# MATHEMATICAL MODELLING OF GRANULATION PROCESSES

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# Abstract

Granulation is an industrial process where fine particles are bound together into larger granules. The process has numerous applications including the manufacture of pharmaceuticals and the production of cosmetics, chemicals, detergents and fertilisers. This thesis studies aspects of wet granulation which involves the application of a viscous binder, usually in the form of a spray, to an agitated bed of powder particles. Individual powder particles may adhere together, joined by small quantities of binder fluid called liquid bridges. By a process of collision and adherence additional particles may join the newly formed agglomerates. Agglomerates may also coalesce together which is a process that leads to granule formation. On the completion of this process, granules are typically dried.

This thesis studies wet granulation on three different levels. First, micro-level investigations of liquid bridges between two and three particles are performed. For the two-particle case, the fluid profile of static (stationary) and dynamic (moving) liquid bridges is investigated. For the static case, a numerical solution to the Young-Laplace equation is obtained; this relates the volume of binder fluid to liquid bridge properties such as the inter-particle force. An analytic solution is also obtained, providing the liquid bridge profile in terms of known mathematical functions. For both solutions, the radii of the (spherical) primary particles may be different. The dynamic case is then studied using the Navier-Stokes equations with the low Reynolds number approximation. The motion of the approaching particles is shown to be damped by the viscosity of the liquid bridge. Static liquid bridges between three equally sized primary particles are then studied. Symmetry of the problem is used to obtain a numerical solution to the Young-Laplace equation. Liquid bridge properties are calculated in terms of the binder fluid volume. Experimental agreement is provided.

Secondly, a model to estimate the stickiness (fractional wet surface area) of agglomerates is proposed. Primary particles are approximated as spheres and are added one at a time in a closely packed arrangement. The model includes parameters to control the inter-particle separation distance and the fluid saturation state. Computational geometry is used to obtain results which relate the number of particles and the volume of binder fluid to the stickiness of the agglomerates.

Finally, a population balance model for wet granulation is developed by extending an earlier model to incorporate the effects of binder fluid. Functions for the inter-particle collision rate and drying rate are proposed, including functions which are derived from the geometric model, described above, for the case of maximum particle consolidation. The model is solved numerically for a range of coalescence kernels and results are presented which show the effect of binder volume and the drying rate.

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# Nomenclature

Constants and variables are defined when they first appear in the text. Commonly used variables from Chapters 2 to 5 are listed below.

## Chapter 2 Variables (Liquid Bridges Between Two Particles)

**Static**

| Variable | Description |
|---|---|
| $H_0$ | Mean curvature |
| $\Delta p$ | Pressure difference |
| $\gamma_{lv}$ | Fluid surface tension |
| $r$ | Vertical coordinate |
| $x$ | Horizontal coordinate |
| $r_1$ | Radii of curvature in the $r-x$ plane |
| $r_2$ | Radii of curvature in the $r-y$ plane |
| $r_0$ | Bridge height at $x=0$ |
| $\theta$ | Contact angle |
| $\alpha$ | Half angle for particle 'A' |
| $\beta$ | Half angle for particle 'B' |
| $\sigma$ | Scaling variable |
| $R$ | Non-dimensional bridge vertical coordinate |
| $X$ | Non-dimensional bridge horizontal coordinate |
| $R_0$ | Non-dimensional bridge height at $X=0$ |
| $\Delta P$ | Non-dimensional pressure difference |
| $S$ | Non-dimensional inter-particle separation distance |
| $V$ | Non-dimensional liquid bridge volume |
| $A$ | Non-dimensional liquid bridge surface area |
| $F$ | Non-dimensional inter-particle binding force |
| $E$ | Non-dimensional Gibbs free surface energy |
| $C$ | Non-dimensional normalised force $\left(C=\frac{F}{2\pi}\right)$ |

**Dynamic**

| Variable | Description |
|---|---|
| $t$ | time |
| $r$ | Vertical coordinate |
| $\mathbf{R}$ | Sphere radius |
| $z$ | Vertical Coordinate |
| $h$ | Separation function |

**Variables used in Chapter 2 (continued)**

| | |
|---|---|
| $h_0$ | Closest separation |
| $\vec{v}$ | Velocity vector |
| $\rho$ | Fluid density |
| $g$ | Acceleration due to gravity |
| $\mu$ | Dynamic Viscosity |
| $P$ | Pressure within liquid bridge |
| $P_{\text{amb}}$ | Ambient pressure |
| $\overline{P}$ | Vertically averaged pressure |
| Re | Reynolds number |
| $F_{\text{bridge}}$ | Force |
| $V_0$ | Constant bridge volume |

## Chapter 3 Variables (Static Liquid Bridges Between Three Particles)

| **Variable** | **Description** |
|---|---|
| $H_0$ | Mean curvature |
| $\Delta p$ | Pressure difference |
| $\gamma$ | Fluid surface tension |
| $(X, Y, Z)$ | Cartesian coordinates |
| $(r, \theta)$ | Cylindrical coordinates |
| $z$ | Liquid bridge surface in cylindrical coordinates |
| $\mathbf{Z}$ | Liquid bridge surface in Cartesian coordinates |
| $\alpha$ | Contact angle |
| $\delta$ | Half-filling angle |
| $r_0$ | Central point of liquid bridge |
| $r_s$ | Intersection between the sphere and fluid |
| $r_p$ | Intersection between the symmetry plane and fluid |
| $C_1$ | Contour of the three-phase contact line |
| $C_2$ | Contour of the symmetry plane |
| $a$ | Sphere radius |
| $S$ | Inter-particle separation distance |
| $n_{\text{surface}}$ | Fluid surface outward pointing normal vector |
| $n_{\text{sphere}}$ | Sphere outward pointing normal vector |
| $n_{\text{sym}}$ | Symmetry plane outward pointing normal vector |
| $\hat{z}$ | Mesh approximation to $z$ |
| $V$ | Liquid bridge volume |

**Variables used in Chapter 3 (continued)**

| | |
|---|---|
| $S$ | Liquid bridge surface area |
| $F$ | Inter-particle binding force |

## Chapter 4 Variables (Modelling the Agglomeration Process)

| Variable | Description |
|---|---|
| $S$ | Matrix containing coordinates of primary particles |
| $\sigma$ | Minimum separation distance between sphere centres |
| $\delta$ | Fluid saturation parameter |
| $T$ | Tetrahedra matrix |
| $F$ | Face matrix |
| $\kappa$ | Skewness number |
| $\Xi$ | Objective function for optimisation |
| $\mathbf{S_i}$ | Polyhedron representation of the sphere centred at $S_i$ |
| $\mathbf{T_i}$ | Polyhedron representation of the expanded tetrahedron centred at $T_i$ |
| $V_{\text{binder}}$ | Binder volume |
| $A_{\text{binder}}$ | Agglomerate wet surface area |
| $A_{\text{particle}}$ | Agglomerate dry surface area |
| $W$ | Surface wetness |
| $V^*$ | Fluid-to-solid volume ratio |
| $N$ | Agglomerate size (number of primary particles) |
| $V_{\text{solid}}$ | Volume of a primary particle |
| $A_{\text{solid}}$ | Surface area of a primary particle |

## Chapter 5 Variables (Population Balance Modelling)

| Variable | Description |
|---|---|
| $N$ | Total number of particles |
| $i$, $j$ | Particle size |
| $n_i$ | Number of particles of size $i$ |
| $K_{i,j}$ | Coalescence kernel (in the absence of binder) |
| $K_0$ | Size independent component of the coalescence kernel |
| $M$ | Total mass of particles |
| $m_i$ | Mass of particles of size $i$ |
| $N_0$ | Initial number of particles |

**Variables used in Chapter 5 (continued)**

| | |
|---|---|
| $t$ | Time |
| $\overline{i}$ | Mean particle size |
| $\sigma$ | Variance |
| $b_i$ | Wet binder mass for all particles of size $i$ |
| $B_i$ | Wet binder mass of a particle of size $i$ |
| $\overline{B}$ | Total wet binder mass |
| $\Phi_{i,j}$ | Sticking efficiency function |
| $\Phi_0$ | Size independent component of the sticking efficiency function |
| $\hat{K}_{i,j}$ | Coalescence kernel (in the presence of binder) |
| $D_i$ | Drying rate of particles of size $i$ |
| $D_0$ | Drying rate constant |
| $\frac{D_0}{K_0}$ | Collision-to-drying ratio |
| $s_{\max}$ | Maximum particle size |

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