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MATHEMATICAL MODELLING OF
GRANULATION PROCESSES

A THESIS PRESENTED IN PARTIAL
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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
Δp	Pressure difference
γ_{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 $x = 0$
θ	Contact angle
α	Half angle for particle 'A'
β	Half angle for particle 'B'
σ	Scaling variable
R	Non-dimensional bridge vertical coordinate
X	Non-dimensional bridge horizontal coordinate
R_0	Non-dimensional bridge height at $X = 0$
ΔP	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 ($C = \frac{F}{2\pi}$)

Dynamic

Variable	Description
t	time
r	Vertical coordinate
R	Sphere radius
z	Vertical Coordinate
h	Separation function

Variables used in Chapter 2 (continued)

h_0	Closest separation
\vec{v}	Velocity vector
ρ	Fluid density
g	Acceleration due to gravity
μ	Dynamic Viscosity
P	Pressure within liquid bridge
P_{amb}	Ambient pressure
\bar{P}	Vertically averaged pressure
Re	Reynolds number
F_{bridge}	Force
V_0	Constant bridge volume

Chapter 3 Variables (Static Liquid Bridges Between Three Particles)

Variable	Description
H_0	Mean curvature
Δp	Pressure difference
γ	Fluid surface tension
(X, Y, Z)	Cartesian coordinates
(r, θ)	Cylindrical coordinates
z	Liquid bridge surface in cylindrical coordinates
\mathbf{Z}	Liquid bridge surface in Cartesian coordinates
α	Contact angle
δ	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_{surface}	Fluid surface outward pointing normal vector
n_{sphere}	Sphere outward pointing normal vector
n_{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
σ	Minimum separation distance between sphere centres
δ	Fluid saturation parameter
T	Tetrahedra matrix
F	Face matrix
κ	Skewness number
Ξ	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_{binder}	Binder volume
A_{binder}	Agglomerate wet surface area
A_{particle}	Agglomerate dry surface area
W	Surface wetness
V^*	Fluid-to-solid volume ratio
N	Agglomerate size (number of primary particles)
V_{solid}	Volume of a primary particle
A_{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
\bar{i}	Mean particle size
σ	Variance
b_i	Wet binder mass for all particles of size i
B_i	Wet binder mass of a particle of size i
\bar{B}	Total wet binder mass
$\Phi_{i,j}$	Sticking efficiency function
Φ_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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