

Lagrangian modelling of agglomeration during spray drying processes

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Abstract

Numerical calculations of spray drying are rather complex due to the large number of elementary processes affecting the droplet behaviour, the agglomeration and eventually the properties of the product powders. The modelling of spray drying and involved agglomeration processes requires the consideration of the changes of material properties which the dispersed phase is undergoing during the drying procedure. This change of material properties due to drying leads to significant differences of the collision mechanisms of the droplets. Therefore, a Lagrangian model approach is presented based on the distinction of different drying states, namely surface tension and viscous dominated droplets and dry particles. The model accounts for several collision modes between droplets of different drying states, enabling the calculation of the agglomerate properties.

1. Introduction

Spray drying is a process which is widely used in the food and dairy sector, in the pharmaceutical and chemical industry and other areas. It is characterised by the change of material properties of the droplets during the drying process. Up to now, spray drying is mostly handled as a “Black Box” process where the elementary microscale events are not accounted for. So the relations between process conditions, fluid properties and final product (powder) characteristics can rarely be predicted. That leads to a “trial and error” empiric or semi-empiric handling unique for each dryer, for each product and each equipment-layout.

Beside the modelling of the elementary processes of atomisation of fluids and the drying of droplets the description of collision, coalescence and agglomeration mechanisms should enable the conclusion about properties of the final powder, as there are particle size distribution and agglomerate structure. These final powder properties may be used as a base for the judgement of subordinate properties like dispersability and solubility.

Here a Lagrangian model is introduced which describes agglomeration processes of particles of different drying states as occurring during spray drying processes. The approach is intended to be used in the frame of Euler-Lagrange calculations of spray dryers, combined with appropriate tracking, collision and droplet-drying models as well as the consideration of impact efficiency. The agglomeration model itself is taking into account different collision mechanisms by considering different drying states and material properties which droplets face during a spray drying process.

2. Characteristics and material properties

Spray drying is a process applied for liquids which can have very different material properties, as these are product composition, dynamic viscosity, surface tension, density and others. The products might be solutions consisting of a solvent and a dissolved solid component or they might be suspensions consisting of a fluid in which small scale particles are dispersed. The present study is focused on milk products of the food industry. Here we find the milk droplets being composed out of a mixture of dissolved sugar components, proteins and suspended fat globules. During the drying, the solvent evaporates at the surface of the droplets. That leads to a thickening of the solution, to a decrease of the droplet diameter and a change of properties like density, viscosity and surface tension. Especially the change of viscosity is relevant for the outcome of collision events. The creation of more complex agglomerate structures, which are essential for an eventual later dispersion and dissolution behaviour, relies on the collision of higher viscous droplets or collisions of higher viscous droplets with dry particles which do not coalesce completely. So the classification of particles in the considered spray drying process is based on properties which are directly related with to collision behaviour. The following three main basic particle classes are defined.

Surface tension dominated droplets (further referred to as std-droplets):

The collision behaviour of droplets belonging to this class is dominated by the interface surface tension forces. They are distinguished from droplets dominated by viscous forces in terms of the Ohnesorge – number:

$$Oh^2 = \frac{\rho_{Droplet}^2}{d_{Droplet} \cdot \eta_{Droplet} \cdot \sigma_{A,Droplet}} < 1 \quad (1)$$

Note that viscosity, surface tension, density and diameter are functions of the drying state and therefore time-dependent properties.

Droplets dominated by viscous forces (further referred to as vd-droplets):

$$Oh^2 > 1 \quad (2)$$

Investigating drying processes it was observed that the surface tension changes only very slightly. The changes can be related to changes of the density. The dynamic viscosity is the decisive variable to change strongly during the drying process, so a chart can be drawn showing the change of the boundary viscosity between the two regions (*Fig. 1*).

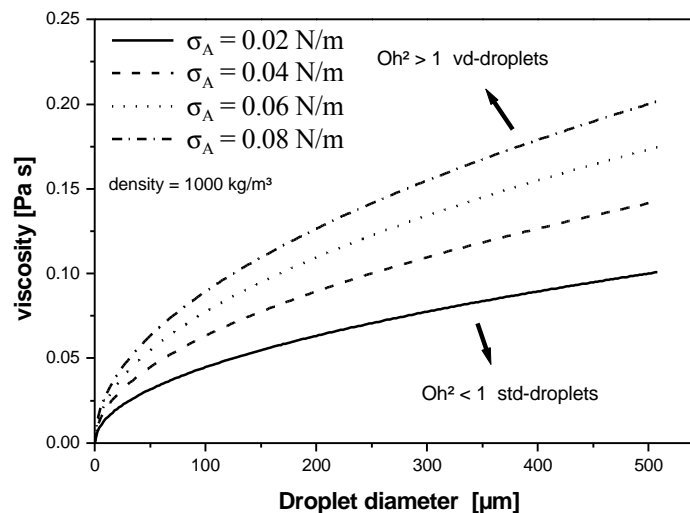


Fig. 1: Boundary between surface tension – and viscous dominated droplets derived from formula 1

Dry particles:

The definition when a particle is considered as dry depends on the material it consists of. If the origin of the droplet is a suspension, the boundary between droplet and dry particle can be determined by the solvent content. Below a certain solvent content the particle is considered as dry. Some fluids which are solutions with high solvent contents change their state to a non-equilibrium amorphous structure. Here the glass transition temperature is usable as a criterion to distinguish between viscous droplets consisting of supercooled liquid, and dry particles whose material is in the amorphous glassy state [6].

3. Modelling of agglomeration processes

Based on the previous classification of particle classes occurring during spray drying processes a number of collision modes can be identified. These collision modes are characterised by different mechanisms which determine the outcome of binary particle impacts. In the frame of the present approach, collisions will occur between the described surface tension dominated (std-) droplets, droplets dominated by viscous forces (vd-droplets) and dry particles. That leads to the following 4 modes of collision mechanisms which are considered in the present model:

Table 1: Collision modes dependent on the particle drying state

std-droplet	-	std-droplet
std-droplet	-	dry / viscous particle
vd-droplet	-	vd / dry particle
dry particle	-	dry particle

The models describing the different collision events should be able to predict the change of droplet sizes, material properties, velocities and probable structure due to impacts.

Collisions between surface tension dominated droplets (std-std)

The model describing impacts of surface tension dominated droplets still has to be capable of considering a range of viscosity, density and surface tension values. These properties change during drying processes and lead to a modified collision behaviour. Such differences were pointed out by previous works on droplet collisions, summarised for example by Orme [4].

The studies of Podvysotsky et al. [5] was the only one considering a range of material property values. This work represents an empirical relation extracted from experiments which were performed with different liquids and various collision conditions.

The empirical relation of Podvysotsky et al. [5] describes a parameter $f_{1,2}$ which represents the mathematical expectation of the ratio between a collector droplet's mass change and the total mass of "projectile" droplets impacting on the collector:

$$E \left(\frac{dm_2}{m_1} \right) = f_{1,2} = 1 - 0.246 \cdot \text{Re}_{1,2}^{0.407} \cdot \text{Lp}_2^{-0.096} \cdot \left(\frac{d_2}{d_1} \right)^{-0.278} \quad (3)$$

Here $\text{Re}_{1,2}$ is the droplet Reynolds number $\text{Re}_{1,2} = \frac{u_{1,2} \cdot d_1 \cdot \rho_1}{\eta}$; Lp_2 the Laplace number

$\text{Lp}_2 = \frac{d_2 \cdot \rho_2 \cdot S_{A,2}}{\eta^2}$ and d_1, d_2 are the diameters of the colliding droplets with $d_2 > d_1$.

Considering the whole collision time t , one can calculate an average mass of the droplets after the collision event which can be expected:

$$m_{2,ac} = m_{2,bc} + \mathbf{f}_{1,2} \cdot m_{1,bc} \quad (4)$$

(ac – after collision
bc – before collision)

The parameter $\mathbf{f}_{1,2}$ should be in a range between $[-1 \leq \mathbf{f}_{1,2} \leq 1]$. According to the authors [5], the correlation is based on experiments in the range $30 \leq \text{Re}_{1,2} \leq 6000$; $5 \leq \text{Lp}_2 \leq 300000$ and $1.9 \leq \frac{d_2}{d_1} \leq 12$.

The velocity of the droplets after the collision event is calculated using the impulse equation under consideration of the exchanged amount of liquid mass. Podvysotsky et al. [5] also gave a correction of the parameter $\mathbf{f}_{1,2}$ for collisions of different liquids.

Collisions between surface tension dominated droplets and viscous or dry particles

For this collision mode the same characteristics can be observed as for std-std droplet collisions. The material properties change with the drying state, which requires a model that accounts for such changes. Here a similar approach as for the std-std collisions is chosen, based on the work of Dubrovsky et al. [1]. The collisions between small droplets and larger solid particles, and small solid particles with larger droplets have to be distinguished. Due to the complexity of introduced empirical relations the reader is referred to the original work [1].

Collisions between droplets dominated by viscous forces

During the drying process droplets of fluids which are typical for spray drying (milk products, flavours, cheese whey etc.) undergo a change of viscosity in the range of a few milli-Pascal seconds up to 10^7 Pascal seconds, in which the latter is a generally accepted value for the occurrence of stickiness and agglomeration related to the phase-states of polymer-like fluids in the glassy or rubbery state [6], [8]. The influence of surface tension on collision processes of droplets can clearly be neglected for such fluids. The collision of viscous droplets with each other or with dry particles is the major mechanism which leads to structured agglomerates during spray drying. Colliding viscous droplets keep roughly their shape and do only partly penetrate into each other. This is a difference to collisions of surface tension dominated droplets, where mostly new spherical droplets are generated in case of coalescence.

For the modelling of viscous droplet collisions different assumptions are made:

- only viscous forces are considered for the collision processes (no influence of surface tension)
- collisions are treated as fully plastic collisions without elastic restitution
- only Newtonian fluids are considered
- the viscous droplets do not change their (spherical) shape during the collision process

The collision process itself is seen as a penetration of the droplet with the higher viscosity into the liquid of the droplet with the lower viscosity. The collision outcome is calculated using a force balance. The movement of the penetrating higher viscous droplet is resisted by the viscous forces created in the liquid of the lower viscous droplet. The force balance is formulated for the Stokesian flow regime, which is justified for high viscous fluids, in a spherical frame of reference, having the origin of coordinates in the centre of the lower viscosity droplet. The coordinates of the centre of the higher viscous partner are given as r , \mathbf{J} and \mathbf{j} and the

components of the relative velocity as u_r , u_J and u_j by transforming them from the global into the spherical frame of reference.

The movement of the higher viscosity body in radial (r) direction is resisted by the Stokesian drag force:

$$m_{High} \cdot \frac{du_r}{dt} = -3 \cdot \mu \cdot m_{Low} \cdot d_{contact} \cdot u_r \quad (5)$$

The penetration in radial direction can be calculated as

$$dr = u_r \cdot dt \quad (6)$$

The values of the lower viscosity droplet are referred to as “low”, that of the droplet with the higher viscosity as “high”. u_r is the time-dependent radial velocity in the spherical frame of reference, μ is the dynamic viscosity, $d_{contact}$ the time-dependent diameter of the contact area between the two droplets. This diameter depends on the penetration depth:

$$Penetration = \frac{d_{High} + d_{Low}}{2} - r \quad (7)$$

The diameter of the contact area is estimated by:

$$d_{contact} = 2 \cdot \sqrt{h \cdot d_{Low} - h^2} \quad \text{with} \quad \begin{aligned} h &= Penetration & \text{if } r > 0 \\ h &= \frac{d_{Low}}{2} + r & \text{if } r \leq 0 \end{aligned} \quad (8)$$

Moreover, it is assumed that the movement in the two tangential directions is resisted by the viscous shear force:

$$m_{High} \cdot \frac{du_{\Lambda}}{dt} = -m_{Low} \cdot \frac{\mu}{4} d_{contact}^2 \cdot \frac{u_{\Lambda}}{\left| \frac{d_{Low}}{2} - h \right|} \quad (9)$$

$$d\Lambda = \frac{u_{\Lambda} \cdot dt}{r} \quad (10)$$

with Λ as alias for the angles J and j .

The penetration and all values u , $d_{contact}$, r , J and j and h are time-dependent variables and therefore the equations are solved numerically. The result of a vd-vd collision depends on the penetration depth and the dissipation of the relative kinetic energy. They are classified as:

- Separation: - The distance between the droplet centres raises above $\frac{d_{High} + d_{Low}}{2}$, and the relative velocity is still above zero.
- Coalescence: - The penetration exceeds 50% of the smaller droplet diameter. At the same time the whole relative kinetic energy is dissipated.
- Agglomeration: - The penetration depth does not exceed 50% of the smaller droplet diameter and the entire relative kinetic energy is dissipated.

After the completion of the time-dependent penetration process the remaining velocity components are transformed back into the global frame of reference.

Collisions between dry particles

In case that two dry particles collide with each other the model described by Ho and Sommerfeld [3] is applied. A critical velocity, calculated out of an energy balance, is used as criterion whether agglomeration occurs or not. In the present case only Van-der-Waals forces are taken into account, so the critical velocity is calculated according to Hiller [2]:

$$u_{crit} = \frac{1}{d_{real}} \cdot \frac{\sqrt{1-e^2}}{e^2} \cdot \frac{A}{\mathbf{p} \cdot \mathbf{z}_0^2 \cdot \sqrt{6 \cdot p_{pl} \cdot \mathbf{r}}} \quad (11)$$

Here e is the energetic restitution coefficient, A the Hamaker constant, z_0 the contact distance and p_{pl} the material limiting contact pressure. An agglomerate is formed if the normal relative velocity between the impacting particles is less than the critical velocity u_{crit} :

$$|\vec{u}_{rel}| \cdot \cos \mathbf{f} \leq u_{crit} \quad (12)$$

\mathbf{f} is the impact angle, \vec{u}_{rel} the relative velocity vector between both particles. Due to the short range of the Van-der-Waals forces agglomeration events can only be expected for dry particles smaller than 20 μm .

The post collision velocity for the case of rebound is calculated by solving the momentum equation distinguishing between sliding and non-sliding collision:

condition for non-sliding collision:
$$\frac{u_{real}}{v_{real}} < \frac{7}{2} \cdot \mathbf{m}_{friction} \cdot (1+e) \quad (13)$$

post collision velocity u-component:
$$u_{real}^* = u_{real} \cdot \left(1 - \frac{1+e}{1 + \mathbf{m}_{real}/\mathbf{m}_{fict}} \right) \quad (14)$$

v-component sliding collision

$$v_{real}^* = v_{real} \cdot \left(1 - \mathbf{m}_{friction} \cdot (1+e) \cdot \frac{u_{real}}{u_{fict}} \cdot \frac{1}{1 + \mathbf{m}_{real}/\mathbf{m}_{fict}} \right) \quad (15)$$

v-component non-sliding collision

$$v_{real}^* = v_{real} \cdot \left(1 - \frac{2/7}{1 + \mathbf{m}_{real}/\mathbf{m}_{fict}} \right) \quad (16)$$

Combined collision and agglomeration model

The agglomeration model is embedded into an Euler-Lagrange approach, where a number of particles are tracked in a fluid flow field and a stochastic collision model is applied. This approach is described in detail by Sommerfeld [7]. Here, only the coupling of the agglomeration and collision model will be discussed.

This stochastic collision model uses fictitious collision partners which are created out of the local particle population by a statistical approach. The collision probability for a collision of the real particles with their fictitious counterparts is calculated according to kinetic theory of gases. The specific characteristics of spray drying processes require the sampling of a number of statistical values for each control volume. These are the number frequency distribution discretised in size-classes (i.e. particle size distribution), the mean and rms velocity components, the mean and rms

values of the particle temperatures, the number fraction of dry particles and the mean and rms values of the solvent content of the droplets (not counting the dry particles). These values have to be sampled for each size class of the particle population.

During each time step of the particle tracking procedure a fictitious collision partner with its properties is sampled. The fictitious particle size is determined by help of a random process in which first the particle size class is estimated and second the fictitious particle diameter is sampled. The mean velocity of the fictitious particle corresponds to the mean velocity of its size class. The fluctuation velocity is correlated with the fluctuation velocities of the flow field and that of the potential real collision partner (see Sommerfeld [7]). The temperature of the fictitious particle is calculated as

$$T_{fict} = \bar{T}_i + GRN \cdot \mathbf{s}_{T,i} \quad (17)$$

For that the mean temperature \bar{T}_i of the actual size class i and the temperature rms-value $\mathbf{s}_{T,i}$ of this size class are used. GRN is a gaussian random number sampled from a distribution which has a mean value of zero and a standard deviation of 1.

Further in the calculations the drying state of the fictitious particle has to be determined. Here the number fraction of dry particles is used. If a random number sampled from a uniform distribution in the range [0...1] is smaller than the number fraction of dry particles of that size class, the fictitious particle is dry, having a solvent content zero. Otherwise the fictitious particle is a droplet.

The droplets solvent content SC is now calculated as:

$$SC_{fict} = \bar{SC}_i + GRN \cdot \mathbf{s}_{SC,i} \quad (18)$$

Using the temperature and the solvent content, other material properties are estimated out of property functions which depend on these two values. These property functions, applied for density, dynamic viscosity and surface tension, are dependent on the fluid of the droplets and have to be known.

The collision probability of the real and fictitious particle is calculated using the time step size Δt and the collision frequency f_{coll} :

$$P_{coll} = f_{coll} \cdot \Delta t = \frac{\mathbf{p}}{4} \cdot (d_{real} + d_{fict})^2 \cdot |\vec{u}_{real} - \vec{u}_{fict}| \cdot n_d \cdot \Delta t \quad (19)$$

If a random number, again created out of a uniform distribution [0...1], is smaller than the collision probability, a collision between the real and fictitious particles will take place:

$$RN < P_{coll} \quad (20)$$

Depending of the drying states of the colliding partners, the appropriate coalescence/agglomeration model is chosen to calculate the post collision properties. The point of impact on the surface of the fictitious particle is sampled randomly using the procedure described by Sommerfeld [7].

In case of coalescence or agglomeration, the information of the primary particles is kept. The agglomerate values like equivalent diameter and agglomerate mass are stored using a separate agglomeration population. Further, the tracking of the merged primary droplets or particles is stopped, instead the new agglomerate is tracked furtheron, but the drying is still continued for the primary particles inside the agglomerates.

4. Conclusions

In the present paper an agglomeration model in the frame of an Euler-Lagrange approach is introduced. The model is based on the knowledge of drying states of multi-component droplets which can be observed during spray drying processes and which are crucial for the collision

The combined tracking, drying, collision, coalescence and agglomeration models should enable the prediction of powder properties after a spray drying process. Further extensions concerning the prediction of agglomerate structure and agglomerate strength are under work.

5. Nomenclature

6. References

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