Deformation of spherical pellets: determination of the yield pressure and E-modulus of wet microcrystalline cellulose pellets

Abstract

The deformation of wet pellets under shear is described, based on results from simple horizontal impact measurements of a wet pellet against a wall. The pellets were made of microcrystalline cellulose (MCC) and different amounts of water. The coefficient of restitution (resulting from the elastic deformation) after a pellet – wall collision was found to be about 0.3. The irreversible pellet deformability is represented by a dimensionless deformation number, which either is based on viscous (strain rate dependent) or plastic (strain rate independent) deformation, and could be calculated from the deformed volume during impact. At low impact velocities, the deformation was found to be viscous, at higher impact velocities a more plastic deformation was found. The yield pressure of the wet pellets was calculated from the deformed volume and the energy used for deformation. The E-modulus was calculated with a force-displacement model. The yield pressure of wet MCC-pellets was about 10 times lower compared to the values of dry MCC powder; the reduced E-modulus of wet MCC-pellets was about 100 times lower compared to dry MCC powder. From the experiments it was found that the moisture content is a key-variable for the value of the yield pressure as well as the E-modulus. The moisture content reduced both the inter-particle friction and the capillary pressure, resulting in a decreasing yield pressure and E-modulus with increasing moisture content. These experiments have great potential to help understanding and describing the influence of material properties on the granulation processes in high-shear mixers.
5.1. Introduction

Granulation is a size enlargement method, which has been used in pharmaceutical, food, detergent and fertiliser industry for many years. Most investigations in granulation processes are aimed at understanding the process: describing and explaining what happens. Because a total understanding of the granulation process is still missing, many changes in the granulation conditions are mostly based on trial and error. The aim of fundamental granulation studies is to be able to predict the outcome of the granulation process through the knowledge of the apparatus, starting materials, and granulation conditions. This finally should reduce the costs of developing the process.

The properties of the granules change continuously during the granulation process. Take for example the relationship between the binder content and the granule size and strength. This relationship has been described to a large extent but still has not been understood yet. The only way to monitor the granulation process on-line is to measure the energy-input by measuring the power consumption, torque or the temperature. The relationship between the power consumption and the granule size distribution is a nice tool to estimate the processing time. But a complete understanding of what goes on during the granulation process is still not provided.

It is possible to measure some granule properties off-line the granulation process. For example the granule size distribution, granule shape, porosity or even a measure for the granule strength (mostly obtained by making tablets from dry granules and measure the tablet strength), and wet mass consistency can be measured. However, for a good understanding of the granulation process, it is necessary to know the properties of the wet granules instead of the properties of the dry granules. The wet granule properties provide insight in the effect of the granule conditions needed to form the desired product. Also questions like: “why are granules formed and broken during specific time-periods occurring in the granulation process” can be answered. Finally, it can be stated that to understand the granulation process in depth it is necessary to measure the properties of wet granules. This chapter makes a contribution to this part of the granulation research.

The aim of this study is to measure the material properties of wet pellets with simple impact experiments. An attempt is made to describe the deformation of the pellet in terms of viscous deformation (in this chapter referred to as strain rate dependent deformation) and plastic deformation (in this chapter referred to as strain rate independent deformation), and to measure the influence of the moisture content on the measured material properties.
5.2. Theoretical considerations

5.2.1. Collision between two pellets

The collision between two pellets starts with the approach of two pellets. The contacts between the binder on the two surfaces cause the formation of liquid bridges. Deformations of the surfaces of both pellets occur at high kinetic energies (impact velocities) of both pellets. Initially this deformation is an elastic deformation (e.g. a reversible deformation), at higher impact velocities the deformation can also be a plastic or viscous deformation (e.g. a non-reversible deformation). Deformation of the surface results in a small local densification of the pellet. As a consequence, liquid from the inside of the pellet is squeezed out to the surface of the pellet, increasing the amount of liquid on the surface. This results in more binder liquid available for maintaining the liquid bridge. Another effect of the deformation of the surface is the increase in effective contact area between the two pellets. (If no liquid is present at the surface, the contact area is increased to the size of the deformed surface. A deformed surface increases the solid-solid interaction, and therefore the chance on solid-binder bonds or interlocking bonds between the pellets.) After maximal deformation, the stored elastic energy causes rebound of the two pellets. By removing the deformation pressure, partial relaxation of the deformed surface occurs. The internal structure of the pellet partly recovers, resulting in partial re-uptake of the binder liquid into the pellet. This diminishes the amount of binder liquid on the surface, and therefore decreases the strength of the binder liquid bridge. Because the velocities of the pellets are now orientated in the opposite directions, an elongation of the binder liquid bridge occurs. The strength of the binder liquid bridge, the amount of liquid in the bridge, and the relative velocities of the pellets determine whether or not the liquid bridge breaks. If the liquid bridge breaks, the coefficient of restitution can be estimated as the ratio of the linear velocity differences between the two pellets after and before impact (see also appendix to chapter 4). If the liquid bridge does not break, coalescence occurs and the coefficient of restitution is zero.

5.2.2. Deformation and coalescence

Iveson et al. investigated the influence of the binder content on the deformability of wet pellets. They considered the pellet as a rigid-plastic material with yield pressure independent of the strain rate. For a cylinder of such a material impacting on a plate surface at velocity \( v_i \), the increase in contact area of the cylinder due to plastic deformation has been adapted from Hawkyard and can be used to estimate the dynamic yield pressure of a material (\( \sigma_{yH} \), in which H stands for the Hawkyard method):

\[
2 \left[ \frac{A_0}{A} - 1 + \ln \left( \frac{A}{A_0} \right) \right] = \frac{\rho_p v_i^2}{\sigma_{yH}} \tag{5.1}
\]
where $A_0$ and $A$ are the contact area of the cylinder before and after impact, respectively. To characterise the extent of deformation, a dimensionless deformation number ($De$) was proposed\cite{Ivesonetal2010}, which is given by:

$$De = \frac{\rho_p v_i^2}{\sigma_{sh}}$$ \hspace{1cm} (5.2)

where $\rho_p$ is the density of the pellet, $v_i$ is the impact velocity (which is of order $\pi ND$ for granulations, where $N$ is the rotational speed and $D$ is the impeller diameter). In high-shear granulation the velocity of the impeller arm is much higher than the velocity of the pellets. Whereas pellet-impeller collisions can cause breakage of the pellets, the pellet-pellet collisions are supposed to cause deformation of the pellets resulting either in a change of sphericity or in coalescence. It therefore would be necessary to use the relative velocity between the moving pellets (e.g. impact velocity of a pellet-pellet collision) instead of the impeller rotational speed to characterise the deformation behaviour.

The velocities of the pellets in the torus are about 1 m/s (chapter 3), while the tip-velocity of the impeller arm varied between 6 and 20 m/s. The impact velocities used in the experiments described in this paper are in the order of the velocities of the pellets inside the torus. Therefore, pellet – pellet collisions as well as pellet – wall collisions are taken into account in our experiments.

Tardos et al.\cite{Tardosetal2011} derived the coalescence Stokes number, which indicates a coalescence probability for non-deformable pellets. This coalescence Stokes number is defined as the ratio of the initial kinetic energy of a pellet and the dissipated energy in the bridge, that is:

$$St_{coal} = \frac{8r_p \rho_p v_0}{9\eta_l}$$ \hspace{1cm} (5.3)

where $r_p$ is the radius of the pellet, $v_0$ the relative velocity between two pellets, $\rho_p$ the pellet density, and $\eta_l$ is the viscosity binder liquid at the surface of the pellet. The coalescence Stokes number increases as the pellet size increases during granulation. Higher coalescence Stokes numbers indicate a smaller coalescence probability.

Tardos et al.\cite{Tardosetal2011} have also developed a relationship (comparable to the deformation number of Iveson et al.\cite{Ivesonetal2010}) between the externally applied kinetic energy and the energy required for deformation: the deformation Stokes number (eq. 5.4). This Stokes number is based on the deformation of the non-rigid core of a pellet. The deformation Stokes number is given by the ratio of the kinetic energy applied and the energy required for deformation, that is:

$$St_{def} = \frac{\rho_p v_0^2}{8\sigma_y}$$ \hspace{1cm} (5.4)

where $\sigma_y$ is the yield pressure of the wet pellet. The deformation Stokes number increases as the shear rate increases during granulation. For deformation Stokes numbers larger than the critical Stokes number, deformation and breakage of the pellets occur.
According to the deformation Stokes number, an increased velocity results in an increased deformation probability. Higher impact velocities can cause more deformation of the surface of the pellet, resulting in a larger contact area between two colliding pellets. As a consequence, minor specific bridging force is needed to reach a permanent bond, and coalescence will occur. On the contrary, according to the coalescence Stokes number, an increased velocity also results in a decrease of the coalescence probability and in an increase of the rebound probability. Because deformation of a pellet affects the coalescence probability, a comparison between deformation probability and coalescence probability is not as simple as suggested by Tardos et al.\textsuperscript{6} using both Stokes numbers.

5.2.3. Moisture-dependent deformation

Deformation of the pellets not only depends on the viscosity of the binder liquid and the yield pressure of the wet pellet, it also depends on the liquid content at the surface of the pellets. The saturation ($S$) is a characteristic measure for the fractional amount of liquid inside the pores of a pellet:

$$S = H \frac{\rho_p}{\rho_l} \left(1 - \frac{\rho_l}{\rho_p} \right)$$

(5.5)

where $H$ is the mass ratio of liquid to solid, $\rho_p$ and $\rho_l$ the density of the solid and the liquid respectively, and $\varepsilon$ the porosity of the wet pellet.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{fig5.1.png}
\caption{Relationship between tensile strength of wet granules and the saturation. $S_p$ denotes the end of the pendular state, and $S_c$ the end of the capillary state.}
\end{figure}

The relationship between the pellet tensile strength and the saturation, as derived by Schubert\textsuperscript{7}, is schematically given in figure 5.1. During the funicular and the capillary state (saturation is between $S_p$ and $S_c$), an increased saturation results in an increase of the tensile strength. At the end of the capillary state (saturation $S_c$), the maximal tensile strength has been reached. Above the maximal tensile strength (at $S > S_c$), the tensile strength decreases drastically with increasing saturations (figure 5.1).

Consider two pellets of equal masses and equal saturations colliding with a relative velocity $v_0$. For saturations of the pellets between $S_p$ and $S_c$, a collision between the pellets results in
local surface deformation. Because all pores are not completely filled with liquid, the amount of liquid at the surface does not increase. The saturation at the surface of the pellet increases, but not enough to result in coalescence. The collision results in rebound.

If the saturation of both pellets is higher than $S_c$, a collision between the pellets results in deformation of the surfaces of the pellets. Because all pores are completely filled with liquid, the amount of liquid at the surface increases: there is free liquid available on the surface of the pellets to form a liquid bridge. The collision results in coalescence.

### 5.3. Modelling of pellet deformation

For the modelling of the collision-induced deformation of a pellet, a simplified concept of bouncing of a spherical pellet on a flat plate was used. During deformation of a sphere on a plate, a segment of the sphere is assumed to be deformed into a cylinder. The radius of the sphere is $r_p$, the mass of the sphere is $m_p$, and $v_i$ is the impact velocity. The deformed segment of the sphere is characterised by its radius $r_i$ (which is also the radius of the cylinder) and its height $h$ (which is not equal to the height of the cylinder, see also figure 5.2).

The volume of the deformed sphere segment ($V_{\text{seg}}$) is given by:

$$ V_{\text{seg}} = \pi h^2 \left( r_p - \frac{h}{3} \right) $$

(5.6)

The volume of the formed cylinder ($V_{\text{cyl}}$) is given by:

$$ V_{\text{cyl}} = \pi r_i^2 y $$

(5.7)

The height of the deformed layer is given with the Pythagorean rule:

$$ h = r_p - \sqrt{r_p^2 - r_i^2} $$

(5.8)

The square of the deformation radius is given by:

$$ r_i^2 = h(2r_p - h) $$

(5.9)

The height of the formed cylinder ($y$) can be obtained by combining eqs. 5.6 - 5.9:

$$ y = \left( r_p - \sqrt{r_p^2 - r_i^2} \right)^2 \left( \frac{2r_p + \sqrt{r_p^2 - r_i^2}}{r_i^2} \right) $$

(5.10)

The deformed volume can now be written as a function of the pellet radius and the impact radius by using eqs. 5.7 and 5.10.

The movement of the pellet during deformation is given by the equations of motion:

$$ -\frac{dh}{dt} = v $$

(5.11)

$$ \frac{dv}{dt} = \frac{F_g}{m_p} $$

(5.12)

It is possible to calculate the energy of deformation ($U$) with:

$$ U = -\int F_g dh $$

(5.13)
5.3.1. Plasticity-viscosity method

The plasticity-viscosity method (shortened as PV-method) describes two kinds of deformation: the viscous (strain rate dependent) deformation and the plastic (strain rate independent) deformation. Modelling the viscous and the plastic deformation results in two dimensionless numbers. One involves viscous deformation ($K_v$) and one involves plastic deformation ($K_p$). These numbers have the same numerical value (as will be seen when comparing eqs. 5.18 and 5.23), but contain partly different physical parameters. By plotting the dimensionless number against the strain rate, a distinction can be made between viscous and plastic deformation.

5.3.1.1. Viscous deformation

Viscous deformation is the dynamic deformation of a body and is a function of the velocity. The viscous deformation force ($F_v$) is proportional to the impact area, the viscosity of the wet pellet ($\eta$) and the deformation rate ($G$), which is the velocity divided by the pellet size ($v/2r_p$):

$$F_v = -\pi r_p^2 \eta G = -\frac{\pi \eta v}{2r_p} h(2r_p - h)$$  \hspace{1cm} (5.14)

The final deformation of the pellet can be obtained by integration of the equation of motion (eq. 5.11 and 5.12). Combining these equations with eq. 5.14 gives:

$$\frac{dv}{dt} = \frac{F_v}{m_p} = -\frac{\pi \eta v h(2r_p - h)}{2r_p m_p} = \frac{\pi \eta}{2r_p m_p} \frac{dh}{dt} h(2r_p - h) \Rightarrow \frac{dv}{dh} = \frac{\pi \eta}{2r_p m_p} h(2r_p - h)$$  \hspace{1cm} (5.15)

The solution of this differential equation is given by:

$$\int_{v_i}^{0} dv = \frac{\pi \eta}{2r_p m_p} \int_0^{h_{max}} h(2r_p - h) dh \Rightarrow v_i = \frac{\pi \eta}{2r_p m_p} \left( r_p h_{max}^2 - \frac{1}{3} h_{max}^3 \right)$$  \hspace{1cm} (5.16)
This equation can be simplified using a dimensionless length of deformation ($f$):

$$h_{\text{max}} = r_p f_{\text{max}}$$  \hspace{1cm} (5.17)

Combining eqs. 5.16 and 5.17 results into:

$$\frac{1}{3K_v f_{\text{max}}^3} - \frac{1}{K_v f_{\text{max}}^2} + 1 = 0$$  \hspace{1cm} (5.18)

Here $K_v$ is given by:

$$K_v = \frac{2m_p v_i}{\pi \eta r_p^3} = \frac{8r_p \rho_p v_i}{3\eta}$$  \hspace{1cm} (5.19)

Combining eqs. 5.8 and 5.17 - 5.19 it now is possible to estimate the viscosity of the wet pellets by measuring the wet radius ($r_p$) and the impact radius ($r_i$) of a pellet impacting on a plate.

This dimensionless $K_v$-value is comparable to the coalescence Stokes number as derived by Tardos et al. (see eq. 5.3). The $K_v$-value describes the deformation of the pellet, whereas the coalescence Stokes number describes the deformation of the liquid bridge. The viscosity of the $K_v$-value given in eq. 5.19 is the viscosity of the wet pellet as a whole, while the viscosity of the coalescence Stokes number is the viscosity of the binder liquid bridge.

### 5.3.1.2. Plastic deformation

If the deformation force on the pellet is not a function of the velocity, plastic deformation occurs, which is characterised by a constant yield pressure. The force is proportional to the impact area, and the yield pressure ($\sigma_{yPV}$):

$$F_p = \pi r_p^2 \sigma_{yPV} = \pi h(2r_p - h)\sigma_{yPV}$$  \hspace{1cm} (5.20)

The maximum deformation of the pellet is obtained by integration of the equation of energy conservation (eq. 5.13):

$$\frac{dU}{dh} = -\pi h(2r_p - h)\sigma_{yPV} \Rightarrow \int_0^{U_{\text{max}}} dU = -\pi \sigma_{yPV} \int_0^{h_{\text{max}}} h(2r_p - h)dh = \pi \sigma_{yPV} \left( \frac{1}{3} h_{\text{max}}^3 - r_p h_{\text{max}}^2 \right)$$  \hspace{1cm} (5.21)

This equation can be rearranged to a dimensionless form using the dimensionless length of deformation ($f$). The maximum energy of deformation is proportional to the fraction of the kinetic energy used for non-elastic deformation, e.g.:

$$U_{\text{def}} = -\frac{1}{2} \left( 1 - e^2 \right) m_p v_i^2$$  \hspace{1cm} (5.22)

Combining eqs. 5.17 and 5.20-5.22 results into:

$$\frac{1}{3K_p f_{\text{max}}^3} - \frac{1}{K_p f_{\text{max}}^2} + 1 = 0$$  \hspace{1cm} (5.23)
Here \( K_p \) is given by:

\[
K_p = \frac{2(1-e^2)n_p v_i^2}{\pi r_p^3 \sigma_{yPV}} = \frac{8(1-e^2)p_p v_i^2}{3\sigma_{yPV}}
\]  

(5.24)

This dimensionless \( K_p \)-value is very similar to the deformation Stokes number derived by Tardos et al. (eq. 5.4) and the deformation number derived by Iveson et al. (eq. 5.2). Differences are, except some constants, the inclusion of the coefficient of restitution in our dimensionless \( K_p \)-value, and the use of the impact velocity instead of the impeller tip velocity as used by Iveson et al. Especially considering pellet-pellet collisions, the impact velocity is much lower than the tip-velocity.

Besides the possibility to calculate the dimensionless plastic deformation value (\( K_p \)) it also is possible to estimate the yield pressure (\( \sigma_{yPV} \)) of a wet pellet using eqs. 5.8, 5.17, 5.23 and 5.24.

The values of \( K_p \) and \( K_v \) (which are actually the same, see eqs. 5.18 and 5.23) can be calculated with the dimensionless length of deformation (\( f \)). The plasticity index (\( n \)) is a measure for the dependency of the \( K \)-value on the strain rate and is defined as the power of the strain rate:

\[
K(\eta \text{ or } \sigma) = c \cdot G^n
\]

(5.25)

where \( c \) is a constant, \( G \) is the impact strain rate, and \( n \) is the plasticity index. At \( n = 1 \), a pure viscous behaviour is described, at \( n = 2 \) the pure plastic behaviour is described. The value of the plasticity index can be estimated as the slope of the \( \log(K) \) versus \( \log(G) \) curve. So, by estimating the plasticity index, a distinction between viscous deformation and plastic deformation can be made.

### 5.3.2. The Hawkyard method

The Hawkyard method can be used to estimate the exact value of the yield pressure (\( \sigma_{yH} \)). A model derived by Hawkyard\(^5\) describing the deformation of a cylinder has been adapted to be used in our experiments. Hawkyard has found a relationship for the increase in area of the cylinder due to deformation of a cylinder. In our model, we assume that a cylinder is formed during deformation of the surface of the sphere. This requires changes in the model of Hawkyard, but the model is still based on the same approach. The deformation of the sphere and the subsequent formation of the cylinder (see figure 5.2) require the following amount of work (\( dU_p \)):

\[
dU_p = \sigma_{yH} 2\pi r_i y d r_i
\]

(5.26)

where \( \sigma_{yH} \) is the yield pressure of the pellet, and \( V_{def} \) is the volume of the deformed segment given by eq. 5.6. The total required work of deformation (\( U_p \)) is given by:
\[ U_p = \sigma_{yH} \frac{2\pi}{r_p} \int_{r_i}^{r_p} r_i y dr_i \]  

(5.27)

where \( y \) stands for the height of the deformed layer (see eq. 5.10). The integral-function of this function is related to the deformed volume of the pellet. The following relationship between the integral-function and the deformed volume of the pellet (\( V_{def} \)) was experimentally found:

\[ 2\pi \int_{r_i}^{r_p} r_i y dr_i = \left( 1 - 0.252 \cdot \sqrt{\theta} - 0.261 \cdot \theta \right) \cdot V_{def} \]  

(5.28)

where \( \theta \) stands for the fractional volume deformed (\( \theta = V_{def}/V_{pellet} \)). By combining eqs. 5.27 and 5.28 it was possible to calculate the deformation energy from the experimentally found deformed volume of the pellet.

The available amount of energy that can be used for the deformation is equal to the kinetic energy at impact. The kinetic energy at (\( U_{kin} \)) impact is given by:

\[ U_{kin} = \frac{2\pi}{3} r_p^3 \rho_p v_i^2 \]  

(5.29)

where \( v_i \) is the velocity at impact, \( r_p \) the radius of the wet pellet and \( \rho_p \) the density of the wet pellet. A part of the kinetic energy of the pellet causes a rebound. The coefficient of restitution is defined as the square root of the fraction of the energy used for the rebound of the pellet. The coefficient of restitution is based on linear velocity difference (\( e \)) for a pellet-wall collision and is given by (see also appendix to chapter 4):

\[ e = \frac{v_r}{v_i} \]  

(5.30)

where \( v_i \) is the impact velocity and \( v_r \) is the rebound velocity.

The energy used for the deformation of the volume of the segment is equal to the loss in kinetic energy during impact. The fraction of the kinetic energy dissipated into plastic energy is defined as \( 1 - e^2 \). In this case frictional forces are neglected. The amount of kinetic energy used for the deformation of the volume of the segment is therefore equal to:

\[ U_{def} = \frac{2\pi}{3} r_p^3 \rho_p v_i^2 \left( 1 - e^2 \right) \]  

(5.31)

Because \( U_p = U_{def} \), the yield pressure can be estimated graphically by plotting the deformed volume (see eq. 5.6) vs. the deformation energy (eq. 5.31). The slope of the graph represents \( 1/\sigma_{yH} \).

5.3.3. Plasticity-viscosity method versus Hawkyard method

A comparison of the two methods used for the estimation of the yield pressure can be made. Generally, in the range of our measurements (until 1.5% deformed volume) the yield pressure according to the Hawkyard-method is four times lower compared to the PV-method. The size
of the deformed volume taken into account in each method differs. Therefore, a difference in yield pressures between those two methods is found.

The differences between the Hawkyard-method and the plasticity-viscosity method (PV-method) can be eliminated if a smaller deformed segment volume (with height \( h_y \)) is used instead of the original deformed segment volume (height \( h \) as given in eq. 5.8, see also figure 5.2). We decided to show both methods, because both methods have their own application. The PV-method makes it possible to distinguish between plasticity and viscosity. Similarity between PV-method and Stokes or deformation numbers is noticeable. The Hawkyard-method is a method, which is easy to understand and makes it very appropriate to calculate a yield pressure of wet granules.

### 5.3.4. Estimation of the E-modulus

A theoretical model for the bounce behaviour of one visco-elastic sphere against a wall, as recently described by Thornton et al.\(^8\), was used for the estimation of the E-modulus. During impact a Herznian pressure-profile inside the pellet was assumed (see figure 5.3) by Thornton et al.

![Figure 5.3. Deformed volume of the sphere (left) and the accompanying Herznian pressure distribution in the sphere and the deformed volume of the sphere (right).](image)

The force-displacement relationship during deformation of a sphere is given in figure 5.4. Pure elastic deformation occurs at minor loading. At a certain loading force \( F_{\text{yield}} \), the pressure at the deformed layer becomes higher than the yield pressure, resulting in a plastic deformation of the pellet. The plastic deformation of the pellet is characterised by a linear relationship between the force and the displacement. At the maximal loading force, a maximal deformation of the pellet occurs resulting in the maximal impact radius of the pellet. During plastic deformation, part of the deformation energy will be stored as elastic energy, resulting in a rebound after removal of the force. The area under the elastic unloading force-displacement curve (the elastic unloading energy), as given in figure 5.4, is assumed to be equal to the rebound energy of the pellet. The ratio of the rebound energy and the maximal
impact energy gives the coefficient of restitution. At impact velocities lower than the yield velocity \( v \leq v_y \) no deformation of the pellet occurs (ignoring the energy losses due to friction between the pellet and the wall) and the coefficient of restitution is 1. At higher impact velocities, the pellet starts to deform plastically, and the rebound kinetic energy is equal to the work done during elastic recovery.

![Force-displacement relationship for the deformation of a sphere.](image)

In case of a sphere impacting a plane surface, Thornton et al.\(^8\) derived the following equations for the coefficient of restitution and the velocity at which the sphere starts to yield \( v_y \):

\[
e = \left( \frac{6 \sqrt{3}}{5} \right)^{1/2} \left( \frac{\sqrt{5}}{2 \sqrt{6}} \right)^{1/4} \left( \frac{v_y}{v_i} \right)^{1/4} = 1.185 \left( \frac{v_y}{v_i} \right)^{1/4}
\]  

\[
v_y = 1.56 \left( \frac{\sigma_y}{\rho v_i^2} \right)^{1/2}
\]

Substituting eq. 5.33 in eq. 5.32 results into:

\[
E^* = 1.754 \left( \frac{\sigma_y}{\rho v_i^2} \right)^{1/4} \left( \frac{1}{e^2} \right)
\]

Here \( E^* \) is the reduced E-modulus (e.g. \( 1/E^* = 1/E_1 + 1/E_2 \) in case of a mixture of two compounds). Knowing the coefficient of restitution (eq. 5.30) and the yield pressure (from eqs. 5.24 and 5.27), the value of \( E^* \) can be calculated.

### 5.4. Experimental set-up

#### 5.4.1. Preparation of the pellets

Pellets were made of microcrystalline cellulose (MCC) (Pharmacel PH101, DMV, Veghel) and different amounts of demineralised water. The pellets were made in a Moulinex coffee
grinder rotating at 200 rps. Liquid was added during 30 seconds by using a syringe. After removing the wall addition, the wet mass was mixed for another 90 seconds. From the wet mass, pellets with a diameter of 2-5 cm were carefully shaped by hand, in such a way that more densification of the pellets was minimised. To obtain pellets with different moisture contents, the pellets were air-dried during periods of different duration (up to 3 hours). In order to obtain a homogeneous liquid distribution, the pellets with the desired moisture content were stored in closed jackets until the experiment was performed. From the dry and wet pellets, the masses and diameters were measured.

5.4.2. Horizontal impact experiments

The experimental set-up of the horizontal impact experiments was partly adapted from Iveson et al.\textsuperscript{4} Compared to the experimental set-up of Iveson et al. no ‘cradle’ was used. A cotton thread was directly laced through the pellets. A schematic diagram of the experimental set-up is given in figure 5.5. The wall was covered with coloured chalk powder in order to measure the deformation area on the pellets.

![Figure 5.5. Experimental set-up of the horizontal impact experiments.](image)

A video camera was used to measure the initial angle and rebound angle of the pellets. From these data the initial height, pellet – plate impact velocity ($v_i$), rebound height, and rebound velocity ($v_r$) were calculated. Each measurement was at least repeated twice.
Because of the swelling properties of microcrystalline cellulose\textsuperscript{9} and the formation of a crystallite gel during pelletisation\textsuperscript{10}, it was experimentally not possible to measure the wet porosity of the pellets and to calculate the saturation of the wet porosity in a reproducible way. In our experiments, we therefore were compelled to use the liquid to solid mass ratio (= moisture content $H$) instead of the saturation of the pellet.

5.4.3. Calculations

The moisture content ($H$) of the pellets was calculated as:

$$H = \frac{m_{wet} - m_{dry}}{m_{dry}}$$\hspace{1cm}(5.35)$$

The deformation rate ($G$) is calculated as:

$$G = \frac{v}{2r_p}$$\hspace{1cm}(5.36)$$

The coefficient of restitution was calculated as the ratio or the rebound velocity and the impact velocity (see eq. 5.30).

The deformed radius could be obtained from the chalk print on the pellet. The coloured-chalk prints on the pellet represent the maximal deformation before rebound (situation at $h_{max}$ in figure 5.4). The volume of deformed material was calculated from the initial wet radius of the pellet and the impact diameter of the pellet using eqs. 5.6 and 5.8. The yield pressure was graphically obtained by plotting the deformed volume (eq. 5.6) versus the deformation energy (eq. 5.31). The slope of this graph represents $1/\sigma_yH$. The reduced E-modulus was then calculated with eq. 5.34.

5.4.4. Scaling effect

Large pellets of about 2 - 5 cm were used in order to make it possible to measure the deformation of the surface of the pellets after impact. It is supposed that these pellets are representative for the small pellets (1 mm) used in pelletisation.

Considering the dimensionless plastic deformation number $K_p$ (eq. 5.24), and the fractional volume deformed ($\theta$), it can be seen that both relations are independent of the pellet size. It therefore is expected that the yield pressure of the wet pellets is also independent of the pellet size. As a consequence, the E-modulus (eq. 5.34) is also a size-independent value. Thus, the yield pressure and E-modulus of the large pellets used in the experimental part of this study are representative for the values found for small pellets during the pelletisation process.
5.5. Results and discussion

5.5.1. Coefficient of restitution

The relationship between the kinetic energy on impact and the coefficient of restitution is given in figure 5.6. The coefficient of restitution varies between 0.17 and 0.33, which is about ten times higher than the coefficients of restitution found by Iveson et al.\textsuperscript{4}

The highest coefficients of restitution are found at the lowest impact energies. Increasing the impact velocity (and consequently the impact energy) results in lower coefficients of restitution. For impact energies > 0.004 Joule, the coefficient of restitution becomes independent of the impact energy.

At low impact energies, a relative high amount of the impact energy is stored as elastic energy, causing the highest rebound (e.g. coefficient of restitution) of the pellet. At higher impact energies, more energy is used to deform the pellet, resulting in a lower coefficient of restitution. The absolute value of the coefficient of restitution differs for varying moisture contents, and depends on the E-modulus as well as the yield pressure of the pellet.

![Figure 5.6. Relationship between the coefficient of restitution and the kinetic energy at impact for different moisture contents. Key: (△) H=0.99, (★) H=1.19, (+) H=1.25, (⊙) H=1.31 and (■) H=1.35.](image)

5.5.2. Yield pressure

Figure 5.7 suggests a linear relationship between the deformation energy and the deformed volume after impact. The inverse slope of the lines in figure 5.7 gives the yield pressure of wet pellets according to the Hawkyard method.

As can be seen in this figure, a linear relationship is found between the deformation energy and the deformed volume, indicating that the yield pressure is independent of the deformation energy. A decrease in yield pressure (e.g. an increase in the slope) is found for increasing moisture contents (figure 5.8). This can be explained by the plasticising effect of water on the MCC pellets, resulting in a decrease in inter-particle friction.
Another explanation can be made in terms of a gel-like network formed by MCC and water, which has been called a crystallite-gel by Kleinebudde\textsuperscript{10}. More water dilutes the network, resulting in a lower network-strength and a lower yield pressure.

The yield pressure of (dry) MCC-powder is about 20 MPa\textsuperscript{11}. The yield pressure of wet MCC-water pellets is about 40 times lower. This can be explained by the plasticising effect of liquid, and the following decrease of the glass transition temperature, resulting in a more rubber-like behaviour of the wet mass at higher moisture content.
5.5.3. Viscous or plastic deformation

In order to make a distinction between viscous deformation and plastic deformation, the relationship between the log($K$)-value and the log($G$) is given in figure 5.9.

![Graph showing log(K) vs log(G) with varying moisture contents](image)

Figure 5.9. Relationship between log($K$) and log($G$) for varying moisture contents. The slopes of the lines represent the plasticity index ($n$). Key: (△) $H=0.99$, (★) $H=1.19$, (+) $H=1.25$, (⊙) $H=1.31$ and (■) $H=1.35$.

In general, a decreasing plasticity index is found at increasing moisture contents (see figure 5.10). At a moisture content of 1, the plasticity index is found to be 1.8, indicating almost pure plastic behaviour. At higher moisture contents, the plasticity index decreases to 1.2 at a moisture content of 1.35, indicating rather more viscous behaviour than plastic behaviour.

![Graph showing plasticity index vs moisture content](image)

Figure 5.10. The relationship between the plasticity index and the moisture content of the wet pellets. The standard deviation is also given.
5.5.4. Reduced E-modulus

The relationship between the reduced E-modulus and the moisture content is given in figure 5.11. The reduced E-modulus of the wet pellets is about 100 times lower than the E-modulus of MCC powder\(^1\) (stored at 76% RH), which is 6.6 GPa. Addition of liquid to MCC powder and a consecutive mixing of the batch results in a decrease of the rigidity of the material. This can be seen in figure 5.1 at \(S > S_c\) and in figure 5.11. As a consequence of the method of calculation of the reduced E-modulus, the influence of the impact velocity is also included. Higher impact velocities cause a little decrease in the reduced E-modulus (see eq. 5.34). Experimentally, a minor influence of the impact velocity on the reduced E-modulus is found.

![Figure 5.11. Relationship between reduced E-modulus and moisture content.](image)

With horizontal impact measurements as introduced by Iveson et al.\(^4\), it is possible to measure material properties of wet pellets (or granules), like the coefficient of restitution, the yield pressure and the E-modulus. In this study, pellets were prepared of a mixture of microcrystalline cellulose (MCC) and water. The strain rates used in this study are comparable to the relative velocities of the pellets inside the high-shear mixer (about 1 m/s, chapter 3).

The coefficient of restitution was found to be about 0.3. Higher impact velocities resulted in lower coefficients of restitution. A restitution of about 30 percent of the original impact velocity is expected in the collisions of the pellets with the stirrer or the wall. The restitution of two colliding pellets is expected to be higher since both pellets are able to store an amount of elastic energy, which is more than the amount of elastic energy stored by one pellet during a pellet-wall collision.

5.6. Conclusions

With horizontal impact measurements as introduced by Iveson et al.\(^4\), it is possible to measure material properties of wet pellets (or granules), like the coefficient of restitution, the yield pressure and the E-modulus. In this study, pellets were prepared of a mixture of microcrystalline cellulose (MCC) and water. The strain rates used in this study are comparable to the relative velocities of the pellets inside the high-shear mixer (about 1 m/s, chapter 3).

The coefficient of restitution was found to be about 0.3. Higher impact velocities resulted in lower coefficients of restitution. A restitution of about 30 percent of the original impact velocity is expected in the collisions of the pellets with the stirrer or the wall. The restitution of two colliding pellets is expected to be higher since both pellets are able to store an amount of elastic energy, which is more than the amount of elastic energy stored by one pellet during a pellet-wall collision.
The estimation of the yield pressure of the pellets is based on the energy conservation model of Hawkyard, and is calculated with the deformed volume and the deformation energy. The estimation of the E-modulus is based on a force-displacement model proposed by Thornton et al.

Our results indicate that pellets deform plastically during preparation in the high-shear mixer. From the experiments it was found that the moisture content is a key variable for the value of the yield pressure as well as the E-modulus (figure 5.8 and figure 5.11). Modelling of the deformation of spherical wet pellets under impact results in a distinction between viscous deformation and plastic deformation. The deformation of wet MCC pellets is plastic at moisture contents $H = 1$ and becomes more viscous at higher moisture contents.

The mean yield pressure of MCC pellets is 0.5 MPa, and the mean reduced E-modulus of MCC pellets is 50 MPa. The yield pressure as well as the E-modulus are highly influenced by the moisture content of the MCC-pellets. The moisture content reduces both the inter-particle friction and the capillary pressure (figure 5.1), resulting in a decreasing yield pressure and E-modulus with increasing moisture content.

Horizontal impact experiments have great potential to help understanding and describing the influence of material properties, like coefficient of restitution, yield pressure and E-modulus, on the granulation process in high-shear mixers.
5.7. Nomenclature

A contact area (m²)
c constant
De deformation number (-)
e coefficient of restitution based on linear velocity differences (-)
E* reduced elasticity modulus (Pa)
f dimensionless length of deformation (-)
F force (N)
G deformation rate (v/dₚ) (1/s)
h height of the deformed layer (m)
H moisture content (mass liquid / mass solid) (-)
K dimensionless constant (-)
m mass (kg)
n plasticity index (-)
r radius (m)
S saturation (-)
St Stokes number (-)
t time (s)
U energy (J)
v velocity (m/s)
V volume (m³)
y height of the formed cylinder (m)

Greek symbols
ε wet pellet porosity (-)
η viscosity (Pa.s)
ρ density (kg/m³)
σᵣ yield pressure (Pa)
θ dimensionless volume of deformation (-)

Subscripts
0 relative
H Hawkyard
i impact
kin kinetic
l liquid
p plastic
p pellet or granule
PV plasticity-viscosity
r rebound
v viscous
y yield
5.8. References
