain a \( \sqrt{2} \) relationship. The breakage interval \( (x = \text{number of size classes to which breakage occurs}) \) was either 2 or 3. Because minor effect of the breakage interval on the outcome of the model was found, no further values of the breakage interval were modelled.

For the modelling, the breakage mechanisms fragmentation, shattering and abrasion were calculated separately for the three different process conditions (coffee grinder low and high impeller speed and Gral experiment).

The breakage distribution function \( f(i,j) \) of equation 6.17 was replaced by one of the breakage distribution functions as given in table 6.1. The over-all coalescence kernel \( (\beta_0) \) is defined as: \( \beta_0 = \beta_{i-1,j} = \beta_{i-1,i-1} = \beta_{i,j} \). The breakage kernel \( (\alpha_i) \) is a size independent value (see appendix to chapter 6).
For this reason, the over all breakage kernel $\alpha_0 = \alpha_i$ was used. During modelling, optimisations of the over all coalescence kernel ($\beta_0$) as well as the over all breakage kernel ($\alpha_0$) were made, based on the pellet size distribution and the colour distribution. The goodness of fit was expressed as the residual sum of squares (SS$_{\text{res}}$) and was calculated for the pellet size distribution as well as the colour distribution by the sum of the square of the differences between the experimental values and the outcomes of the model divided by the experimental values:

$$SS_{\text{res}} = \sum \left( \frac{\text{experiment} - \text{model}}{\text{experiment}} \right)^2$$

(6.19)

A summation of the SS$_{\text{res}}$ of the mass distribution and the colour distribution results in the total sum of squares. Optimisation of $\beta_0$ (over all coalescence kernel) and $\alpha_0$ (over all breakage kernel) was performed towards a minimal total sum of square value.

The difference between the modelled values and the experimental values was calculated with the chi-square test. Barlett’s chi-square test on a system of models was used to compare the three breakage mechanisms. By comparing the calculated sum of square values (3 breakage mechanisms, 5% uncertainty), discrimination between the three breakage mechanisms within each apparatus was tried to be made.

### 6.4. Modelling results

#### 6.4.1. Results

In table 6.3, the modelling results for the three selected breakage mechanisms of each experiment (both coffee grinder experiments and Gral experiment) are given in case of a breakage interval ($\alpha$) of 2. The modelling results with a breakage interval of 3 showed similar results. Therefore, the results of a breakage interval of 3 are not presented.

From the residual sum of square values (SS$_{\text{res}}$, see table 6.3), the following information is obtained. According to the SS$_{\text{res}}$-values found for shattering in both coffee grinder experiments, shattering is excluded as a possible breakage mechanism for the coffee grinder experiments. For both coffee grinder experiments, low and high impeller speed, the most important breakage mechanisms are therefore abrasion and fragmentation.

On the basis of the SS$_{\text{res}}$-values, no distinction between the three breakage mechanisms could be made for the Gral experiment. So, all modelled breakage mechanisms are important at the high-shear pelletisation process in the Gral.

Generally, high coalescence kernels are found for modelling results for the high impeller speed experiment in the coffee grinder. Furthermore, high breakage kernels are also found for the same experiment. This implies fast growth as well as fast breakage of pellets for the high impeller speed experiment. This is confirmed by the results presented in chapter 3, where a...
rapid homogeneous colour distribution over all pellets was found during a coffee grinder experiment at high impeller speed. 

Because a high coalescence kernel often correlates with a high breakage kernel, the ratio of the coalescence kernel and the breakage kernel is also given in table 6.3. The coalescence/breakage ratio for fragmentation is somewhat higher compared to the ratio for abrasion, whereas the highest ratios are found in case of shattering.

Table 6.3. Modelling results of the three selected break-up mechanisms, breakage interval = 2.

<table>
<thead>
<tr>
<th>experiment</th>
<th>breakage mechanism</th>
<th>$\beta_0$</th>
<th>$\alpha_0$</th>
<th>ratio $\beta_0/\alpha_0$</th>
<th>SS res mass</th>
<th>SS res colour</th>
<th>SS res total</th>
</tr>
</thead>
<tbody>
<tr>
<td>cg low N</td>
<td>fragmentation</td>
<td>336</td>
<td>$3.0 \times 10^{-2}$</td>
<td>$1.1 \times 10^4$</td>
<td>5.8</td>
<td>3.0</td>
<td>8.8</td>
</tr>
<tr>
<td>cg low N</td>
<td>shattering</td>
<td>185</td>
<td>$3.9 \times 10^{-4}$</td>
<td>$4.7 \times 10^5$</td>
<td>10.3†</td>
<td>8.0†</td>
<td>18.4†</td>
</tr>
<tr>
<td>cg low N</td>
<td>abrasion</td>
<td>345</td>
<td>$4.1 \times 10^{-2}$</td>
<td>$8.5 \times 10^3$</td>
<td>5.1</td>
<td>3.1†</td>
<td>8.2</td>
</tr>
<tr>
<td>cg high N</td>
<td>fragmentation</td>
<td>1.3 $\times 10^4$</td>
<td>4.3</td>
<td>$3.1 \times 10^3$</td>
<td>24.2</td>
<td>11.8</td>
<td>36.0</td>
</tr>
<tr>
<td>cg high N</td>
<td>shattering</td>
<td>363</td>
<td>$9.0 \times 10^{-3}$</td>
<td>$4.0 \times 10^4$</td>
<td>28.6†</td>
<td>70.4†</td>
<td>99.0†</td>
</tr>
<tr>
<td>cg high N</td>
<td>abrasion</td>
<td>1.2 $\times 10^4$</td>
<td>4.5</td>
<td>$2.6 \times 10^3$</td>
<td>29.5</td>
<td>11.4</td>
<td>40.9</td>
</tr>
<tr>
<td>Gral</td>
<td>fragmentation</td>
<td>459</td>
<td>$2.8 \times 10^3$</td>
<td>$1.6 \times 10^5$</td>
<td>29.4</td>
<td>24.4</td>
<td>53.9</td>
</tr>
<tr>
<td>Gral</td>
<td>shattering</td>
<td>395</td>
<td>$1.3 \times 10^5$</td>
<td>$3.0 \times 10^7$</td>
<td>30.6</td>
<td>29.9</td>
<td>60.4</td>
</tr>
<tr>
<td>Gral</td>
<td>abrasion</td>
<td>489</td>
<td>$4.6 \times 10^3$</td>
<td>$1.1 \times 10^5$</td>
<td>29.5</td>
<td>23.5</td>
<td>53.0</td>
</tr>
</tbody>
</table>

*: cg stands for coffee grinder experiment.
†: significant excluded with the Barlett’s chi-square test.

Shattering is a very ‘effective’ breakage mechanism, because it results in many small particles, and no larger pellet remains after breakage. Fragmentation is less effective compared to shattering, because it results in larger fragments, and more breakage-steps are needed to obtain particles as small as the shattering results. Abrasion, with the lowest coalescence/breakage kernel ratio, is the least-effective breakage mechanism, because a large pellet remains after breakage. Many breakage-steps are needed to obtain particles as small as the shattering results. The breakage kernel, seems to indicate an ‘effectiveness of the breakage mechanism in order to form very small particles’, which will be discussed in greater depth in section 6.5.2.

6.4.2. Three break-up distributions

For the coffee grinder experiments, two breakage mechanisms (abrasion and fragmentation) are included as possible breakage mechanisms. It therefore is assumed that both mechanisms occur simultaneously during pelletisation.

Comparable results are obtained for the Gral experiment, where no breakage mechanism could be excluded on basis of the Barlett chi-square test. This could indicate that the three
breakage mechanisms occur simultaneously in the Gral during high-shear pelletisation. Furthermore, it is likely that each breakage mechanism has its own optimal location in the bowl. So, the three breakage mechanisms (fragmentation, shattering and abrasion) are proposed to occur simultaneously during the pelletisation process, each dominating at different locations within the bowl. It is possible to localise the three different breakage mechanisms in the bowl as follows.

6.4.2.1. Fragmentation

During the pelletisation process, fragmentation does not occur at all locations within the mixer bowl. Because high impact forces are needed to fragment a pellet, it is likely that fragmentation occurs only at locations where the highest shear and impact forces are present (e.g. locations where the differences in velocity are high). It therefore is supposed that fragmentation occurs at the chopper and the impeller, because of the high tip speed resulting in high impact forces.

6.4.2.2. Shattering

Just like the fragmentation, it is also likely for shattering to occur at the impeller and the chopper site, because of the high impact forces at these locations. A difference between fragmentation and shattering is the force needed to generate these breakage mechanisms. Thornton et al. observed shattering at high impact velocities, and fragmentation (semi-brittle fracture) at moderate impact velocities\(^{12}\). The mechanism of breakage is influenced by the strength of the pellets and the location of the pellets in the bowl (near the impeller or the chopper, which also disturbs the flowing profile of pellets even if it has been set-off). As a result of shattering, many fine particles are formed, which can cause an increase of the mass of the smallest sieve fraction, followed by growth by layering.

6.4.2.3. Abrasion

Less impact is needed for the abrasion of material from the outer shell of a pellet. The force to remove a primary particle from a pellet can be estimated with the following equation:

\[
F_p = \frac{1}{4} \pi d_p^2 \frac{1 - \varepsilon}{\varepsilon} \frac{F_{pp}}{d_{pp}^2} \Rightarrow \frac{F_p}{F_{pp}} \propto \left( \frac{d_p}{d_{pp}} \right)^2
\]  

(6.20)

Where \(d_p\) is the diameter of the pellets (~1 mm), and \(d_{pp}\) is the diameter of the primary particles (63 \(\mu m\)). Because the diameter of a primary particle is about 20 times smaller than the diameter of a pellet, the force needed to break a primary particle from a pellet is 400 times smaller than the force needed to break a whole pellet. Consequently, abrasion can occur everywhere inside the
mixer bowl, and not only at locations with high velocity differences. Beside the impeller and
the chopper site, abrasion also can occur at the wall as a result of pellet-wall collisions. Even
pellet-pellet collisions somewhere in the torus can cause breakage by abrasion.

6.5. Physical interpretation of the fit-parameters

In order to find a physical interpretation of the two fit-parameters used during the population
balance modelling, the following assumptions have been made.
The over-all coalescence kernel ($\beta_0$) is supposed to represent an ‘effectiveness of the
coaalescence mechanisms’, which can be related to a number of successful pellet-pellet
collisions in the bowl during pelletisation. After all, growth only proceeds after a successful
pellet-pellet collision.
The over-all breakage kernel ($\alpha_0$) is supposed to represent an ‘effectiveness of the breakage
mechanism’, which will be related to a number of pellet-impeller collisions. It is reasonable to
expect that breakage of pellets is caused by the highest impact e.g. impeller-impact or
chopper-impact. The coffee grinder does not have a chopper at all, and during the Gral
experiment the chopper was set-off. For this reason, the breakage kernel is proportional to the
‘number of pellet-impeller impacts per unit time’ and the pellet-chopper collisions will be
neglected.

Growth and breakage of pellets is supposed to happen at different locations within the bowl.
Growth can occur at locations where low-impact pellet-pellet collisions occur frequently,
while breakage can occur at locations where high-impact pellet-impeller collisions occur. We
thus have to try to find a way to calculate the coalescence kernel by calculating a number of
pellet-pellet collisions during pelletisation. This can be calculated with the power input and
the kinetic energy.

6.5.1. Physical interpretation of the coalescence kernel

The coalescence kernel, or the number of pellet-pellet collisions inside the apparatus, can be
estimated from the energy input. In fact, the energy input is used to move the pellets in the
bowl causing pellet-pellet interactions. Hence, the energy input is dissipated into kinetic
energy by the pellet-pellet collisions. The power input is given by:

\[ P = m \omega C_p \Delta T \Delta t \]  

(6.21)

The temperature changes were estimated at the different scales of operations. The kinetic
energy of one pellet is given by:

\[ E_{\text{kin}} = \frac{1}{2} m_p v_r^2 \]  

(6.22)

Where $v_r$ is the relative velocity between two layers of moving pellets, with a thickness of $d_p$,
which is a function of the height of the torus ($h_{\text{torus}}$) and the size of a pellet (see figure 6.5):
The calculated number of collisions per pellet per second \( (N_{\text{collisions}}) \) is given by:

\[
N_{\text{collisions}} = \frac{P}{2(1-e^2)E_{\text{kin}}} \tag{6.24}
\]

Here \( e \) is the coefficient of restitution based on linear velocity differences (see also appendix chapter 4). The factor 1/2 is introduced in order to prevent double counting of each collision between two pellets.

A collision between two particles can result in coalescence. The chance on a collision depends on half of the square of the number of pellets present and a kind of growth-characteristic rate constant, which is represented by the coalescence kernel. The total number of collisions \( (N_{\text{collisions}}) \) per unit time is given by:

\[
N_{\text{collisions}} = \frac{1}{2} \beta_{0,\text{calc}} n_p^2 \tag{6.25}
\]

(Note that the number of collisions per unit time can also be derived from the second term of the right-handed site of equation 6.2.)

In which \( n_p \) is the total number of pellets, given by:

\[
n_p = \frac{m_{\text{tot}}}{m_p} \tag{6.26}
\]

Combining equations 6.22 - 6.26 results in an expression for the calculated coalescence kernel \( (\beta_{0,\text{calc}}) \):

\[
\beta_{0,\text{calc}} = \frac{2C_p \Delta T}{(1-e^2) n_p V_r^2} \tag{6.27}
\]

Some input parameters and the results of the calculations are presented in table 6.4. The values of the coalescence kernel obtained with the population balance, \( \beta_{0,\text{model}} \), are also given. It is clear that there is still a large difference between the modelled values and the calculated values. Apparently, more factors are involved than given in the simplified calculations.
6.5.2. Physical interpretation of the breakage kernel

The breakage kernel, or the number of pellet-impeller collisions in the different scales of operation, can also be calculated with the power input. The total power input has already been given with equation 21. The power input initially is dissipated to the pellets with direct pellet-impeller collisions. This is the only way in which the pellets can obtain a velocity, which will be in the order of the tip velocity of the impeller \( (v_{\text{tip}}) \):

\[
E_{\text{kin,a}} = \frac{1}{2} m_p v_{\text{tip}}^2
\]  

(6.28)

The number of impeller-pellet collisions per unit time for the breakage kernel \( (N_{\text{collisions,a}}) \) can be calculated using equation 6.28 for the kinetic energy.

\[
N_{\text{collisions,a}} = \frac{P}{2(1 - e^2)E_{\text{kin,b}}}
\]

(6.29)

The total number of collisions \( (N_{\text{collisions}}) \) resulting in breakage according to the population balance (last term of eq. 6.17) is given by:

\[
N_{\text{collisions}} = \alpha_{0,\text{calc}} n_p
\]

(6.30)

Combining equations 21, 6.28-6.30 results in an expression for the calculated breakage kernel \( (\alpha_{0,\text{calc}}) \):

\[
\alpha_{0,\text{calc}} = \frac{C_p \Delta T}{(1 - e^2)^2 v_{\text{tip}}^2}
\]

(6.31)

As can be seen in eq. 6.31, the calculated breakage kernel is independent of the pellet size (which is discussed in further detail in the appendix). Therefore, the same breakage kernel can be used for all sieve fractions.

Some input parameters and the results of the calculations are presented in table 6.4. The values of the breakage kernel obtained with the population balance, \( \alpha_{0,\text{model}} \), are also given. It is clear that there is still a large difference between the modelled values and the calculated values. Apparently, more factors are involved than given in the simplified calculations.

The ratio of the coalescence and the breakage kernel gives a nice indication of the differences between the three processes. At higher impeller speeds, it can be seen for the coffee grinder experiment, that this ratio becomes smaller. The coalescence kernel increases ten times at higher impeller speeds, while the breakage kernel increases more than one hundred times.

A higher impeller speed results in more collisions (which is also confirmed by the modelling of the toroidal flow pattern\(^{13}\)), and in larger conversion rates constants as previously found in chapter 3. In the Gral, the ratio between the coalescence kernel and the breakage kernel is ten times higher compared to the low impeller speed experiment in the coffee grinder. This can be caused by two factors. Firstly, the higher amounts of pellet present in the Gral cause more collisions, and secondly the relatively lower impeller rotational speed results in a lower breakage kernel.
Table 6.4. Data used for the calculation of $\beta_0$ and $\alpha_0$.

<table>
<thead>
<tr>
<th></th>
<th>cg low N</th>
<th>cg high N</th>
<th>Gral</th>
</tr>
</thead>
<tbody>
<tr>
<td>$N$ (rps)</td>
<td>82</td>
<td>187</td>
<td>6.7</td>
</tr>
<tr>
<td>$\Delta T/\Delta t$ (K/min)</td>
<td>20</td>
<td>30</td>
<td>6</td>
</tr>
<tr>
<td>$C_p$ (J/g K)</td>
<td>2</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>$m_{tot}$ (kg)</td>
<td>0.036</td>
<td>0.036</td>
<td>1.2</td>
</tr>
<tr>
<td>$e$ (-)</td>
<td>0.3</td>
<td>0.3</td>
<td>0.3</td>
</tr>
<tr>
<td>$v_{pellets}$ (cm/s)</td>
<td>10</td>
<td>10</td>
<td>19</td>
</tr>
<tr>
<td>$v_r$ (cm/s)</td>
<td>0.72</td>
<td>0.53</td>
<td>0.62</td>
</tr>
<tr>
<td>$m_p$ ($\mu$g)</td>
<td>0.25</td>
<td>0.14</td>
<td>0.69</td>
</tr>
<tr>
<td>$h_{tora}$ (cm)</td>
<td>2.0</td>
<td>2.3</td>
<td>6.3</td>
</tr>
</tbody>
</table>

| $\beta_0$,model        | 336      | $1.3 \times 10^4$ | 459  |
| $\beta_0$,calc         | 92       | 150       | 3.2  |
| ratio $\beta_0$,model/$\beta_0$,calc | 3       | 74        | 180  |

| $\alpha_0$,model       | 0.030    | 4.3       | $2.8 \times 10^{-3}$ |
| $\alpha_0$,calc        | 1.7      | 0.5       | 8.3  |
| ratio $\alpha_0$,model/$\alpha_0$,calc | 0.015    | 7.6      | $4 \times 10^{-4}$ |

| ratio $\beta_0$,calc/$\alpha_0$,calc | 53       | 300      | 0.4  |
| ratio $\beta_0$,model/$\alpha_0$,model | $1 \times 10^4$ | $3 \times 10^3$ | $2 \times 10^5$ |

### 6.6. Conclusions

The population balance modelling was used to describe the continuous growth and breakage of pellets during an equilibrium stage of growth.

Until now, population balances only included growth of particles; the breakage of particles was not included into the balance. Especially for the high-shear granulation processes, one has to include breakage of the agglomerates into the population balance. Therefore a population balance with separate growth and breakage kernels was performed. The development of the particle size distribution and a tracer component distribution of pellets during an equilibrium stage of growth was modelled using different breakage mechanisms. The population balance was principally based on number distributions but, for the sake of convenience, represented in mass distributions. Three breakage mechanisms were selected from literature and their importance at high-shear pelletisation was modelled with the population balances.

From the population balance modelling, it can be concluded that the most important breakage mechanisms in the coffee grinder are fragmentation and abrasion. For the Gral-experiment, the three breakage mechanisms (fragmentation, shattering and abrasion) showed similar contributions. A simultaneously occurring combination of two of the three possible breakage...
mechanisms within one apparatus is proposed, each mechanism occurring at its own location within the apparatus. Fragmentation and shattering are expected to occur at the locations with the highest shear forces present at the impeller and chopper site. Abrasion is expected to occur at locations with less shear forces e.g. at the wall or inside the torus as a result of pellet-wall and pellet-pellet collisions respectively.

The effect of higher impeller speeds on the different fitting parameters was also investigated. At higher impeller speeds, an increase in the coalescence and breakage kernels was found, which could be explained by an increase of the total numbers of collisions. More collisions between the pellets and the impeller arm cause faster growth and breakage of pellets.

The population balance showed to be a useful tool to describe pellet growth and pellet breakage during the high-shear pelletisation process. A physical interpretation of the fitted parameters is still rather speculative, and has to be studied in further detail. Apparently, more factors are involved than given in the simplified calculations. After modelling the exact toroidal flow pattern of pellets in a high-shear mixer, one must be able to gain further insight in the physical interpretation of the coalescence and breakage kernels. The first approximation of the torus flow pattern modelling\textsuperscript{13} showed to be very useful for the interpretation of the coalescence and breakage kernel. This remains to be investigated more exactly.
6.7. Nomenclature

\( C \)  
\text{colour concentration (mg/g)}

\( C_p \)  
\text{specific heat (J/kg K)}

\( d \)  
\text{diameter (m)}

\( e \)  
\text{coefficient of restitution based on linear velocity differences (-)}

\( E \)  
\text{energy (J)}

\( F \)  
\text{force (N)}

\( f(i,j) \)  
\text{break-up distribution function}

\( h \)  
\text{height of the torus (m)}

\( m_i \)  
\text{mass of one pellet from size class } i \text{ (kg)}

\( m_p \)  
\text{mean mass of one pellet (kg)}

\( \Delta m_{i,j} \)  
\text{amount of transferred mass from class } i \text{ to } j \text{ (kg)}

\( n_p \)  
\text{number of pellets}

\( n(v) \)  
\text{density function (-)}

\( N \)  
\text{impeller rotational speed (1/s)}

\( N_{\text{collisions}} \)  
\text{number of collisions per unit time (1/s)}

\( N_i \)  
\text{number of pellets in size class } i

\( P \)  
\text{power input (Watt)}

\( t \)  
\text{processing time (s)}

\( T \)  
\text{temperature (K)}

\( v \)  
\text{velocity (m/s)}

\( v_0 \)  
\text{volume of one pellet (m}^3\text{)}

\( V \)  
\text{volume of size class (m}^3\text{)}

\( \Delta V_{i,j} \)  
\text{volume change from size class } i \text{ to } j \text{ (m}^3\text{)}

\( x \)  
\text{break interval, number of size classes to which breakage occurs}

\text{Greek symbols}

\( \alpha \)  
\text{break-up kernel (1/s)}

\( \alpha_0 \)  
\text{over all break-up kernel (1/s)}

\( \beta \)  
\text{coalescence kernel (1/s)}

\( \beta_0 \)  
\text{over all coalescence kernel (1/s)}

\( \varepsilon \)  
\text{porosity}

\( \rho \)  
\text{density of the pellet (kg/m}^3\text{)}

\( \omega \)  
\text{mass fraction}

\text{Subscripts}

\( i \)  
impact

\( i,j,k \)  
size classes

\( n \)  
total number of size classes

\( p \)  
pellet

\( pp \)  
primary particles

\( r \)  
relative

\( \text{tot} \)  
total
6.8. References

Appendix to chapter 6

Explanation why the breakage of pellets is independent of the mass of the pellets.

Break-up of particles is described on basis of two forces:
1. Impact force of the impeller arm as derived in chapter 2.
2. Tensile strength of the pellet as described by Rumpf\(^1\).

Ad-1:
The **impact force** of the impeller arm is given by:

\[
F_i = m_p \left( \frac{d}{dt} \nu \right) = m \frac{\Delta \nu^2}{\Delta s} = \frac{\pi}{6} d_p^3 \left( 1 - \epsilon_p \right) \rho_s \frac{\Delta \nu^2}{\Delta s} \propto \frac{\pi}{6} d_p^3 \left( 1 - \epsilon_p \right) \rho_s \left( \pi ND \right)^2 \frac{1}{c_1 d_p} \\
\]

where \(m_p\) is the mass of the pellet, \(\Delta \nu\) the relative velocity between the impeller and the pellet, \(\Delta s\) the height of the deformation layer of the pellet, which is a function of the pellet size (\(\Delta s \propto c_1 d_p\)). \(c_1\) is a constant, \(\rho_s\) is the density of the starting materials, \(d_p\) the size of the pellet, and \(\epsilon_p\) the porosity of the pellet. In case of breakage, the relative velocity is a function of the tip speed of the impeller (\(\pi ND\)). The impact pressure is given as the impact force divided by the surface of a pellet (\(\pi d_p^2\)):

\[
\sigma_i = \frac{F_i}{A_p} \propto \frac{\pi}{6} d_p^2 \left( 1 - \epsilon_p \right) \rho_s \left( \pi ND \right)^2 \frac{1}{c_1} \\
\]

Here \(c\) is a constant.

As can be seen with equation A2, the impact pressure is independent of the pellet size.

Ad-2:
The **tensile strength** (\(\sigma_t\)) according Rumpf\(^1\) is given by:

\[
\sigma_t = \frac{1 - \epsilon_p}{\epsilon_p} \cdot \frac{F_{pp}}{d_{pp}^2} \\
\]

where \(d_{pp}\) is the size of the primary particles (starting material), and \(F_{pp}\) is the force holding these primary particles together. This either can be a capillary force or a viscous force.

The **capillary** force according to Rumpf\(^1\) is given by:

\[
F_c = \pi \cdot \gamma \cdot d_{pp} \\
\]

In case of capillary forces, the tensile strength becomes:

\[
\sigma_{t,c} = C \cdot S \cdot \frac{1 - \epsilon_p}{\epsilon_p} \cdot \frac{\pi \gamma}{d_{pp}} \propto \frac{\gamma}{d_{pp}} \\
\]

Here \(C\) is the co-ordination number, \(S\) is the saturation, and \(c_e\) is a constant. Note that the
capillary force depends on the size of the primary particle, but is independent of the pellet size.

The **viscous** force is given by:

\[
F_v = \frac{3\pi \eta d_{pp}^2 \Delta \nu}{8z}
\]

where \( \eta \) is the viscosity of the binding liquid, and \( z \) the interparticle distance.

The ratio of the volume of solids \( (V_s) \) to the volume of liquids \( (V_l) \) is given by (assuming that all pores are filled with liquid):

\[
\frac{V_s}{V_l} = \frac{1 - \varepsilon_p}{\varepsilon_p}
\]

(A7)

The ratio of the primary particle size and the interparticle distance (distance between the primary particles) can be estimated similar to the ratio of equation A7, as:

\[
\frac{d_{pp}}{z} = \frac{1 - \varepsilon_p}{\varepsilon_p} \Rightarrow z = \frac{\varepsilon_p}{1-\varepsilon_p} d_{pp}
\]

(A8)

In case of viscous dissipation, the tensile strength becomes:

\[
\sigma_{t,v} = C \cdot S \left( \frac{1 - \varepsilon_p}{\varepsilon_p} \right)^2 \cdot \frac{3\pi \eta \Delta \nu}{8} \cdot \frac{\Delta \nu}{d_{pp}^2} = c_v \frac{\Delta \nu}{d_{pp}^2}
\]

(A9)

Here \( c_v \) is a constant.

Note again that the strength of the pellets depends on the primary particle size, but is independent of the pellet size.

Breakage occurs if the impact pressure of the impeller exceeds the tensile strength of the pellet \( (\sigma_{\text{impact}} > \sigma_{\text{tensile}}) \). Here the impact pressure of the impeller is given by equation A2, and the tensile strength of the pellets is given either by equation A5 or A9. In both situations, the impact pressure of the impeller arm as well as the tensile strength of the pellet are independent of the pellet size. Therefore, the breakage kernel \( (\alpha_0) \) of the population balance is also independent of the pellet size.
Nomenclature to appendix

A surface area (m²)
c constant
C co-ordination number
d diameter pellet (m)
D diameter impeller (m)
e coefficient of restitution based on linear velocity differences (-)
F force (N)
m mass (kg)
N impeller rotational speed (1/s)
s deformation layer (m)
S saturation (-)
t processing time (s)
v velocity (m/s)
z interparticle distance (m)

Greek symbols
ε porosity (-)
γ surface tension (N m⁻¹)
η viscosity (Pa s)
ρ density of the pellet (kg/m³)
σ strength (Pa)
ω mass fraction (-)

Subscripts
c capillary
i impact
i,j,k size classes
n total number of size classes
p pellet
pp primary particles (starting material)
r relative
s solid
v viscous

References to appendix
