Powder Flow In Vertical High Shear Mixer Granulators

Anh Lan Ho Tran
B.Eng (Hons)

A thesis submitted for the degree of Doctor of Philosophy at The University of Queensland in 2015
School of Chemical Engineering
Abstract

An integral component in understanding the fundamental mechanisms of granulation is the **powder flow behaviour** within mixer granulators. All three classes of granulation rate processes: wetting and nucleation, consolidation and coalescence, and attrition and breakage require detailed knowledge of flow fields in mixing systems to enable rational approaches to the prediction of granule attributes. The three objectives of this thesis are to 1) develop a powder flow regime map to characterise the powder flow field, 2) investigate the effect of impeller speed, powder cohesion and blade-rake angle on powder flow and mixing behaviour and, 3) outline a framework for compartment models of vertical-axis high shear mixer (vHSM) granulators.

In the development of the powder flow regime map, two distinct regimes were identified. Ideal **roping regime** occurs at high impeller speeds when the rotating blades pushes the powder out and up the mixer wall, and then back down the free surface forming the “roping” flow pattern. This stable flow behaviour promotes high vertical bed turnover which is essential for controlled granulation. In this regime, the powder in the blade region has sufficient rotational inertia to overcome gravitational forces (i.e. powder Froude number greater than unity). If this criterion is not met, the mixing system remains in the undesirable **bumping regime**. This regime is characterised by poor bed turnover and non-uniform shear distribution within the powder bed. In the bumping regime, multiple transitional states are observed with increasing impeller speeds for this particular system, which has not been fully characterised in the literature before. An explanation for the observed **apparent roping behaviour** based on bed resonance is given. These intermediary transitional states can be differentiated by blade-powder interactions.

From these observations, a powder flow regime map for vertical-axis mixer granulators is proposed based on two dimensionless groups: **powder Froude number**, the ratio of the powder rotational inertial to gravitational forces and the **Bed Resonance number**, which characterises the interaction of the bulk motion of the powder bed with the passage of impeller blades beneath it. The regime map was validated by experimental results utilising
the well-established, non-invasive tracking technique of Positron Emission Particle Tracking (PEPT) and torque measurements.

Investigations into the effect of impeller speed, powder cohesion and blade-rake angle on powder flow and mixing behaviour were conducted using PEPT. Results showed that powder velocity, bed porosity and shear rates varied spatially within the mixer granulator. Studies of the stable roping regime revealed that the average powder velocity in the impeller zone was 1.66 m/s (≈25% of the impeller tip speed). In comparison, the average powder velocity in the circulation and surface zone was approximately half of the impeller zone at 0.9 and 0.74 m/s, respectively (12% and 11% of the impeller tip speed). With the addition of moisture (cohesion) to the dry powder, the average velocity of the powder decreases by 10%. Increasing blade-rake angle from 10 to 90 degrees increased the average powder velocity by approximately 38%. Vertical mixing improved with higher blade-rake angle due to an increased thickness of the shear transmission layer however no significant improvements were observed with changes in bed cohesion.

Compartment models are developed to reduce the highly complex flow field into spatial similar phenomenological compartments. A general framework is outlined to describe the methodology used to identify compartments and powder exchange rates between compartments. A simple two-compartment model was proposed for a system in which growth and breakage occurred simultaneously. The mixing vessel was partitioned into two compartments, the impeller zone and the circulation zone. Expanding on this, a more complex multi-compartment model was developed to incorporate wetting and nucleation processes into a third compartment defined as the spray zone. Both models represented reasonable approximations to the experimental data. These fitted models, in combination with population balance modelling should provide a useful tool to predict wet granulation processes as well as a rational approach to controlling final granule attributes.
Declaration by author

This thesis is composed of my original work, and contains no material previously published or written by another person except where due reference has been made in the text. I have clearly stated the contribution by others to jointly-authored works that I have included in my thesis.

I have clearly stated the contribution of others to my thesis as a whole, including statistical assistance, survey design, data analysis, significant technical procedures, professional editorial advice, and any other original research work used or reported in my thesis. The content of my thesis is the result of work I have carried out since the commencement of my research higher degree candidature and does not include a substantial part of work that has been submitted to qualify for the award of any other degree or diploma in any university or other tertiary institution. I have clearly stated which parts of my thesis, if any, have been submitted to qualify for another award.

I acknowledge that an electronic copy of my thesis must be lodged with the University Library and, subject to the General Award Rules of The University of Queensland, immediately made available for research and study in accordance with the Copyright Act 1968.

I acknowledge that copyright of all material contained in my thesis resides with the copyright holder(s) of that material. Where appropriate I have obtained copyright permission from the copyright holder to reproduce material in this thesis.
Publications during candidature

Conference papers


Publications included in this thesis

No publications included.
The PEPT experiments would not have been possible without the technical laboratory support of Professor David Parker and Dr Xianfeng Fan who assisted in the preparation of the radioactive tracer particles and operation of the PEPT equipment.
Statement of parts of the thesis submitted to qualify for the award of another degree

None.
Acknowledgements

This research could not have been possible without the help and support of many people. Firstly, I would like to thank my supervisors Professor Jim Litster, A/Professor Tony Howes and Professor Jonathan Seville - all of whom have contributed valuable ideas, discussions, support, guidance and patience for the successful completion of this work.

Thanks must also be extended to the particle groups at the University of Queensland and the University of Birmingham in which lively debate was encouraged. In particular I would like to thank technical input from Dr Rachel Mansa, Dr Martijn van der Hoeven, Dr Stephan Tait, Dr Richard Dombrowski, Dr Michael Wallis, Dr Rachel Smith, Dr Lian Liu, Dr Andy Ingram, Dr Phil Robbins and Dr Serafim Bakalis.

Technical laboratory work would not be possible without Dr Xianfeng Fan and Professor David Parker who assisted with the PEPT experiments and David Page with particle characterisation.

I also wish to thank my mum, dad, my siblings Nhi and Hung for their unwavering love and support and to my future - Alex and Lara.

Lastly, my biggest thanks goes to my schkuet husband Dinis who has taught me all I know about MATLAB programming and most importantly, for the sanity check.
Keywords

powder flow, particulate flow, granulation, high shear mixer, granulator, Positron Emission Particle Tracking (PEPT), powder flow regime map, compartment model, powder Froude number, Bed Resonance number

Australian and New Zealand Standard Research Classifications (ANZSRC)

ANZSRC code: 090406, Powder and Particle Technology, 90%
ANZSRC code: 111504, Pharmaceutical Sciences, 10%

Fields of Research (FoR) Classification

FoR code: 0904, Chemical Engineering, 90%
FoR code: 1115, Pharmacology and Pharmaceutical Sciences, 10%
Abstract ii
Declaration by author iv
Publications during candidature v
Acknowledgements ix
Keywords x
List of Figures xv
List of Tables xxi
List of Abbreviations xxiii

1 Introduction 1
1.1 Background ................................................. 1
1.2 Granulation Mechanisms .................................. 1
1.3 Powder flow characteristics for granulation ................. 5
1.4 Granulation Equipment ...................................... 6
1.5 Flow visualisation and modelling ............................ 8
1.6 Thesis Objectives ........................................... 9
1.7 Thesis Outline ............................................. 9

2 Literature Review 11
2.1 Introduction .................................................. 11
2.2 Powder Flow Theory and Regimes .......................... 11
2.3 Powder flow studies ......................................................... 14
  2.3.1 Powder flow regime map ........................................... 16
  2.3.2 Effects of different parameters on powder flow behaviour .......... 21
2.4 Techniques of studying powder flow .................................. 23
2.5 Advances in computational powder flow models ....................... 25
2.6 Critical summary ......................................................... 27

3 Materials and Methodology .............................................. 28
  3.1 Introduction ........................................................... 28
  3.2 Materials ............................................................... 28
    3.2.1 Selection of model powders ...................................... 28
    3.2.2 Powder properties ................................................ 29
    3.2.3 Binder properties ............................................... 30
    3.2.4 Bulk powder properties ........................................ 30
  3.3 Mixer design, construction and control ................................ 34
  3.4 Torque measurements ................................................ 37
  3.5 Positron Emission Particle Tracking (PEPT) Experiments ............. 39
    3.5.1 Ergodicity criterion ............................................ 40
    3.5.2 Tracer segregation ................................................ 40
    3.5.3 Limitations of PEPT detection and resolution ................... 41
  3.6 PEPT Data Analysis .................................................... 42
    3.6.1 Preprocessing of data ....................................... 42
    3.6.2 Single particle trajectory .................................... 43
    3.6.3 Lagrangian properties ....................................... 43
    3.6.4 Eulerian maps .................................................. 44
    3.6.5 Occupancy ....................................................... 44
    3.6.6 Bed density ..................................................... 48
    3.6.7 Eulerian velocities ......................................... 48
    3.6.8 Fourier analysis ................................................. 48
    3.6.9 Shear rates ...................................................... 49
    3.6.10 Mixing ........................................................... 50
  3.7 Summary ............................................................... 50

4 Powder Flow Regime Map ............................................... 52
  4.1 Introduction ........................................................... 53
    4.1.1 Powder flow dimensionless groups ............................. 54
## Contents

### 4.2 Proposed Powder Flow Regime Map ................................................. 55
### 4.3 Experimental methodology .......................................................... 59
  4.3.1 PEPT experiments ........................................................................... 59
  4.3.2 Torque experiments ......................................................................... 59
### 4.4 Results and discussion ................................................................. 60
  4.4.1 Fourier Analysis ............................................................................ 61
  4.4.2 Motion of particles ......................................................................... 63
  4.4.3 Bump height .................................................................................. 65
  4.4.4 Velocity profile .............................................................................. 67
  4.4.5 Velocity vector maps ..................................................................... 70
  4.4.6 Occupancy maps ........................................................................... 72
  4.4.7 Mixing .......................................................................................... 77
  4.4.8 Shear rates .................................................................................... 79
### 4.5 Validation of regime map ............................................................... 79
  4.5.1 Torque measurements of bumping regime ....................................... 81
  4.5.2 Effective shear thickness on roping regime ..................................... 83
### 4.6 Relationship for measurable regime map parameter ....................... 83
### 4.7 Potential application of regime map to granulation ....................... 85
### 4.8 Conclusion ...................................................................................... 85

### 5 The effects of cohesion and blade-rake angle on powder flow and mixing 88
  5.1 Introduction ....................................................................................... 89
    5.1.1 Effect of cohesion on powder flow and mixing ............................... 89
    5.1.2 Effect of blade-rake angle on powder flow and mixing .................. 90
### 5.2 Experimental procedure ............................................................... 90
### 5.3 Results and discussion ................................................................. 91
    5.3.1 Powder flow ................................................................................ 91
    5.3.2 Velocities .................................................................................... 93
    5.3.3 Bed occupancy ............................................................................ 95
    5.3.4 Shear rates ................................................................................ 96
    5.3.5 Mixing ....................................................................................... 97
### 5.4 Implications for granulation ........................................................... 99
### 5.5 Conclusions .................................................................................... 101

### 6 Compartment models ......................................................................... 103
  6.1 Introduction ....................................................................................... 104
Contents

6.2 Model development and theory ........................................ 105
   6.2.1 General framework ........................................... 105
   6.2.2 Defining compartments ....................................... 106
   6.2.3 Global mixing properties ..................................... 109
   6.2.4 Sub-compartment model generation ............................ 110
   6.2.5 Deriving model parameters .................................... 111

6.3 Experimental method .................................................. 112
   6.3.1 Positron Emission Particle Tracking (PEPT) experiments .. 112

6.4 Results and discussion ............................................... 114
   6.4.1 Case study: Two-compartment model (two-CM) ............ 114
   6.4.2 Case study: Multi-compartment mixing model (multi-CM) .. 121

6.5 Implications for granulation processes ............................. 123
6.6 Limitations ............................................................ 123
6.7 Conclusion ............................................................. 124

7 Conclusions and Recommendations .................................... 126
   7.1 Summary ............................................................ 126
   7.2 Original contributions ............................................ 128
   7.3 Future work ......................................................... 128

References ................................................................. 130
## List of Figures

1.1 Granulation processes as classified by Ennis and Litster (1997) .......................... 2

1.2 Nucleation regime map developed by Hapgood et al. (2003) where $\tau_p$ is the dimensionless drop penetration time and $\Psi_a$ is the dimensionless spray flux. .......................... 3

1.3 Growth regime map developed by Iveson et al. (2001a) where $\rho_g$ is the granule density, $U_c$ is the representative collision velocity in the granulator, $Y_g$ is the granule dynamic yield stress, $w$ is the mass ratio of liquid to solid, $\varepsilon_{min}$ is the minimum porosity the formulation and $\rho_s$ and $\rho_l$ is the solid particles and liquid density, respectively. ............................................................... 3

1.4 Design model approach to granulation. ................................................................. 4

1.5 Examples of industrial high shear mixer granulators: a & b) Aeromatic-Fielder (Litster et al., 2002, Plank et al., 2003), c) Niro Pellmix (Knight et al., 2000, Schaefer et al., 1993), d) Collette Gral (Faure et al., 1999), e) Zanchetta (Nilpawar et al., 2006), f) custom design vHSM (Tu et al., 2009, Knight et al., 2001, Bridson et al., 2007), g) planetary mixer (Hiseman et al., 2002), and h) cyclomix mixer granulator (Ng et al., 2007a) ................................................................. 7

1.6 Custom designed cylindrical flat bladed vHSM by a) Stewart et al. (2001a) and b) Knight et al. (2000) ................................................................. 8

2.1 Granulator and reactor scale approach (Tardos et al., 2003) .............................. 13

2.2 Schematic diagram of a) bumping regime and b) roping regime Litster et al. (2002) ................................................................. 18

2.3 Effect of impeller speed on powder surface velocity (Litster et al. (2002)) .... 18

2.4 Effect of impeller speed on torque (Wellm, 1997) .............................................. 20
2.5 Mixer granulator regime map for flows operating in the roping regime where $K_1$ is the ratio of mean particle velocity $\bar{U}_p$ to impeller tip speed $U_i$ and $K_2$ is the ratio of the shear transmission height $\delta$ to mixer radius $R$. Flow regimes are defined in terms of a dimensionless shear rate: i) frictional, $\dot{\gamma}^* < 0.2$; ii) intermediate, fluid-like; and iii) rapid collisional, $\dot{\gamma}^* > 3$ (Mort, 2009).

2.6 Surface velocity with the addition of binder (Plank et al., 2003)

2.7 Compartment model of vertical-axis mixer by Freireich (2010)

3.1 Viscosity of 0.1 Pa.s silicone oil over varying temperatures.

3.2 Determination of powder tapped density a) schematic drawing b) Copley JV1000 setup.

3.3 a) Shear stress plot and resulting yield locus as constructed with a ring shear tester and Mohr stress circles ($\sigma_1$ consolidation stress, $\sigma_c$ unconfined yield strength) from (Schulze, 2006).

3.4 Photographs of a) the high shear mixer granulator and PEPT set-up, b) side view of the vHSM, c) mixer bowl and connector shaft assembly and d) mixer motor.

3.5 Photographs of a) the control box and wiring b) top view of mixer bowl and torque arm, c-d) ball bearing flowing assembly, e) s-bend torque arm measuring device, f) torque reader, g) side view of 0.21 m mixing bowl and h) top view of mixer bowl with rectangular impeller blade.

3.6 Schematic isometric views of mixing vessel with torque arm. Dimensions are in mm.

3.7 Photographs of impeller blades a) top view and b) front view and schematic isometric views of the c) 90° rectangular two-bladed impeller and d) 10° triangular two-bladed impeller. Dimensions are in mm.

3.8 Positron emission particle tracking in a mixer granulator: 1) motor and control box, 2) mixer vessel, 3) impeller, 4) torque arm, 5) PEPT tracer, 6) powder bed, 7) gamma-ray detectors.

3.9 The presence of dead zones showed that the ergodicity criteria was not achieved using dry sand with either the a) rectangular or b) triangular impeller at low impeller speed of 100 rpm. The blade boundary is indicated by $\ldots \ldots$.

3.10 Geometric efficiencies (Seville et al., 2009).

3.11 Tracer experiments at a) 33 rpm and b) 566 rpm.

3.12 Typical time data series of the Cartesian coordinates of PEPT experiments.
3.13 The vertical bed turnover frequency was determined by the rotational frequency at which the tracer moved over a certain threshold height. The dots represent the maximum and minimum position of each vertical rotation cycle in the a) radial-axial plane and b) axial position versus time plot. ............................................ 46
3.14 Velocity gradient method used to estimate velocity component from a tracer trajectory in each coordinate direction. ................................................................. 46
3.15 Generating a virtual cluster of particles from a single particle trajectory using the a) box method and b) the constant time interval method from Doucet et al. (2008) ........................................................................................................... 47
3.16 Diagram of 2D shear rate calculations. ................................................................. 49
4.1 Schematic diagram of the powder flow regime showing the interaction between the tracer with the two-bladed impeller (1 and 2) with increasing rotational speed. In the bumping regime a) the powder interacts with each blade. The combination of bump period and blade interaction period define the gravity dominated powder flow of a) bumping, b) apparent roping, c) bumping and d) apparent roping. At high speeds, rotational forces overcomes gravity resulting in roping behaviour (e). The blades are shown stationary with the particle moving flowing over it. ................................................................. 56
4.2 Schematic diagram of the powder bump height $h_a$, powder bump period $t_a$ and the blade interval period $t_b$ measurements used to calculate Bed Resonance number. ........................................................................................................... 57
4.3 Proposed powder flow map for bladed, vertical high shear mixers. ................. 58
4.4 Tracer axial trajectory at varying impeller speeds for cohesive sand using the rectangular impeller. ................................................................. 60
4.5 Power spectral density of particle axial trajectory. ............................................. 62
4.6 Streamline diagrams of particle trajectories of the three powder flow regimes. 63
4.7 Snapshots of powder flow regime using 2 kg of cohesive sand and the rectangular blade in the Hobart mixer ($R = 0.15$ m) at a) 235 rpm, bumping regime, b) 360 rpm, bumping regime (apparent roping behaviour), c) 370 rpm, bumping regime and d) 560 rpm, roping regime. ................................................................. 63
4.8 Sample trajectories of a tracer particle with cohesive sand using rectangular impeller at a) 100 rpm (100 s), b) 200 rpm (50 s), c) 250 rpm (50 s), d) 300 rpm (30 s), e) 400 rpm (20 s), f) 500 rpm (20 s) and g) 600 rpm (40 s). The location of the impeller is shown by - - . The first order bumping behaviour (A) and the second order roping motion (B) are labelled. ................................................................. 64
4.9 Average circulation frequency of cohesive sand using rectangular impeller... 66
4.10 Axial position of tracer particle of cohesive sand and rectangular blade at a) 300 rpm and c) 600 rpm. The red cross represents the intermediate minimums and maximums of the axial trajectory. Vales of bump heights at b) 300 rpm and d) 600 rpm. ................................. 66
4.11 Average overall powder velocities and velocity components (tangential, axial and radial) of cohesive sand using the rectangular impeller at various rotation speeds. ................................................................. 69
4.12 Velocity distributions plots at varying speeds for rectangular blade a) average powder velocity and b) powder tangential velocity. ............ 70
4.13 Comparison of average particle velocity of all bladed experiments. The shaded symbols denote overall particle velocity \( v_p \) values and the hollow symbols are the tangential velocity \( v_\theta \) values. ................................................................. 71
4.14 Comparison of dimensionless particle velocity of all bladed experiments. ... 72
4.15 Azimuthally averaged normalised tangential velocity \( v_\theta/v_{tip} \) (shading) and radial-axial \( v_{rz} \) velocity vectors (arrows) maps for cohesive sand and rectangular impeller at a) 100 rpm, b) 200 rpm, c) 250 rpm, d) 300 rpm, e) 400 rpm, f) 500 rpm, and g) 600 rpm. ......................................................... 73
4.16 Average powder tangential velocity as a function of a) height and b) radial distance of cohesive sand with the rectangular impeller. —— marks the critical particle tangential velocity for this mixer geometry. The impeller boundary is indicated by ................................................................. 74
4.17 Occupancy plots of cohesive sand using rectangular impeller at a) 100 rpm, b) 200 rpm, c) 250 rpm, d) 300 rpm, e) 400 rpm, f) 500 rpm, and g) 600 rpm. 76
4.18 Bed density of the powder bed at various impeller speeds. ............ 77
4.19 Effect of impeller speed on degree of mixing of cohesive sand using a rectangular blade in the a) horizontal and b) vertical azimuthal plane. ................................................................. 78
4.20 Contour maps of shear rates of cohesive sand and rectangular impeller at a) 100 rpm, b) 200 rpm, c) 250 rpm, d) 300 rpm, e) 400 rpm, f) 500 rpm, and g) 600 rpm. Note the different shear rate (s\(^{-1}\)) colour scales. The location of the impeller was shown by - ................................. 80
4.21 Results of powder flow map for two-bladed impellers in a vertical high shear mixer. ................................................................. 81
4.22 Effect of cohesion and blade-rake angle on impeller torque at various impeller rotating speeds for fill load of H/D = 0.29. Bed resonance is observed for all experiments except for dry sand with triangular blade. ................................. 82
4.23 Effective shear thickness for a) gravity dominated bumping regime and b) inertia dominated roping regime. Figure from Mort (2009).

5.1 Tracer axial trajectory for a) dry sand and rectangular impeller b) cohesive sand and rectangular blade c) dry sand and triangular blade and d) cohesive sand and triangular blade at 500 rpm ($v_{tip} = 5.4$ m/s).

5.2 Velocity profiles in the tangential direction at the a) centroid and b) wall. The location of the impeller is shown by - - - -.

5.3 Azimuthally averaged normalised tangential velocity $v_\theta/v_{tip}$ (shading) and radial-axial $v_{rz}$ velocity vectors (arrows) maps at 500 rpm ($v_{tip} = 5.4$ m/s) for a) dry sand and rectangular impeller b) cohesive sand and rectangular blade c) dry sand and triangular blade and d) cohesive sand and triangular blade.

5.4 Normalised frequency distribution for Lagrangian velocity components at 500 rpm for various materials and blade-rake angles. Velocities have been normalised by dividing by impeller tip speed. Frequency bin width are 0.02 m.s$^{-1}$. 

5.5 Occupancy plots at 500 rpm ($v_{tip} = 5.4$ m/s) for a) dry sand and rectangular impeller b) cohesive sand and rectangular blade c) dry sand and triangular blade and d) cohesive sand and triangular blade.

5.6 Contour maps of shear rates (s$^{-1}$) at 500 rpm ($v_{tip} = 5.4$ m/s) for a) dry sand and rectangular impeller b) cohesive sand and rectangular blade c) dry sand and triangular blade and d) cohesive sand and triangular blade. The location of the impeller was shown by - - - -.

5.7 Horizontally averaged slices of shear rates (s$^{-1}$) of a) dry sand and rectangular impeller b) cohesive sand and rectangular blade c) dry sand and triangular blade and d) cohesive sand and triangular blade at 500 rpm. The location of the impeller is shown by - - - -.

5.8 Effects of cohesion and blade-rake angle on degree of mixing in the a) horizontal and b) vertical azimuthal plane.

6.1 General framework to develop compartment models for mixing systems.

6.2 a) Schematic representations of granulation processes occurring in the mixer and the simplified compartment model diagrams of the b) two-CM and c) multi-CM where $C_{imp}$, $C_{cir}$ and $C_{sur}$ are the impeller, circulation and surface compartments respectively and $F$ are particle fluxes between the compartment.
6.3 Cumulative distribution plot of occupancy of cohesive sand and rectangular blade at 600 rpm. Only cells in which the tracer was observed have been considered in the distribution plots and compartments. The plateau in $F(x)$ demarcates a region of low occupancy, providing the threshold value for the surface compartment ($\text{occ} < 0.0009$). 108

6.4 Schematic diagram to define the surface compartment in the azimuthal projection. Using a particle occupancy criterion ($0 < \text{occ}_{\text{cell}} < 0.009$), the shaded green region are cells in which the tracer was observed shows all the cells which in at least one particle (yellow circles) is present and falls between the criterion values. All internal interconnected cells are also included in the surface compartment. 109

6.5 Schematic of sub-compartment model for a) the single well-mixed CSTR model and b) the combination model consisting of a single well-mixed CSTR in parallel with a series of well-mixed CSTR. $\tau_1$ is the average time spent in the single well-mixed tank, $\lambda$ is the proportion of flow entering the single mixed tank, $\tau_3$ is the average time spent in the single well-mixed tank, $\tau_4$ is the average time spent in the series of well-mixed tanks and $N_2$ is the number of tanks in the series for Eqn. 6.1 and 6.3. 110

6.6 Results of the spatial distribution of compartments in the a) two-CM and b) multi-CM. The red squares ■, blue circles ● and green triangles ▲ represent cells within the impeller, circulation and surface compartment, respectively. 115

6.7 Results of the residence time distribution and fitted mixing model for the a) impeller compartment (identical for both two-CM and multi-CM), b) two-CM circulation compartment, c) multi-CM circulation model and d) multi-CM surface compartment. The plots are truncated to highlight interesting behaviours however Table 6.1 shows maximum residence time values. 116

6.8 Lagrangian particle trajectories grouped by residence times of individual passes through the a) impeller and b) circulation compartments over 100 s or 1000 s period for cohesive sand using the rectangular impeller at 100 - 600 rpm. The location of the impeller was shown by - - - - - - - - - - - - - - - - - - - - 120

6.9 Average particle fluxes between interconnecting compartments of the multi-CM. 122
List of Tables

2.1 Summary of vertical high shear mixer studies ................................. 15
2.2 Description of flow regimes in the vertical-axis mixer granulators literature . 17
3.1 Properties of sand particles ..................................................... 29
3.2 Properties of binder .............................................................. 30
3.3 Parameters for flow experiments ................................................. 31
3.4 Compressive and shear loads used in shear test. ............................... 32
3.5 Flowability of powders according to Jenike’s Flow Function ................ 33
3.6 Parameters for flow experiments ................................................. 34
3.7 Summary of powder flow experiments using bladed impellers ............... 40
3.8 Cylindrical grid properties ....................................................... 45
4.1 Velocities of cohesive sand with rectangular and triangular impeller blades at various rotating speeds. ......................................................... 67
4.2 Surface velocities of cohesive sand with rectangular and triangular impeller blades at various rotating speeds. ................................................. 68
4.3 Powder regime map parameters. .................................................. 79
4.4 Shear layer thickness. ............................................................. 84
5.1 Summary of powder flow experiments using bladed impellers ............... 91
5.2 Resulting values of occupied volume vol, bed density \( \rho_{\text{bed}} \) and velocities for various cohesion and blade-rake angle experiments at 500 rpm. ............... 95
6.1 Properties of each compartment from experimental PEPT data for the two-CM and multi-CM: occupied volume vol, occupancy occ, average particle velocity \( v_p \), average dimensionless particle velocity \( v_p^* \), average residence time \( \tau \), one standard deviation of average residence time \( \sigma_\tau \) (68% percentile) and maximum residence time \( \tau_{\text{max}} \). ......................................................... 117
6.2 Model parameters fitted to PEPT experimental data for the two-CM and multi-CM. .......................... 118
<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>AR</td>
<td>Apparent roping</td>
</tr>
<tr>
<td>CM</td>
<td>Compartment model</td>
</tr>
<tr>
<td>CSTR</td>
<td>Continuously stirred tank reactor</td>
</tr>
<tr>
<td>DAE</td>
<td>Differential algebraic equations</td>
</tr>
<tr>
<td>DEM</td>
<td>Discrete element modelling</td>
</tr>
<tr>
<td>LSF</td>
<td>Least squares fitted</td>
</tr>
<tr>
<td>PBM</td>
<td>Population based model</td>
</tr>
<tr>
<td>PEPT</td>
<td>Positron Emission Particle Tracking</td>
</tr>
<tr>
<td>PFR</td>
<td>Plug flow reactor</td>
</tr>
<tr>
<td>PIV</td>
<td>Particle image velocimetry</td>
</tr>
<tr>
<td>rpm</td>
<td>Revolutions per minute</td>
</tr>
<tr>
<td>rps</td>
<td>Revolutions per second</td>
</tr>
<tr>
<td>RTD</td>
<td>Residence time distribution</td>
</tr>
<tr>
<td>StD</td>
<td>Standard deviation</td>
</tr>
<tr>
<td>vHSM</td>
<td>Vertical-axis high shear mixer</td>
</tr>
<tr>
<td>2D</td>
<td>Two-dimensional</td>
</tr>
<tr>
<td>3D</td>
<td>Three-dimensional</td>
</tr>
</tbody>
</table>
All truths are easy to understand once they are discovered; the point is to discover them.

Galileo Galilei
1.1 Background

Wet granulation is an important size-enlargement process that converts fine powders, which are notoriously difficult to handle, to granular material that has improved flow and compression properties that are more easily controlled. This process also reduces the potential risk of explosion due to dust and improves product attributes by limiting segregation of granule constituents (Iveson et al., 2001a). The application of wet granulation is prevalent in many industries such as pharmaceutical, chemical, agricultural, food and metallurgical (Ennis and Litster, 1997). However, despite the industrial significance of this process, large recycle ratio and frequent plant shut downs are still a common problem due to the lack of understanding of the fundamental mechanisms involved in granulation (Liu and Litster, 2002).

1.2 Granulation Mechanisms

Significant progress has been made to elucidate the mechanisms involved in granulation processes. Ennis and Litster (1997) have classified the three crucial mechanisms of granulation as: wetting and nucleation; consolidation and coalescence; and attrition and breakage (see Figure 1.1). An integral part of these mechanisms is understanding the impact of powder
flow and mixing patterns within the granulator. The theoretical prediction of granule size distribution from the fundamental properties of granules and binder is extremely difficult without the knowledge of force and velocity distributions within different types of granulators.

Predictive regime maps have been developed for nucleation and growth mechanisms based on estimated and measured properties of powder velocities and forces within the mixer granulator, however challenges and discrepancies still exist. The nucleation regime map developed by Hapgood et al. (2003) required powder velocities data through the spray zone in order to calculate the dimensionless spray flux ($\Psi_a$) (see Figure 1.2). High speed videoing techniques were used. However, difficulties in observing the bed surface due to dust hindered accurate velocity measurement. Also, the granulator lid was required to be removed in order to observe the surface which may not be possible in industrial equipment due to safety lock mechanisms Litster et al. (2002).

Iveson et al. (2001b) proposed a growth regime map and validated it using a range of formulations and granulation equipment. Drum granulation results correlated well with the proposed map however, high shear mixer results were over predicted (see circled results in Figure 1.3). The authors hypothesised the reason for the discrepancies was an overestimation
1.2 Granulation Mechanisms

Figure 1.2: Nucleation regime map developed by Hapgood et al. (2003) where $\tau_p$ is the dimensionless drop penetration time and $\Psi_a$ is the dimensionless spray flux.

Figure 1.3: Growth regime map developed by Iveson et al. (2001a) where $\rho_g$ is the granule density, $U_c$ is the representative collision velocity in the granulator, $Y_g$ is the granule dynamic yield stress, $w$ is the mass ratio of liquid to solid, $\varepsilon_{min}$ is the minimum porosity the formulation and $\rho_s$ and $\rho_l$ is the solid particles and liquid density, respectively.
of the characteristic impact velocity which were estimated from the chopper tip speed, instead of measured powder velocities. Techniques to accurately measure velocities and forces within the mixer would be useful to validate these regime maps.

Despite a better understanding of granulation mechanisms, there are still challenges which must be overcome before a priori determination of granulation behaviour from fundamental properties are realised. Figure 1.4 outlines the design model approach to granulation used in this thesis. The approach used to achieve the ultimate designer particle breaks down granulation into five main components: 1) physico-chemical interactions, 2) powder flow behaviour, 3) granulation rate mechanisms, 4) population balance models and 5) distributed granulator flow models or compartment models. It can be seen that powder flow behaviour is an integral component to this overall picture.

Prior to the 1990s, powder flow and mixing research predominately centred on empirical studies relevant only to a few specialised industrial devices. A small number of fundamental researchers made some progress analytically, though generally applicable knowledge was still elusive (Bridgwater, 2010).

In the past two decades, advances in measurement and computational power have seen
research in powder and granular flow expand significantly (Bridgwater, 2010, Muzzio et al., 2004). Despite this surge in research, fundamental understanding of powder flow linking particle-scale characteristics to bulk powder behaviour is still lacking due to three main reasons:

1. The fundamental dynamics of powder flow are poorly understood. Currently, constitutive models to predict flow behaviour analogous to the Navier-Stokes equation or viscosity property in the fluid mechanic literature still does not exist (Tardos, 1997).

2. Standard test methods (e.g. shear cell testers) for bulk powder properties are still not applicable across all conditions experienced in mixers where the powder bed can be highly aerated.

3. Powder flow profiles are highly dependent on equipment geometry and design hence the characterisation of flow is generally not applicable across a range of formulations, operating conditions and mixing devices (Litster and Ennis, 2004).

Thus, the development of predictive models of powder flow behaviour based on material properties, operating conditions and mixer design still remains a challenge.

### 1.3 Powder flow characteristics for granulation

Powder flow characteristics important for controlled granulation are considered for each granulation rate mechanism. For wetting and nucleation, characterising surface flows is crucial. Operation within the ideal drop controlled nucleation regime dictates that a continual renewal of fresh powder should be brought to the spray area at the bed surface. The measure of binder density on the powder surface is the dimensionless spray flux (Litster et al., 2001):

\[ \Psi_n = \frac{3 \dot{V}}{2 \dot{A} d_d} \]  

(1.1)

where \( \dot{V} \) is the volumetric spray flowrate, \( d_d \) is an average drop size and \( \dot{A} \) is the powder flux through the spray zone given simply by: \( \dot{A} = v W \) where \( v \) is the powder velocity past the spray and \( W \) is the width of the powder being wet. Ideally, the penetration time of each binder droplet should be shorter than the time it takes for the powder surface to move through the spray zone. In the non-ideal scenario where the powder velocity past the spray is too slow, pooling of liquid binder will occur at the surface. Redistributed of the localised wet mass will then be require to move the wet clumps into the region of high shear, high impact around the impeller or chopper. Spatial and temporal knowledge of mixing patterns
and forces within the mixer is required to predict growth and consolidation, and breakage and attrition rate processes.

1.4 Granulation Equipment

The lack of knowledge of granulation fundamentals has also been attributed to the existence of a multitude of equipment in industry used to deal with this common purpose. It includes fluidised beds, spouted beds, tumbling drums and pans, rotary processor and mixer granulators (Litster and Ennis, 2004). The main differences between these granulators are the way the materials are agitated. The fluidised and spouted beds utilise air to create mixing and flow patterns that produce granules that are more porous, while the tumbling drum, tumbling pan and rotary processor are rotating vessels or plates that transfer momentum via friction and gravity to produce granules that are very spherical. High shear mixers (also called mixer granulators) employ a mechanical agitator to convey the material and tend to make smaller, denser, less spherical granules than other granulators. These mixers are generally more robust and will handle a wider range of feed material including very fine cohesive powders and very viscous liquid binders (Ennis and Litster, 1997).

There are two types of high shear mixers: horizontal or vertical-axis mixers, both equipped with a least one impeller rotating at high velocities (typically between 60 - 800 rpm) (Litster and Ennis, 2004). Vertical mixers are typically operated in a batch while horizontal mixers can be batch, semi-continuous or continuous. A liquid binder is typically sprayed onto a moving bed of powder. Most industrial designs incorporate a chopper mounted on the wall, rotating at very high speeds (500 - 3500 rpm) to break up large lumps of granules from poorly distributed binder systems. Baffles are occasionally added to eliminate solid body rotation. Other design features, auxiliaries and fittings include tapered vessel design, multiple impeller configurations, fluidised air to aid circulation and wall linings.

The powder flow in horizontal mixers have been researched extensively e.g. ploughshare mixers (Jones and Bridgwater, 1998, Forrest et al., 2003, Martin et al., 2007), helical ribbons blade mixers (Broadbent et al., 1993, Laurent and Bridgwater, 2002) and Lödige mixers (Saleh et al., 2005). There have, however, been less studies in vertical high shear mixer (vHSM), which is surprising considering their prevalent use, especially in the pharmaceutical industry. Work on understanding powder flow has mainly focussed on industrial vHSM with rotating impellers at the base e.g. Fielder, Diosna, Collette Gral, Niro Pellmix (Knight et al., 2000, Schaefer et al., 1993), though other studies also include the cyclomix (Ng et al., 2007a,b, Hassanpour et al., 2009) and planetary mixer (Hiseman et al., 2002) (see Figure 1.5). A number of researchers have designed custom mixers in order to simplify complex flow
1.4 Granulation Equipment

Figure 1.5: Examples of industrial high shear mixer granulators: a & b) Aeromatic-Fielder (Litster et al., 2002, Plank et al., 2003), c) Niro Pellmix (Knight et al., 2000, Schaefer et al., 1993), d) Collette Gral (Faure et al., 1999), e) Zanchetta (Nilpawar et al., 2006), f) custom design vHSM (Tu et al., 2009, Knight et al., 2001, Bridson et al., 2007), g) planetary mixer (Hiseman et al., 2002), and h) cyclomix mixer granulator (Ng et al., 2007a)
patterns (see Figure 1.6). However, many studies are still carried out at very low impeller speeds which are not directly relevant to industrial practices (Stewart et al., 2001a). In fact, studies have shown that contrary to their name, many high shear granulators are commonly operated at conditions that actually produce low shear forces (Tardos et al., 2004). This leads to poor mixing and non-uniform particle size distributions.

1.5 Flow visualisation and modelling

The study of powder motion within high shear mixers has been inhibited in the past due to an inability to visualise the flow beyond the surface layer. Recent technological advances have lead to the innovation of Positron Emission Particle Tracking (PEPT), a non-invasive method of investigating particulate systems (Broadbent et al., 1993). Thus, it is now possible to investigate opaque dense powder flows and track particles in dynamic motion. PEPT can provide information on particle motions, velocity fields and occupancies as a function of position and time. Physically-based kinetic parameters from measured PEPT data can be integrated with Population Balance Models (PBM) to provide better estimations of granulation rate processes (Litster and Ennis, 2004). PEPT information can also be used to aggregating spatial similar flow properties into compartments (e.g a high strain rate compartment around the impeller and low strain rate compartment in the remainder of the bed) also know as compartment models (CM). The combination of these two models into a single compartment-based PBM, could provide a natural multi-scale modelling approach of integrating particle flow characterisation given by the CM (distributed macro level) with kinetic physical mechanisms given by PBM (mesoscale) which are still missing in the literature (Michaels, 2003).
1.6 Thesis Objectives

A greater understanding of the effects of material properties, operating conditions and design parameters on granulator performance and scale-up of vHSM, hinges on the ability to accurately and efficiently characterise powder motion. The main objective of this thesis therefore, is to develop an improved understanding of powder flow behaviour in granulator mixers, specifically to:

1. Develop a **powder flow regime map** to determine the dominant flow behaviour in a “simple” mixer granulator, across operating conditions relevant to industrial practices.

2. Investigate the **effects of powder properties, operating conditions and design parameters** on powder flow behaviour using Positron Emission Particle Tracking and torque measurements.

3. Develop a general **framework for compartment models** to utilise powder flow information from PEPT experiments in compartment models suitable for integration into compartment-based population balance models for granulation processes.

This thesis will focus on granulation in custom-design vHSM granulator as a case study.

1.7 Thesis Outline

The outline of the thesis is presented as follows:

Chapter 2 consists of a literature review that summarises the current understanding of granulation, focussing on powder flow behaviour in vertical high shear mixers and the developments in technology that will enable greater understanding of the system.

Chapter 3 describes the experimental methodologies and techniques used in this study, including a detailed analysis of how useful information was extracted from the large amount of PEPT data.

Chapter 4 presents the proposed powder flow regime map for bladed vertical high shear mixers. This will enable a rational approach to the scale-up of mixer granulators.

Chapter 5 investigates the effect of changing formulation properties and design specification on velocity profiles, occupancies and shear rates using the 3D imaging capabilities of PEPT.
Experimental results are compared with existing theories.

Chapter 6 utilises the acquired knowledge from previous chapters to develop a general framework for compartment models of vHSM granulators for future application incorporating population based models.

Finally, in Chapter 7 conclusions are drawn and recommendations are made for further research.

Nomenclature

\[ \dot{A} \] area flux of powder through the spray zone \((m^2.s^{-1})\)

\[ d_d \] average liquid drop size \((m)\)

\[ \dot{V} \] volumetric spray rate \((m^3.s^{-1})\)

\[ U_c \] representative collision velocity in the granulator \((m.s^{-1})\)

\[ w \] mass ratio of liquid to solid \((-)\)

\[ Y_g \] granule dynamic yield stress \((kPa)\)

Greek symbols

\[ \varepsilon_{min} \] minimum porosity the formulation \((-)\)

\[ \rho_g \] granule density \((g/mL)\)

\[ \rho_l \] liquid density \((g/mL)\)

\[ \rho_s \] solid particles density \((g/mL)\)

\[ \Psi_a \] dimensionless spray flux \((-)\)
2.1 Introduction

Flow behaviours of powders and granular materials are essential to the understanding of granulation processes. In this chapter, the current state of knowledge of powder flow in mixer granulators is examined in four sections. The first part of this literature review gives a summary of the research into powder flow theory and existing regimes. In the second part, previous studies of powder flows in mixer granulators are reviewed. Also, the opportunities and challenges of existing measuring techniques and simulation tools are summarised. Thirdly, powder flow models are explored with a focus on all length scales from micro to macro-scale. Finally, a critical summary is given that identified gaps in the knowledge that will be addressed in this thesis.

2.2 Powder Flow Theory and Regimes

Historically, the study of fluid motion has been a subject of extensive research and is well documented. Contrary to this, the subject of powder flow is still poorly understood. One reason for this is that currently, no universal mathematical model exists to predict powder flow behaviour in every situation, analogous to the Navier-Stokes equation for fluid flow, nor
is there a relationship between powder parameters that might be comparable to viscosity (Paul et al., 2004).

The main challenge in developing predictive models of powder flow behaviour in granulation systems is the coexistence of multiple, history dependent granular states. Traditionally, two limiting granular flow regimes have been identified. On one side of the spectrum lies the slow frictional flow regime (also known as quasi-static regime in the solid mechanics literature) and, on the other side, the fully dynamic rapid granular flow regime also defined as the grain-inertia regime by Bagnold (1954). Between the two extreme powder flow regimes, lies a region where both collisional and frictional interactions between particles are significant. In this so called intermediate flow regime, industrially significant flow exists such as in high shear mixing though controversy still surrounds the existence of this regime for powders.

Extensive research in the soil mechanics literature has allowed the development of powder flow models based on the slow frictional flow regime in which frictional forces between particles are dominant. These models have been applied to the design of hoppers with relative success (Jenike, 1964). At very high shear rates and low concentration, the rapid granular flow regime model is based on the kinetic theory of gas due to the physical similarities of the motion of granules in rapid granular flow with the motion of molecules in a gas (Savage and Jeffrey, 1981, Jenkins and Savage, 1983, Lun, 1991, Sela and Goldhirsch, 1998). Rapid granular flow is analogous to ideal gas behaviour in that only brief collisions between particles and random fluctuating velocities determine the character of the flow. It was assumed that the particles interact though monodisperse binary instantaneous collisions like molecules of gas, except that the energy is not conserved during the collisions between granules.

The intermediate regime was first proposed by Hibler (1977) and observations by other researchers (Johnson and Jackson, 1987, Tardos et al., 2003) also indicated its existence. Johnson and Jackson (1987) observed when the blade moved through the horizontal bed of granule material, the inter-granule contact time lies between the two extremes of slow and rapid movement. An intermediate regime was developed by an ad hoc patching together of stresses from the two limiting regimes. A more refined model of the intermediate regime has been developed by Tardos (1997) and co-workers based on the simple geometry of the Couette and hopper device.

A new approach to model the flow in the intermediate regime was proposed by Savage (1998). Granular flows are considered at very fine scales where particle velocities are divided into fluctuating and mean transport components. The constitutive equations relate the standard deviation to fundamental quantity of granular temperature in the powder. This idea is used quite extensively in rapid granular flow models (Savage, 1998). A relationship between the mean stress and the mean strain rate reveals a viscous-like behaviour. For
low shear rates, the effective viscosity decreases with increasing temperature, whereas for rapid granular flows, the viscosity increases with increasing granular temperature as in a gas. Tardos et al. (2003) have extended this model to obtain an expression for average stress that reduces to the slow frictional regime limit when fluctuations go to zero while, in the limit of large fluctuations, a “viscous character” is displayed by the bulk powder. This model proposes a smooth transition from one regime to another (see Figure 2.1). The dimensionless shear rate given by Tardos et al. (2003) is:

\[ \gamma^* = \gamma \sqrt{\frac{d_p}{g}} \]  

(2.1)

where \( \gamma \) is the shear rate, \( d_p \) is the bed depth and \( g \) is the gravity constant.

GRD Midi (2004) further adapted the dimensionless shear rate relating inertia to normal stress in the particle bed given by:

\[ I = \frac{\dot{\gamma} \cdot d_p}{\sqrt{P/\rho}} \]  

(2.2)

where \( \dot{\gamma} \) is the shear rate, \( d_p \) is the particle diameter, \( P \) is a characteristic normal stress.

---

**Figure 2.1**: Granulator and reactor scale approach (Tardos et al., 2003)
and $\rho$ is the bulk density. In vertical free surface flow, the shear rate can be approximated with $\dot{\gamma} = \frac{v_{\text{tip}}}{h_{\text{bed}}}$ where $h_{\text{bed}}$ is the static bed height and $v_{\text{tip}}$ is the impeller tip speed and the characteristic normal stress with $P = \rho \cdot g \cdot h_{\text{bed}}$.

$$I = \frac{v_{\text{tip}} \cdot d_p}{\sqrt{g \cdot h_{\text{bed}}^3}}$$  \hspace{1cm} (2.3)

The transition between the quasi-static regime (where the effective friction coefficient is constant) and the dense inertial regime occurs when $I$ is approximately 0.01 (GRD Midi, 2004).

These approaches to model the intermediate flow regime have only investigated very simple limiting cases to predict powder behaviour. Particle interactions are currently assumed to be binary (Savage, 1998). To enable the application of these models to more general flow problems, improvements should include multiple collisions which often occur in high shear mixers.

While there has been considerable progress toward a theory-based approach to understanding granular flow and mixing, no predictive models exist that are applicable to all granular systems. Until such time, the advancement of powder technology will still continue to be a combination approach of fundamental theory, experimental investigation and simulation. The experimental approaches to powder flow in mechanically agitated systems are reviewed next.

## 2.3 Powder flow studies

Many studies involving high shear granulation are responses to the practical needs of the pharmaceutical industry and hence are concerned with the optimisation of granulation equipment and operating parameters (Hapgood, 2000). These areas of research include operating parameters (e.g. speed, residence time, temperature), method of liquid addition, binder and powder properties, binder and powder ratio, control, equipment design and scale up. In the few cases where powder flow behaviour is mentioned, it is rarely distinguished from other mechanisms that are occurring simultaneously in the system. Table 2.1 provides a useful reference of vHSM studies.

Knight et al. (2000) investigated the effect of impeller speed on granule growth in a Niro Pellmix with a calcium carbonate and polyethylene glycol mix. The molten binder was poured directly onto the powder surface whilst the impeller was stationary. A clear bimodal distribution is observed in which local regions of highly saturated powder grew preferentially. High shear rates were required to mechanical distribute these granules though even after
## 2.3 Powder flow studies

### Table 2.1: Summary of vertical high shear mixer studies

<table>
<thead>
<tr>
<th>Author/s</th>
<th>Materials</th>
<th>Equipment</th>
<th>Powder flow/mixing</th>
<th>Powder properties</th>
<th>Binder properties</th>
<th>Impeller speed</th>
<th>Fill level</th>
<th>Liquid-powder ratio</th>
<th>Temperature</th>
<th>Time</th>
<th>Control</th>
<th>Design</th>
<th>Scale up</th>
</tr>
</thead>
<tbody>
<tr>
<td>Litster et al. (2002)</td>
<td>Lactose; water, HPC solution</td>
<td>A</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
<tr>
<td>Plank et al. (2003)</td>
<td>Lactose, MCC, Starch, Sucrose; Water</td>
<td>A</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
<tr>
<td>Tardos et al. (2004)</td>
<td>Lactose, MCC, Starch, Sucrose, Mannitol; Water, HPC, Ethanol</td>
<td>A F</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
<tr>
<td>Knight (1993)</td>
<td>Sodium phosphate</td>
<td>F</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
<tr>
<td>Westell (1997)</td>
<td>Sand, zeolite, polypropylene; glycerol, PEG</td>
<td>1</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
<tr>
<td>Knight et al. (2001)</td>
<td>Sand</td>
<td>1</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
<tr>
<td>Bridson et al. (2007)</td>
<td>Lactose</td>
<td>1</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
<tr>
<td>Tu et al. (2009)</td>
<td>MCC; PEG</td>
<td>1</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
<tr>
<td>Stewart et al. (2001a)</td>
<td>Glass beads</td>
<td>2</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
<tr>
<td>Knight et al. (2000)</td>
<td>Calcium carbonate; Polyethylene glycol</td>
<td>N</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
<tr>
<td>Ramaker et al. (1998)</td>
<td>MCC, Lactose; Water</td>
<td>O C</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
<tr>
<td>Faure et al. (1999)</td>
<td>Lactose, Starch; Water</td>
<td>C</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
<tr>
<td>Horsthuys et al. (1993)</td>
<td>Lactose; PVP, Water</td>
<td>C</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
<tr>
<td>Ghorab and Adeyeye (2007)</td>
<td>βCD, Ibuprofen, Magnesium steerate; Water, Isopropanol</td>
<td>C</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
<tr>
<td>van den Dries et al. (2003)</td>
<td>Lactose; HPC</td>
<td>C</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
<tr>
<td>Nilpawar et al. (2006)</td>
<td>Calcium carbonate; Glycerol, PEG</td>
<td>Z</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
<tr>
<td>Darelius et al. (2007b)</td>
<td>MCC</td>
<td>M</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
<tr>
<td>Lekhal et al. (2006)</td>
<td>Sand; Water</td>
<td>3</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
<tr>
<td>Remy et al. (2010b)</td>
<td>Glass beads</td>
<td>3</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
<tr>
<td>Mort (2009)</td>
<td></td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
</tbody>
</table>

✓ Tick indicates parameters studied. Abbreviations: A - Aeromatic Fielder, C - Collette Gral, F - Fukae, M - MiPro, O - Moulinex coffee grinder, N - Niro Pellmix, Z - Zanchetta, 1 - Custom mixer (Birmingham), 2 - Custom mixer (Cambridge), 3 - Custom mixer (Rutgers), MCC - Microcrystalline cellulose, βCD - Beta-cyclodextrin, HPC - Hydroxypropylcellulose, PEG - Polyethylene glycol, PVP - polyvinylpyrrolidone.
some time, the bimodal distribution still persisted. Higher shear rates were able to more efficiently distribute these regions to reduce the large clumps. The authors hypothesise that the bimodal distribution was caused in some part to breakage however, it is probably a remnant of the poor binder distribution from the start.

One approach to simplify the complex mechanisms occurring in an agitated mixer is to design simple testing equipment instead of using real process equipment, to measure specific meso-scale properties e.g. powder viscosity (Michaels, 2003). Various geometries have been used as test systems for powder properties such as the shear cell tester, ring shear tester and Couette devices (Talu et al., 2001, Tardos et al., 2003).

Another approach is to use simplified agitated systems. Several researchers studied the flow of powder over a single blade moving linearly through a trough using photography of tracer particles through a glass wall (Bagster and Bridgwater, 1967, Bagster, 1969, Bagster and Bridgwater, 1970). Two motions were identified in the convective action of the blade: a net displacement of the main bed in the direction of the moving blade, and a bypassing of the bed via a recirculation region in front of the moving blade. Dynamic similarities were identified using four key dimensionless groups: the blade height to particle diameter, the blade immersion height to particle diameter, the tracer particle immersed height to particle diameter and the impeller Froude number.

While these investigations provided useful information, the application is limited as the studies were concerned with low velocity, frictional behaviour and carried out at operating conditions much lower than those commonly used in industry. Also, the movement of the powder bed in a mechanical mixer has the compounding effect of bed dilation created by the passage of a series of impeller blades through a bed of powder which is not captured in a single bladed set-up. Bagster and Bridgwater (1970) hypothesised the effect of blade impact on the powder in the inertia regime as “materials may well fly many blade heights downstream before landing”. Here, these insights are built upon to develop a classification system for the motion of powder in vertical-axis high shear mixers.

### 2.3.1 Powder flow regime map

One of major challenges in comparing vHSM studies is the lack of universally accepted flow regimes. Flow regimes and their corresponding transitions can be graphically presented using regime maps. This simple approach can determine the flow patterns without the need to directly observe the flow. Regime maps have been developed to analyse the flow in a range of process equipment including Rushton turbine, horizontal flows, vertical flows and fluidised beds.
Table 2.2: Description of flow regimes in the vertical-axis mixer granulators literature

<table>
<thead>
<tr>
<th>Author/s</th>
<th>Flow descriptions and regimes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Schaefer et al. (1993)</td>
<td>Torus (helical shape)</td>
</tr>
<tr>
<td>Ramaker et al. (1998)</td>
<td>Toroidal ring</td>
</tr>
<tr>
<td>Stewart et al. (2001a)</td>
<td>Rising and falling regions around blade with stagnant regions between blades</td>
</tr>
<tr>
<td>Knight et al. (2001)</td>
<td>Fully lifted powder bed; Partially lifted powder bed; Toroidal</td>
</tr>
<tr>
<td>Tardos et al. (2004)</td>
<td>Stationary; plug flow solid-body rotation with impeller</td>
</tr>
<tr>
<td>Litster et al. (2002)</td>
<td>Bumping; Transition; Roping</td>
</tr>
<tr>
<td>Plank et al. (2003)</td>
<td>Surface stagnation; Toroidal</td>
</tr>
<tr>
<td>Mor (2007)</td>
<td>Centripetal</td>
</tr>
<tr>
<td>Remy et al. (2010a)</td>
<td>Bumping; Roping</td>
</tr>
</tbody>
</table>

The first step towards a map is the characterisation of the basic flow structure independently of equipment set-up and scale. Table 2.2 lists the flow description from several researchers who have described the powder flow pattern in vHSM resulting from a combination of centrifugal forces, centripetal forces, impact forces and gravity. A large variety of flow regimes and names exist to describe the basic flow structure for two reasons: firstly, qualitative observations of flow regime are subjective and secondly, different names are given to the same regimes.

Litster et al. (2002) used dry lactose to measure powder surface velocities as a function of impeller speed using a high speed camera. They defined two basic flow patterns for bladed vertical-axis mixer granulators: **bumping** and **roping** (see Figure 2.2). A transitional stage could be included to develop a comprehensive map with the following definitions:

1. **Bumping regime** (also referred as pulsing in a horizontal mixer (Laurent, 2006) or shunting in a planetary mixer (Hiseman et al., 2002)): At low impeller speeds, the powder bed surface appears to remain horizontal as the bed is “bumped” in sequence to the passage of the blade beneath it.

2. **Transition**: Increasing the impeller speed somewhat, an unstable behaviour is reported where powder is pushed up and out, creating an unstable powder wall which collapses to the centre of the vessel producing large amounts of dust.

3. **Roping regime** (also referred as toroidal): At high impeller speed, a stable toroidal flow pattern forms with good vertical turn over of the powder bed. The powder is forced from the bottom of the mixer up the wall and then tumbled down the angled bed surface towards the centre of the bowl.
Quantitative definitions of these flow regimes have been based on the measured values of powder velocity as a function of impeller speed. As an experimental precursor to a more generalised regime map, Litster et al. (2002) measured the powder surface velocity of lactose powder in a Fielder mixer with a bowl diameter of 0.4 m. The powder surface velocities were found to increase linearly with impeller speed in the bumping flow regime and were less sensitive to impeller speed in the roping regime (see Figure 2.3). From extrapolation, a critical transition point is observed at an impeller speed of 250 rpm. In general, the powder surface velocities were an order of magnitude lower than the impeller tip speed.

The transition between bumping and roping was approximated to be the balance between the rotational inertia with gravity. Litster et al. (2002) suggested the impeller Froude
number:

$$Fr_i = \frac{DN^2}{g} \quad (2.4)$$

where $D$ is the impeller diameter, $N$ is the impeller rotational speed (rpm) and $g$ is the gravitational acceleration. However, scale-up using $Fr_i$ is only useful for geometrically and kinematic similar mixers, which in most cases, are different even across the same model type (Michaels, 2003).

A more general approach which could be applied to different combinations of experimental set-up and materials would be useful, one that could be used to predict the operating conditions in any equipment. Some authors have suggested a more appropriate term would be to use a modified Froude number based on particle properties (Knight et al., 2001, Forrest, 2004), namely, the powder Froude number:

$$Fr_p = \frac{v^2}{Rg} \quad (2.5)$$

where $v$ is characteristic powder velocity and $R$ is the impeller radius. Here the critical parameter is the characteristic powder velocity which is a complex function of mixer geometry, impeller design, dimensionless fill level or bed height, and cohesion. In addition to this complexity, a broad range of velocities exist in a mixer. Even at very low impeller speeds, Stewart et al. (2001a) measured an order of magnitude difference in the range of particle tangential velocities in a stirred mixer.

Another method used to identify critical changes on a macroscale, is to measure energy input into the powder bed. Wellm (1997) designed a “floating” mixer bowl with an attached load arm to enable torque measurements to be obtained via a load cell. Torque was generally observed to have a quadratic relationship with impeller speed at low values, up until a critical transition at which, the relationship changed to a linear one (see Figure 2.4). These different regions are analogous to the regimes described by Litster et al. (2002). This demonstrates that torque could be used to detect powder flow transitions.

Tardos et al. (2003) introduced the dimensionless shear rate to determine the regime of granular flow (refer to Eqn. 2.1). A mixer granulator flow regime map has been proposed by Mort (2009). Building on previous work by Tardos et al. (2003), Mort identified three flow regimes: 1) frictional, 2) intermediate fluid-like and 3) rapid collisional (see Figure 2.5). This map allows the dominating flow regime to be determined as a function of $K_1$, the ratio of mean particle velocity $\bar{U}_p$ to impeller tip speed $U_i$ and $K_2$, the ratio of the shear transmission height $\delta$ to mixer radius $R$. The limitation of this map is that it is only applicable to systems operating in the roping regime ($Fr_p >> 1$) where centripetal acceleration exceeds gravity.
**Figure 2.4**: Effect of impeller speed on torque (Wellm, 1997)

![Torque Speed Relationship](image)

**Figure 2.5**: Mixer granulator regime map for flows operating in the roping regime where $K_1$ is the ratio of mean particle velocity $U_p$ to impeller tip speed $U_i$ and $K_2$ is the ratio of the shear transmission height $\delta$ to mixer radius $R$. Flow regimes are defined in terms of a dimensionless shear rate: i) frictional, $\dot{\gamma}^* < 0.2$; ii) intermediate, fluid-like; and iii) rapid collisional, $\dot{\gamma}^* > 3$ (Mort, 2009).
To date, no powder flow regime map has been published for vHSM that quantitatively predicts flow regimes and the corresponding transitions across the operating conditions commonly practised in industry.

2.3.2 Effects of different parameters on powder flow behaviour

Impeller and/or chopper speeds

Studies involving variations in impeller speeds to influence powder flow behaviour are by far the most abundant in the powder flow literature. This is not surprising as the rotating devices are the only mechanism in which forces can be transferred to the powder bed. The effects of impeller speed on powder flow profiles from bumping through to roping regimes, have been discussed previously in the previous section.

Binder content

Increasing the binder to dry powder ratio increases the cohesiveness of the powder bed. Figure 2.6 shows a study by Plank et al. (2003) into the effect of binder addition on the surface velocity. It is clear that with increased binder content, the powder velocity at the bed surface increases, albeit non-linearly. Changes in the powder bed profile were reported as the binder was added, forming a “wet mass” that permitted the shear forces from the rotating impellers to be transmitted more effectively to the whole powder bed. Ng et al. (2007b) also observed increases in the average tangential velocity with the addition of a liquid binder at low speeds. However, contrary to these results, at higher speeds, a decrease in the tangential velocity was observed. Further investigation revealed that this result was caused by the complex impeller configuration of the system being studied, in which the impeller did not reach all the way to the mixer wall (see Figure 1.5f). The wet cohesive mixture tended to adhere to the wall creating dead zones.

Another comparative study of wet and dry granules was carried out in a small coffee grinder by Ramaker et al. (1998). A stable flow pattern was observed for wet granules even without the cover on. However, no stable flow patterns were observed for dry granules. The authors hypothesised that the formulation properties were a key factor but provided no further evidence. The conclusion from these studies is that the addition of binder to the granulation system is expected to be an important influence of powder flow behaviour, particularly its role in developing a cohesive “wet mass”.

Litster et al. (2002) studied wet and dry lactose in the 25L Fielder granulator and found that for wet lactose, the transition from bumping to roping occurred at 150-200 rpm, while
Figure 2.6: Surface velocity with the addition of binder (Plank et al., 2003)

for dry lactose, it occurred at higher speeds (250-300 rpm). These effects were attributed to the cohesive properties of wet lactose.

Fill level

The effect of fill level on powder flow behaviour in vHSM has received relatively little attention. Stewart et al. (2001a) varied fill level from 1.4 to 7 kg in vertical bladed mixer and found that the tangential particle velocity distribution was broader at low fill levels where most particles were directly interacting with the moving impellers. At higher fill levels, most particles were located in the bulk circulation regions above the impeller zone resulting in limited motion and the narrowing of the distribution. Remy et al. (2010b) simulated various fill level at low impeller speeds and found that increasing fill levels reduced surface velocities however, above a critical fill level, the velocities profiles are independent of fill level.

Impeller and mixer design

In agitated mixers, powder flow behaviour is highly sensitive to the geometry of the mixer and impeller blades. The motion of the bed is generated through the transferral of momentum from the impeller to the powder through a combination of impact, centrifugal, frictional, shear, gravity and inertial forces (Litster and Ennis, 2004). Hence, it is difficult to obtain a
generalised flow pattern for all mixers.

Simple mixers with one set of impeller blades located at the bottom of the vessel (e.g. Aeromatic-Fielder, Colette Gral, Niro Pellmix, Zanchetta) create powder flow that move from the bottom of the mixer, out towards the wall, up the wall and then cascades down towards the centre of the mixer (Litster et al., 2002, Plank et al., 2003, Knight et al., 2000, Schaefer et al., 1993, Faure et al., 1999, Nilpawar et al., 2006).

More complex mixers include multi-impeller mixers such as the cyclomix in which the flow is observed to move either up or down the wall depending on the impeller locations (Ng et al., 2007a). Impeller design also has significant impact on the flow with the planetary mixer “K-beater” producing highly complex flow patterns (Hiseman et al., 2002).

A generalised approach to characterise powder flow behaviour using ideal systems could solve the problem of equipment dependent parameters (Michaels, 2003). Chemical engineering has long used two ideal reactor types, the continuous stirred tank reactor (CSTR) and plug flow reactor (PFR), to mathematically model mixing behaviour as a function of residence time and rates of reactions (Levenspiel, 1999). A similar approach would progress the study of powder flow towards quantitative characterisation of key transport properties (e.g. powder “viscosity”) and transport kinetics (e.g. growth and breakage kernels) (Michaels, 2003).

### 2.4 Techniques of studying powder flow

Techniques used to explore the dynamics of powder flow and mixing have traditionally been hindered by the inability to observe particles behind the second or third layer. Such techniques include tracers embedded in the powder (Bagster and Bridgwater, 1967, Bridgwater et al., 1969b, Bagster, 1969, Bagster and Bridgwater, 1970), photography through transparent walls of the mixer (Bagster, 1969, Malhotra and Mujumdar, 1990, McCarthy et al., 1996), laser doppler anemometry to measure particle velocity at the wall region (Darelius et al., 2007b) and digital video imaging and recording to measure surface velocities. However, a full understanding of the complex internal mixing circulation of the powder bed cannot be obtained just by the two dimensional characterisation of flow fields at exposed surfaces, either at the free surface or at walls.

A three dimensional visualisation method used by Novosad (1968) involved the dissection of a bed of different colour layers that had been frozen using paraffin. Though a crude and time consuming method, it found that the flow patterns observed at the wall were only qualitatively representative of the bulk powder. In the past twenty years, advances in visualisation technology and computer processing power and has revolutionised the study of
powder flows in industrial systems (Bridgwater, 2010).

The innovation of Positron Emission Particle Tracking (PEPT) has allowed non-invasive, three-dimensional studies of flow patterns of laboratory-scale equipment (Parker et al., 1993). Applications of this technique include a rapid expansion of studies of powder flow (Field et al., 1991, Broadbent et al., 1993, Laurent and Bridgwater, 2002, Forrest et al., 2003), fluid flow and liquid-solid systems. PEPT was developed at the University of Birmingham and utilises radioactive tracer particles that are tracked in space and time within process equipment. This tracer emits a high energy positron that annihilates with a nearby electron to produce two collinear gamma rays. These gamma rays are captured by a camera via two detector plates on opposite sides of the apparatus, giving a line on which the annihilation occurred. The intersection of several of these lines marks the location of the tracer particle to 5 mm for a tracer moving at 1 m/s (Stewart et al., 2001a). The gamma rays can travel through opaque material and hence the three-dimensional motion of a particle can be tracked within an operating high shear mixer. Further advancements of the technique include studies of up to three tracers (Yang et al., 2006, 2007, 2008) and a portable, modular version of PEPT (Leadbeater and Parker, 2010). A detailed technical description of PEPT is provided in Section 3.5.

Research has been conducted using the PEPT to delineate the powder flows in various process equipments, including ploughshare mixers (Jones and Bridgwater, 1998, Forrest et al., 2003), planetary mixers (Hiseman et al., 2002), rotary kilns (Parker et al., 1997, Lim et al., 2003, Kuo et al., 2003) and single-bladed horizontal mixer (Laurent and Bridgwater, 2000).

In a vertical flat bladed mixer, Stewart et al. (2001a) investigated the flow patterns of dry powder flow using PEPT. This in-depth study described the motion of powder material whilst varying the fill level and impeller speed. Significant radial motion was observed. Also, three-dimensional patterns of recirculation in front of the blade were described by Bagster and Bridgwater (1967).

Limitations to this technique involve the deflections of gamma rays by collisions with material allowing the detection of approximately 0.5% of gamma ray pairs. This consequently leads to prolonged experiments, sometimes over an hour or more, to collect many points to obtain accurate estimates of the tracer position (Stewart et al., 2001a). As many granulation systems occur over a period of minutes, this time constraint needs to be taken into consideration (Litster and Ennis, 2004). Also, prolonged mixing experiments results in temperature increases, undesirable for temperature sensitive formulations. The inclusion of a cooling jacket could mitigate any adverse effect of temperature for the duration of the mixing experiment.

The accuracy of PEPT also decreases as the speed of the particles increases due to the
speed at which the camera can collect data (Hawkesworth et al., 1991). The use of tracers that are representative of the bulk powder is also a variable that needs to be addressed as segregation can occur and impact on the results obtained in the PEPT. It is important to note that using the high shear mixer with PEPT, velocity and position of a single particle can be obtained but collision velocities, collision frequencies and all the forces acting on that particle, which is useful for breakage and growth mechanism, are not directly measurable (Forrest, 2004). Even with the use of multiple particles, the resolution and frequency of collisions are too low to be practical.

As for the data analysis, Track is the standard program provided to users for initial visualisation. However the tool is limited in its ability for data manipulation. Despite the large number of PEPT studies in the literature, no common set of analysis techniques or coding is readily available.

### 2.5 Advances in computational powder flow models

Advances in computational modelling have allowed considerable insight into powder flow mechanics at the microscale (particle-level). Discrete element modelling (DEM) is a technique that has also been developed to simulate the motion of powder flows based on real interparticle force laws governing particle-particle interactions. Stewart et al. (2001b) and Chandratilleke et al. (2010) have observed similar flow patterns in experimental data obtained from PEPT in a vHSM using discrete element modelling (DEM). Simple force balance models of particles in high shear mixers were developed and have shown good general correlation with PEPT results at very low speeds (< 160 rpm). However, no one set of assumptions best predicts the detailed motion in all sections of the bed. Other workers who modelled vHSM with DEM are Bertrand et al. (2005). These techniques have increased our ability to characterise the motion of powder in a mixer granulator. However, more work is required to validate these models with experimental data for a comprehensive range of conditions and designs. Perhaps the most useful application of PEPT is its use as an experimental validation tool for particle dynamic modelling (Stewart et al., 2001a,b, Hassanpour et al., 2009, 2011, Marigo et al., 2010). Limitations of this modelling approach relates to the lack of understanding of the inter-particle interactions (Stewart et al., 2001a). Also, the computation time required to solve even the most simple real life problem (i.e. non-spherical particles) is a major drawback for its current application.

It has been suggested that an intermediate solution to the bottleneck in computer processing power is the use of compartment models (CM). CMs are a mathematical model used to describe the transport processes between interconnected volumes of materials or energies
of a system, in this case, a mixing vessel. CM are based on the assumption that perfect mixing occurs in each compartment, allowing volumes of “homogeneous” powder to be grouped together at a mesoscale instead of tracking individual particles which could number in the millions in a typical granulator. This mesoscale modelling approach provides a linkage between the three length scale of micro, meso and macro (see Fig 1.4), capturing key flow behaviours in mixer granulators more efficiently (Michaels, 2003). The main advantage of CM is that it is computationally less demanding than other models (e.g. DEM).

CM have been used extensively to model flows in crystallisers and chemical reactors however, very few studies exist of granulators. One study in which CM was developed for powder material was by Freireich (2010) in a vertical-axis mixer. The residence time distributions were obtain from DEM models to model the surface shearing region as well-mixed tank in parallel with a series of well-mixed tanks. The remainder of the bed was modelled using the same set-up with the addition of a shortcut (see Figure 2.7). The results showed similar behaviour to DEM but with one tenth of the processing time. As far as the author is aware, no CM utilising experimentally fitted parameters from PEPT data has been studied to date.

Building upon the CM, a set of population balance (PB) equations can be written for each compartment. For a well-mixed compartment, the general PB equation is given by (Freireich, 2010):

$$\frac{\partial n}{\partial t} + \frac{\partial}{\partial t_s}(Gn) = \sum_i \frac{Q_{in}}{V_i} n_i - \frac{Q_{out}}{V} n$$  

(2.6)
where \( Q \) is the volume flow rate, \( V \) is the volume of the compartment, \( G \) is the local growth rate and \( t_s \) is the mean residence time. (Freireich, 2010) developed a fully integrated compartment-based population based model (PBM) for the dual-axis paddle mixer however, did not extend the CM model to include a PBM for the vertical-axis mixer.

## 2.6 Critical summary

Granulation has slowly progressed in the past sixty years with the identification and understanding of the fundamental mechanisms involved in granulation. However, substantial gaps in the literature still remain. An understanding of the particle collisional forces and velocities distribution in mixer granulator systems is still lacking at present. This situation has forced researchers to rely on gross macroscopic estimates (Ennis and Litster, 1997). In addition, mixing patterns and constitutive relationships for all powder flow regimes are still poorly understood.

Thus, the three major gaps in the literature covered by this study are:

1. Detailed flow characterisation of cohesive powder in a “simple” vertical-axis bladed mixer granulator at industrially-relevant operating conditions has not been studied. The development of visualisation technology such as PEPT is a powerful tool to enable the characterisation of internal flow of particulate systems without disturbing the bed.

2. The classification of powder flow patterns at the macroscale, into a powder flow regime map for vertical-axis mixer granulators.

3. The development of a general framework to develop compartment models that accounts for the spatial and temporal distribution of powder properties based on experimental PEPT measurements of internal flow characteristics.

It is proposed that a detail study of vHSM granulator be undertaken to characterise powder flow behaviour. PEPT will be used to provide qualitative and quantitative information of the internal flow patterns. These experimental results will be used to develop a powder flow regime map defining key transitional boundaries. A general framework to integrate spatially distributed powder properties from PEPT into a compartment model will be investigated.
3.1 Introduction

This chapter describes the materials, methodology and data analysis techniques used to study and characterise the powder material in the vHSM granulator. These techniques are used throughout the thesis and are discussed here to avoid duplication.

3.2 Materials

3.2.1 Selection of model powders

The complex behaviour of powder flow in the vHSM were studied using model powders that corresponded to a set of criteria. These required the model powders to be inexpensive, widely available in narrow, reproducible size distributions and to vary in flow properties across the range of granulation process of dry blending and wet massing. Based on these criteria, free flowing dry sand was selected to represent dry blending flow. Sand was supplied by the David Ball Group U.K., in the form of natural, uncrushed silica sand, washed, dried and graded free from silt, clay or organic matter.

The other model powder was cohesive sand, a mixture made up of dry sand and silicone
oil. Researchers have broadly classified cohesion in particulate solids into two types: dry and wet cohesion (Alexander et al., 2006). For dry cohesive systems, the particles are typically less than 100 µm and solid flow behaviour is controlled by Van der Waals and electrostatic forces. For wet or moisture-induced cohesion, the addition of small amounts of liquid to powder induces cohesion through capillary forces. For this study, the latter cohesive system was investigated as it best simulated the dynamic flow behaviour of wet masses observed in granulation processes where liquid binders are added to powder.

The cohesive sand mixture was prepared by placing dry sand and silicone oil into a plastic bag in a ratio of 2 kg of dry sand to 50 mL of silicone oil and then hand kneaded for 5 minutes to obtain a uniform and reproducible starting mass. The liquid to solid ratio after binder addition was 2.5 % w/w. Silicone oil was selected over water as the binder as it thermally stable, demonstrating little change in physical properties over the temperature range expected in the granulator mixer. Silicone oil DC 200 at a nominal viscosity of 0.1 Pa.s was supplied by Sigma Aldrich Company, Australia.

### 3.2.2 Powder properties

The particle size distributions measured by laser diffraction using a Malvern Mastersizer 2000. Data was obtained from Forrest (1998) for true particle density measured using helium pycnometry and for particle porosity using nitrogen absorption. The particle properties of sand are shown in Table 3.1.

<table>
<thead>
<tr>
<th>Particle property</th>
<th>Sand</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nominal size $a$</td>
<td>150-300 µm</td>
</tr>
<tr>
<td>Surface mean $d_{32} b$</td>
<td>217 µm</td>
</tr>
<tr>
<td>Volume mean $d_{43} b$</td>
<td>234 µm</td>
</tr>
<tr>
<td>$d_{10} b$</td>
<td>157 µm</td>
</tr>
<tr>
<td>$d_{50} b$</td>
<td>226 µm</td>
</tr>
<tr>
<td>$d_{90} b$</td>
<td>323 µm</td>
</tr>
<tr>
<td>Particle density $\rho_p c$</td>
<td>2500 kg/m$^3$</td>
</tr>
<tr>
<td>Porosity $c$</td>
<td>$\sim 0%$</td>
</tr>
</tbody>
</table>

$^a$ Supplier specification (David Ball Group, UK)  
$^b$ Measured using Malvern Mastersizer 2000 by research assistant David Page.  
$^c$ Data from Forrest (1998)
3.2.3 Binder properties

Viscosity of the binder was measured with an Advance Rheometer AR1000 using a 40 mm steel plate with a gap of 5mm between the plate and the detector. Shear rate was controlled at 10/sec and the temperature was incrementally increased at 1°C/min over a range of 15-50°C. Figure 3.1 shows the results of varying temperature on the viscosity of silicone oil. The viscosity was observed to decrease with increasing temperatures and the measured viscosity of 0.074 Pa.s at 25°C was slightly less than the manufacturers nominal value of 0.1 Pa.s. A summary of the binder properties are listed in Table 3.2.

![Figure 3.1: Viscosity of 0.1 Pa.s silicone oil over varying temperatures.](image)

<table>
<thead>
<tr>
<th>Table 3.2: Properties of binder</th>
</tr>
</thead>
<tbody>
<tr>
<td>Binder property</td>
</tr>
<tr>
<td>-----------------</td>
</tr>
<tr>
<td>Density (20°C)</td>
</tr>
<tr>
<td>Viscosity (25°C)</td>
</tr>
</tbody>
</table>

\(^a\) Supplier specification (Sigma-Aldrich, Australia)

\(^b\) Measured using Advance Rheometer AR1000 at the University of Birmingham

3.2.4 Bulk powder properties

The packing characteristics of a powder are important. They are dependent on the shape, average size, size distribution, surface characteristics of the particles, as well as the state of agglomeration of the powder. Two measurements were used to characterise bulk properties: poured and tapped densities.

Poured density was determined in a 250 ml graduated cylinder, in which a known mass of powder material was transferred into the cylinder held at a 45° angle. An average level in the cylinder to the nearest millilitre was recorded. The cylinder was then placed onto the
3.2 Materials

Copley JV1000 tap master (see Figure 3.2) to determine tapped density. A constant tapping amplitude was applied to the cylinder for a total of 3000 taps and the level was recorded. This technique was repeated three times for each powder material. The results are shown in Table 3.3. The bulk density of dry sand is greater than the cohesive sand due to its ability to pack more closely together.

![Figure 3.2: Determination of powder tapped density a) schematic drawing b) Copley JV1000 setup.](image)

<table>
<thead>
<tr>
<th>Table 3.3: Parameters for flow experiments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bulk Properties</td>
</tr>
<tr>
<td>Poured density $\rho_p$ ($kg/m^3$)</td>
</tr>
<tr>
<td>Tap density $\rho_t$ ($kg/m^3$)</td>
</tr>
</tbody>
</table>

*a* Measured using Copley JV1000 tap master

The bulk solids flow properties were determined using a shear tester, the Schulz Ring Shear Tester RST-XS. The flow properties obtained from the analysis of the shear tests included 1) the effective internal angle of friction, 2) the wall angle of friction, 3) the unconfined yield stress and 4) the flowability of the powder under investigation.

A known mass of powder was carefully filled into an annular shear cell of know volume and covered with a lid. The shear tester applied a series of both compressive and shear loads to simulate flow condition in a mixer (see Table 3.4).

For each compressive load, once the material in the shear cell is consolidated, the material strength is measured by shearing it until the sampled reached the point at which steady-state flow is achieved (i.e. where the shear stress reached a constant value). These measured values are then plotted on a $\tau,\sigma$-diagram, forming the yield locus as seen on the example Figure 3.3. After each different load condition, the sample is brought back to the initial pre-sheared state.
### Table 3.4: Compressive and shear loads used in shear test.

<table>
<thead>
<tr>
<th>Consolidating stress $\sigma_{\text{pre}}$</th>
<th>16 kPa</th>
<th>8 kPa</th>
</tr>
</thead>
<tbody>
<tr>
<td>Normal stress $\sigma_{sh}$ 1</td>
<td>4 kPa</td>
<td>2 kPa</td>
</tr>
<tr>
<td>Normal stress $\sigma_{sh}$ 2</td>
<td>8 kPa</td>
<td>4 kPa</td>
</tr>
<tr>
<td>Normal stress $\sigma_{sh}$ 3</td>
<td>12 kPa</td>
<td>6 kPa</td>
</tr>
<tr>
<td>Normal stress $\sigma_{sh}$ 4</td>
<td>4 kPa</td>
<td>2 kPa</td>
</tr>
</tbody>
</table>

**Figure 3.3:** a) Shear stress plot and resulting yield locus as constructed with a ring shear tester and Mohr stress circles ($\sigma_1$ consolidation stress, $\sigma_c$ unconfined yield strength) from (Schulze, 2006).
To interpret these measurements, two Mohr stress circles are drawn. The larger stress circle represents the stress state in the bulk solid sample at steady-state flow (i.e. the stress state at the end of preshearing). The major principal stress of this Mohr circle defines the consolidation stress $\sigma_1$. The major principal stress of the smaller Mohr stress circle is the unconfined yield stress $\sigma_c$. The tangent of the larger Mohr circle passing through the origin defines the effective yield locus and the angle is the internal angle of friction at steady-state flow $\varphi_{sf}$. For dry powders, Coulomb’s law of friction gives the relationship between shear stress $\tau$ and normal stress $\sigma$ given by:

$$\tau = \sigma \tan \phi + c_f$$  \hspace{1cm} (3.1)

where $\phi$ is the angle of internal friction and $c_f$ is a parameter representing the cohesive properties of bulk solids ($c_f = 0$ for cohesionless materials). For certain applications, it is more useful to define the angle of internal friction as static coefficient of internal friction $\mu_f$ where $\tan \phi = \mu_f$. Cohesive powders in general gain strength under the action of compacting stresses. Similarly, the adhesive frictional forces between granular material and mixer wall can be written as:

$$\tau_w = \sigma_w \tan \phi_w + c_w$$  \hspace{1cm} (3.2)

where $\phi_w$ is the wall angle of friction and $c_w$ is the wall adhesion and $\tan \phi_w$ is the coefficient of wall friction, $\mu_w$.

The flowability $ff_c$ is defined as the ratio of the principle consolidation stress $\sigma_1$ to the unconfined yield stress $\sigma_c$:

$$ff_c = \frac{\sigma_1}{\sigma_c}$$  \hspace{1cm} (3.3)

The classification of the flowability values according to Jenike (1964) used for silo design are outlined in Table 3.5.

<table>
<thead>
<tr>
<th>$ff_c$</th>
<th>Description</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>$&lt; 2$</td>
<td>Very cohesive, non-flowing</td>
<td>Cohesive powders</td>
</tr>
<tr>
<td>$2 &lt; ff_c &lt; 4$</td>
<td>Cohesive</td>
<td></td>
</tr>
<tr>
<td>$4 &lt; ff_c &lt; 10$</td>
<td>Easy flowing</td>
<td>Non-cohesive powders</td>
</tr>
<tr>
<td>$ff_c &gt; 10$</td>
<td>Free flowing</td>
<td></td>
</tr>
</tbody>
</table>

The wall angle of friction $\phi_w$ was also measured with the Schulz Ring Shear Tester RST-XS using a wall friction cell and a sample piece of stainless steel which is the same material as the impeller blades and mixer wall.
Materials and Methodology

The results of the bulk properties tests for dry sand and cohesive sand at two different compressive loads are shown in Table 3.6. According to the Jenike’s flowability classification table (Table 3.5) “cohesive” sand is considered an easy-flowing, non-cohesive powder (Table 3.6). “Cohesive” is used throughout this study as a relative term to distinguish “sticky” cohesive sand with dry free flowing sand. Therefore, caution is advised when inferring results of cohesive sand with other true cohesive powders as classified by Jenike.

<table>
<thead>
<tr>
<th>Bulk Properties</th>
<th>Dry Sand</th>
<th>Cohesive Sand</th>
</tr>
</thead>
<tbody>
<tr>
<td>Consolidating load $\sigma_{pre}$ (Pa)</td>
<td>16 000</td>
<td>8 000</td>
</tr>
<tr>
<td>Unconfined yield stress $\sigma_c$ (Pa)</td>
<td>785</td>
<td>461</td>
</tr>
<tr>
<td>Angle of internal friction $\phi$ (°)</td>
<td>37.4</td>
<td>39</td>
</tr>
<tr>
<td>Bulk density $\rho_b$ (kg/m$^3$)</td>
<td>1 380</td>
<td>1 375</td>
</tr>
<tr>
<td>Cohesion $c_f$ (Pa)</td>
<td>196</td>
<td>115</td>
</tr>
<tr>
<td>Flowability $ff_c$</td>
<td>39</td>
<td>35</td>
</tr>
<tr>
<td>Flow description Free flowing</td>
<td>Free</td>
<td>Easy flowing</td>
</tr>
<tr>
<td>Angle of wall friction $\phi_w$ (°)</td>
<td>10</td>
<td>10.5</td>
</tr>
<tr>
<td>Wall adhesion $c_w$ (Pa)</td>
<td>43</td>
<td>144</td>
</tr>
</tbody>
</table>

3.3 Mixer design, construction and control

To study powder flow, a new vertical-bladed mixer granulator was designed and constructed. The new mixer was designed to be compatible with the detection range of the new positron camera (Parker et al., 2002). The mixer motor is an Eurotherm Drives 5.5 kW AC motor. The bottom-driven motor shaft was controlled by a variable speed control box (Eurotherm 690+ Integrator) that enabled the impellers to rotate at speeds of 0-667 rpm. The speed can be manually adjusted using the up or down button to obtain the desired speed. The drive shaft assembly attaching the motor drive shaft to the mixing bowls is of similar dimensions to that used by (Wellm, 1997, Knight et al., 2001) to take advantage of existing interchangeable bowls and impellers. To enable torque measurements, the mixing vessels rested on a ring of ball bearings allowing free motion of the mixing bowl. An s-bend torque meter is used to measure instantaneous torque and the readings are displayed on an electronic display box.

The 0.21 m diameter mixing bowl with a volume capacity of 6.9 L is used for this investigation (Fig. 3.6). The mixer bowl was constructed from stainless steel pipes and welded closed at one end with a circular steel piece. For the 2kg powder load, the ratio of powder bed height to bowl diameter $H/D$ was approximately 0.29.
Figure 3.4: Photographs of a) the high shear mixer granulator and PEPT set-up, b) side view of the vHSM, c) mixer bowl and connector shaft assembly and d) mixer motor.
Figure 3.5: Photographs of a) the control box and wiring b) top view of mixer bowl and torque arm, c-d) ball bearing flowing assembly, e) s-bend torque arm measuring device, f) torque reader, g) side view of 0.21 m mixing bowl and h) top view of mixer bowl with rectangular impeller blade.
3.4 Torque measurements

Torque monitoring has been used in industry for monitoring wet mass quality and especially for process end-point detection. In this study, mixer torque rheometry was used to investigate the flow behaviour of powders.

Torque experiments were carried out at the University of Birmingham. Torque measurements were sampled every 0.5-1 Hz by a data logger. Errors in the torque measurements arose from friction in the drive shaft bearings. Accuracy were estimated to be accurate to $\pm 10\%$.

Two different types of impellers were used as shown in Figure 3.7. A $90^\circ$ rectangular two-bladed impeller and a $10^\circ$ triangular two-bladed impeller. These impellers were designed to be similar to impellers found in commercially available mixers (e.g. Aeromatic-Fielder) with a height and width of 10 mm and 50 mm, respectively. The triangle impeller was designed with a 1 mm front edge face to avoid sharp edges. These types of impellers produced both impact and shear forces within the mixer. No chopper blades or baffles were present in the mixer to simplify the flow field.

Figure 3.6: Schematic isometric views of mixing vessel with torque arm. Dimensions are in mm.
Materials and Methodology

Figure 3.7: Photographs of impeller blades a) top view and b) front view and schematic isometric views of the c) 90° rectangular two-bladed impeller and d) 10° triangular two-bladed impeller. Dimensions are in mm.
3.5 Positron Emission Particle Tracking (PEPT) Experiments

In order to investigate the motion of particles beneath the surface, the non-invasive PEPT technique was employed to obtain direct measurements of powder flow. This technique, developed at the University of Birmingham, involved the use of a radioactively labelled tracer particle to reveal the three-dimensional Lagrangian trajectory of flowing solid particles (Parker et al., 1993). The resin beads were labelled using ion exchange techniques and have a half life of approximately 110 min (Fan et al., 2006a,b, Parker and Fan, 2008). The radioactive tracer undergoes $\beta^+$ decay emitting positrons. When a positron annihilates with an electron, a collinear pair of $\gamma$-rays are emitted back-to-back in opposite directions. This collision event is captured by the positron camera which is made up of two positron sensitive detectors each 50 x 40 cm$^2$ and situated 54 cm apart. A location algorithm then calculates the incidence coordinates by triangulation of events that converge on a single point (Figure 3.8).

![Figure 3.8: Positron emission particle tracking in a mixer granulator: 1) motor and control box, 2) mixer vessel, 3) impeller, 4) torque arm, 5) PEPT tracer, 6) powder bed, 7) gamma-ray detectors.](image)

The capabilities of the new Birmingham camera can follow a tracer moving at 1 m/s to within 5 mm at a data acquisition rate of over 250 times per second (Parker et al., 2002). This allows studies of powder motion moving at high speeds in vHSM. Detailed description of the technique, construction and operation of the PEPT are discussed by Parker et al.
(1993) and Parker et al. (2002). The set-up for the PEPT experiment with the laboratory scale mixer granulator positioned between the two detectors is shown in Fig. 3.4a.

In a typical experiment, the powder was premixed for two minutes to achieve a steady state before the tracer was added to the bulk flow and data acquisition commenced. Each experiment was run for approximately one hour. A summary of the 14 PEPT experiments carried out is given in Table 3.7.

<table>
<thead>
<tr>
<th>Material</th>
<th>Impeller design</th>
<th>Impeller speed (rpm)</th>
<th>100</th>
<th>200</th>
<th>250</th>
<th>300</th>
<th>350</th>
<th>400</th>
<th>500</th>
<th>600</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cohesive sand</td>
<td>90° Rectangular</td>
<td>□</td>
<td>■</td>
<td>■</td>
<td>▲</td>
<td>▲</td>
<td>▲</td>
<td>▲</td>
<td>▲</td>
<td>▲</td>
</tr>
<tr>
<td>Cohesive sand</td>
<td>10° Triangular</td>
<td></td>
<td>▲</td>
<td>■</td>
<td>▲</td>
<td>▲</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dry sand</td>
<td>90° Rectangular</td>
<td>□</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dry sand</td>
<td>10° Triangular</td>
<td>□</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Powder flow regimes: □ bumping, △ apparent roping; ○ roping. Shading denote experiments where the ergodicity theorem was applicable.

**3.5.1 Ergodicity criterion**

While each PEPT experiment was run for one hour, results showed that for two runs the ergodicity criterion was not achieved, that is the tracer particle did not visit all regions of the powder bed (see Figure 3.9). The dry sand experiments at low Froude number ($Fr \leq 1.2$) using the rectangular and triangular impeller exhibited “dead zones” at the bed surface near the wall or in the inner core region of the bed. These regions represented areas in which the transfer of particles across solid body rotation was not achieved within the given experimental time. Given that the granulation process in a commercial granulator mixer typically takes in the order of five minutes to complete, operating at low Froude number is undesirable. Even though the ergodicity criteria was not achieved for these two experiments, interesting qualitative observations can still be obtained from the results.

**3.5.2 Tracer segregation**

Ideally, a tracer particle would be selected from the bulk sample under investigation to ensure that the PEPT data reflect the behaviour of the bulk flow within the mixer. However due to the small size and frictional attrition of the sand surface in the mixer, an alternative tracer particle was chosen. A 212 µm resin bead of similar size to that of the sand was selected.
3.5 Positron Emission Particle Tracking (PEPT) Experiments

The presence of dead zones showed that the ergodicity criteria was not achieved using dry sand with either the a) rectangular or b) triangular impeller at low impeller speed of 100 rpm. The blade boundary is indicated by ———.

with a density of 1200 kg/m³, which is lower than that of sand 2500 kg/m³. This may be a possible source of error due to segregation. However, previous studies have found these effects to be minimal (C.J. Broadbent and Parker, 1995, Knight et al., 2001).

C.J. Broadbent and Parker (1995) found that the size of the spherical tracer (2-8 mm) within the bulk material (rice grains 5 mm long 2 mm diameter) had little effect on the bulk motion, velocities and the residence time distribution. However, it was noted that that the size of the tracer has significant influence on the transfer rate between the two halves of the horizontal mixer separated vertically along the axis.

Knight et al. (2001) made measurements in the new and more sensitive PEPT camera, that shown that coarse size tracers are not subject to significant segregation compared to smaller tracers.

3.5.3 Limitations of PEPT detection and resolution

The data acquisition rate was not constant, with rates averaging between 12 - 113 Hz. The rate of detection of an event is dependent on three factors: 1) the radioactivity of the
Materials and Methodology

tracer which decreased exponentially with time, 2) the position of the tracer relative to
the detectors described as geometric efficiencies by Seville et al. (2009) and 3) the velocity
of the tracer. With a tracer half-life of 110 min, the decay rate of the tracer was slow
enough so that the experiments were not effected over the duration of the experiment.
Geometric efficiencies effected the tracer’s detection rate as the opposing configuration of
the two detectors increased the probability of detection of collinear event (Seville et al.,
2009). Figure 3.10 demonstrates how those events closest to the centre of the detector
(A) have a higher probability of being accurately detected than those at the outer edge
of the non-detector side (C). Experiments carried out with only the tracer glued onto the
impeller blade at 33 and 566 rpm showed the dramatic effect of increasing tracer speed on
PEPT detection rates (Fig. 3.11). At high tracer speeds, detection rates were lower due to
geometric efficiencies and a reduce ability to triangulate the tracer position. To enable the
use of PEPT data at higher speeds, the raw PEPT data was resampled as described below.

3.6 PEPT Data Analysis

3.6.1 Preprocessing of data

In order to use PEPT data to understand powder flow, further data processing was required.
MATLAB was used as the platform to develop these algorithms. Cartesian coordinates were
assigned so that the x-axis was parallel to the detectors, the y-axis was orthogonal to the
detectors and the z-axis was vertical, with the origin centred at the base of the mixer (see
Figure 3.8). A typical example of the positional-time series measurements obtained by the

\begin{figure}
\centering
\includegraphics[width=0.5\textwidth]{geom.png}
\caption{Figure 3.10: Geometric efficiencies (Seville et al., 2009).}
\end{figure}
PEPT technique in the vHSM is shown in Figure 3.12.

To overcome asymmetrical detection rates, the data was re-sampled at a constant time interval of 120 Hz for all data sets except for cohesive sand using rectangular blade at 600 rpm which was sampled at 500 Hz due to very high particle speeds. Also, the Cartesian coordinates were converted to cylindrical coordinates for useful interpretation of the data of the cylindrical mixer.

3.6.2 Single particle trajectory

The simplest analysis of the flow field using PEPT data consist of following the continuous Lagrangian trajectory of a tracer particle. Lagrangian refers to the method of viewing particle motion where the observer follows an individual particle as it moves in space and time. From the Lagrangian specification, rotational frequencies can be measured. A new parameter \( \phi_V \) was introduced as a measure of vertical powder turnover. This term is the average frequency at which the Lagrangian trajectory of a tracer completed a vertical rotation above a given threshold height. The vertical turnover frequency was measured from the axial trajectory of the tracer, recording the frequency of the tracer as it moves from an absolute minimum through to a distance greater than a threshold distance \( h_t \), and returning to the next absolute minimum (see Figure 3.13). The threshold height was assigned according to the height at which distinct vertical circular motion was observed. This parameter was also used as a simple quantitative measure of mixing in vertical high shear mixers.

Using a similar approach, the \( \phi_H \) was obtained from the horizontal position of the tracer. The trajectory in the plane perpendicular to the two PEPT detector plates (y-axis) was used instead of the parallel plane (x-axis) to reduce errors caused by geometric efficiencies as discussed in Section 3.6.

3.6.3 Lagrangian properties

Secondary analysis required further data processing to obtain information on velocities from positional-time series data. Lagrangian velocities were calculated along the trajectory of the tracer using the velocity gradient method, also used in previous studies (Bakalis et al., 2003). Regression analysis was performed on each Cartesian coordinate time series where a linear segment was fitted to \( i \) successive data points using the following relationships:

\[
v_x = \frac{dx}{dt}, \quad v_y = \frac{dy}{dt}, \quad v_z = \frac{dz}{dt}
\]  

(3.4)
The velocity component $v_x$ was obtained from the gradient of the line. The coordinates of the velocity component was taken as the average of the $i$ data points for each direction (see Figure 3.14). Typically, five consecutive data points were used to estimate the velocity. A number higher than this may smooth out significant bumping motion. The resultant velocity magnitude $v_p$ was calculated by:

$$v_{p, \text{point}} = \sqrt{v_x^2 + v_y^2 + v_z^2}$$ (3.5)

and then converted to cylindrical coordinates using:

$$v_r = v_x \cos \theta + v_y \sin \theta$$ (3.6)

$$v_\theta = -v_x \sin \theta + v_y \cos \theta$$ (3.7)

$$v_z = v_z$$ (3.8)

where the negative in Equation 3.6b denotes clockwise motion.

### 3.6.4 Eulerian maps

While following the Lagrangian trajectory of a single particle can yield useful data, more interesting properties of bulk flow and mixing can be derived from an Eulerian specification, where the motion of the powder flow is focussed on specific locations in the space over time. An analogy of an Eulerian map is viewing a flow field while sitting on the bank of a river and watching the water pass the fixed location. In contrast a Lagrangian specification is viewing the flow while sitting on a boat drifting down a river.

The region of the mixing vessel with powder material was discretised into 19200 cells of equal volume. The number of cells in the radial, axial and tangential direction were $n_r = 20$, $n_z = 24$ and $n_\theta = 40$ respectively. Assuming axial symmetry within the cylindrical vessel, a 2D azimuthal view was presented by collapsing the tangential component into a 2D radial-axial plane with annular rings of equal volume.

### 3.6.5 Occupancy

Using a user-defined cylindrical grid of equal-volume annular cells, the occupancy of each cell $occ_{cell}$ is defined as the fraction of time the tracer spent in each cell of the overall experimental time $t_T$ given by:
3.6 PEPT Data Analysis

![Figure 3.11](image)

**Figure 3.11:** Tracer experiments at a) 33 rpm and b) 566 rpm.

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$n_{cell}$</td>
<td>19200</td>
</tr>
<tr>
<td>$vol_{cell}$</td>
<td>216 mm³</td>
</tr>
<tr>
<td>$vol_{annular}$</td>
<td>8659 mm³</td>
</tr>
<tr>
<td>$n_r$</td>
<td>20</td>
</tr>
<tr>
<td>$n_\theta$</td>
<td>40</td>
</tr>
<tr>
<td>$n_z$</td>
<td>24</td>
</tr>
<tr>
<td>$deg_\theta$</td>
<td>9°</td>
</tr>
<tr>
<td>$h_z$</td>
<td>5 mm</td>
</tr>
</tbody>
</table>
Materials and Methodology

**Figure 3.12**: Typical time data series of the Cartesian coordinates of PEPT experiments.

**Figure 3.13**: The vertical bed turnover frequency was determined by the rotational frequency at which the tracer moved over a certain threshold height. The dots represent the maximum and minimum position of each vertical rotation cycle in the a) radial-axial plane and b) axial position versus time plot.

**Figure 3.14**: Velocity gradient method used to estimate velocity component from a tracer trajectory in each coordinate direction.
where \( \tau_{\text{cell}} \) is the residence time of each pass through the cell. Note that only cells in which the tracer is observed is considered in the compartments. Alternatively, occupancy can be evaluated from a virtual bed of discrete particles generated by dividing the Lagrangian trajectory into strings at constant time intervals (Fig. 3.15b). Occupancy is then the ratio of the number of particles (strings) contained in a cell \( n_p \), to the total number of particles in the mixer \( n_T \).

The average residence time of a cell \( \tau_{\text{cell}} \) is given by:

\[
\tau_{\text{cell}} = \frac{\sum \tau_{\text{cell}}}{n_{\text{pass}}} \approx \frac{n_p}{n_T} \tag{3.10}
\]

where \( n_{\text{pass}} \) is the number of tracer visits through the cell and \( k \) is the constant time interval between each particle. Internal properties of the cells (e.g. particle velocities in each component, overall 3D and 2D (radial-axial) particle velocity, standard deviation of the velocity, tracer acceleration, number and frequency of visits are also obtained.

\[\text{occ}_{\text{cell}} = \frac{\sum \tau_{\text{cell}}}{t_T} \approx \frac{n_p}{n_T} \tag{3.9}\]

Figure 3.15: Generating a virtual cluster of particles from a single particle trajectory using the a) box method and b) the constant time interval method from Doucet et al. (2008)

Compartment properties

The volume of a compartment is the total volume of annular cells with particles present, regardless of if the cells are only partially filled as in the case with surface cells. The occupancy of each compartment \( \text{occ}_{\text{comp}} \) is the sum of \( \text{occ}_{\text{cell}} \) within the compartment (Eqn. 3.9). The average velocities in each compartment were obtained by multiplied the cell properties by \( \text{occ}_{\text{cell}} \) and summing the values of that compartment.
3.6.6 Bed density

Occupancy can also be viewed as roughly equivalent to a time-averaged bed density $\rho_{bed}$ given by:

$$\rho_{bed} = \frac{occ \ast m}{v_{cell}}$$

(3.11)

where $m$ is the overall mass of powder bed and $v_{cell}$ is the volume of an annular cell and $occ$ is the occupancy in that cell. As $m$ and $v_{cell}$ are constant $\rho_{bed} \propto occ$.

3.6.7 Eulerian velocities

The average powder velocity of each cell was calculated by taking the resultant velocity magnitude (Eqn. 3.5) from the velocity component, and averaging over the number of particles visits. The overall powder velocity $v_p$ in the mixer was then obtained by summing all the average powder velocity in each cell $v_{p,cell}$ multiplied by occupancy (Eqn. 3.9):

$$v_p = \sum_{j=1}^{n_{occ}} v_{p,cell} \ast occ$$

(3.12)

where $n_{occ}$ is the total number of occupied cells. It was important to factor in the time spent in each cell as occupancy varies considerable between the cells, particularly near the surface and around the impeller where the cells are only partially filled.

3.6.8 Fourier analysis

Fourier analysis was used on the PEPT data to examine the sinusoidal components of the particle trajectory over time. As the acquisition of the 3D positional coordinates $P_{xyz}(t)$ was determined by the random nature of $\beta^+$ decay and its detection by the PEPT cameras, the data was inherently non-equidistant. Each data set was re-sampled in order to obtain equidistant samples before fourier analysis. High sampling rates of PEPT data (e.g. a minimum of 120 Hz), reduced any potential errors from aliasing. The function to implement the fast fourier transform (FFT) for vectors of length $N$ was given by:

$$X(k) = \sum_{j=1}^{N} x(j)\omega_N^{(j-1)(k-1)}$$

(3.13)

where

$$\omega_N = e^{-2\pi i/N}$$

(3.14)
is an \( N \)th root of unity. The FFT was then multiplied by its complex conjugate to obtain the power spectrum. The resulting plot allowed easy detection of any dominant periodic components of the data.

### 3.6.9 Shear rates

Shear plays an important role in the flow behaviour and mixing of powders in high shear mixers. Shear stress exists whenever there is relative motion of layers of particles and is a function of shear rate. A simplified approach of estimating shear rates (\( \gamma \)) in a vertical mixer is to consider a one-dimensional view of the shear rate (Figure 3.16).

![Figure 3.16: Diagram of 2D shear rate calculations.](image)

Significant shear occurs perpendicular to the direction of the moving impeller and radial and axial velocities are typically an order of magnitude smaller than tangential velocities, therefore only tangential velocity gradients are used in shear rates estimates. The shear rate of each cell is given by:

\[
\begin{align*}
\gamma_{\theta(r)} &= \frac{\Delta v_\theta}{\Delta r} \quad (3.15) \\
\gamma_{\theta(z)} &= \frac{\Delta v_\theta}{\Delta z} \quad (3.16) \\
\gamma_{\theta(rz)} &= \sqrt{\gamma_{\theta(r)}^2 + \gamma_{\theta(z)}^2} \quad (3.17)
\end{align*}
\]

where \( \gamma_{\theta(r)} \) and \( \gamma_{\theta(z)} \) is the tangential shear rate in the radial and axial direction and \( \gamma_{\theta(rz)} \) is the overall magnitude of the tangential shear rates, \( v_\theta \) is the average tangential velocity of each cell, \( r \) is the radial distance and \( z \) is the axial distance.
3.6.10 Mixing

The phenomenon of powder mixing occurs by three mechanisms: convection, dispersion and shear (Portillo et al., 2008). One way to characterise the degree of mixing is by creating a mixture of different colour particles, which have identical properties except for colour. The multi-CM was segregated into three coloured species: red, blue and black particles to identify particles in the impeller, circulation and surface compartments respectively. The evolution of particle mixing was followed over 20 revolutions in the vertical azimuthal projection (Fig. 4.19). All impeller speeds followed a general roping pattern with particles moving from the impeller blade, up the wall and back down the face of the powder bed except for 100 rpm, which had a bi-directional flow. In the analysis of mixing in the horizontal plane, the mixer is split vertically into two halves with one side consisting of red particles and the other blue. The red particles were followed for 20 revolutions (Fig. 4.19).

Statistical analysis was used to determine the homogeneity of the mixture which was represented by the relative standard deviation (RSD) given by (Remy et al., 2009):

\[
RSD = \frac{\sigma_{conc}}{\bar{x}_{conc}}
\]

(3.18)

where \(\sigma_{conc}\) is the standard deviation of the particle concentration of all samples taken and \(\bar{x}_{conc}\) is the overall mean particle concentration. The lower the RSD value, the better the mixing as less variability exists between samples. It should be noted that RSD is highly sensitive to the sample size and sample number and care was taken to obtain a RSD value that was independent of sampling biases by varying the sampling grid.

3.7 Summary

Formulation properties of two model powders, dry and cohesive sand have been characterised. The process parameters and key design features of the vHSM granulators were discussed and used to develop a systematic approach to investigate powder flow behaviour.

A range of tools have been identified for quantitative analysis of the motion of particulates within a mixer granulator. These included the powerful non-invasive imaging technique of PEPT and equipment for torque measurements. In conjunction with these tools, a detailed analysis of the data processing used to obtain useful information from the tools was outlined. In the following chapters, an investigation was carried out using these tools to fully characterise the powder flow behaviour in mixer granulators.
3.7 Summary

Nomenclature

\( \dot{A} \)  
area flux of powder through the spray zone \((m^2.s^{-1})\)

\( d_d \)  
average liquid drop size \((m)\)

\( \dot{V} \)  
volumetric spray rate \((m^3.s^{-1})\)

\( U_c \)  
representative collision velocity in the granulator \((m.s^{-1})\)

\( w \)  
mass ratio of liquid to solid \((-)\)

\( Y_g \)  
granule dynamic yield stress \((kPa)\)

Greek symbols

\( \varepsilon_{\text{min}} \)  
minimum porosity the formulation \((-)\)

\( \rho_g \)  
granule density \((g/mL)\)

\( \rho_l \)  
liquid density \((g/mL)\)

\( \rho_s \)  
solid particles density \((g/mL)\)

\( \Psi_a \)  
dimensionless spray flux \((-)\)
4

Powder Flow Regime Map

Abstract

Particle motion in vertical-axis bladed mixer-granulators is complex, typically showing non-trivial flow behaviour with an increase in blade speed from intermittent vertical motion in sequence with the blade passage (“bumping regime”) to toroidal “roping regime”. The transitions speed depends \textit{inter alia} on blade and mixer design, formulation properties and operational parameters. This investigation observed that cohesive sand exhibited \textit{multiple} transitional states instead of \textit{one} transition reported in the literature (Litster et al., 2002 Litster et al. (2002)). This interesting phenomenon effects scale up of granulating systems as regular roping motion is essential for controlled agglomeration. A powder flow regime map for bladed, vertical-axis mixers is proposed based on \textit{powder Froude number} and dimensionless \textit{Bed Resonance number}. The chapter is an experimental validation of earlier theories relating the motion and torque for stirring powders, using Positron Emission Particle Tracking to follow the particle motion. The powder flow regime map provided a greater understanding of the effects of formulation properties, operating and design parameters on granulator performance and offers a rational approach to scale-up of mixer granulators.
4.1 Introduction

Using qualitative visual observations, Litster et al. (2002) defined two distinct flow regimes in vertical mixer granulators using dry lactose. At low impeller speed, the powder moved slowly around the mixer in the direction of the impeller blade with the surface of the powder remaining generally horizontal. The powder appears to bump as a static bed due to the intermittent vertical motion in sequence with the blade passage beneath the bed (see Figure 2.2a). This behaviour has been described as bumping flow (Litster et al., 2002), and also as shunting (Hiseman et al., 2002). It has been reported by numerous researchers (Stewart et al., 2001a, Litster et al., 2001, 2002, Plank et al., 2003) of whom nearly all focussed on understanding the fundamentals of mixing and segregation at low impeller speeds and not at higher speeds that are typically used for industrial processes.

With increasing impeller speeds, chaotic flow transitions was reported using lactose (Litster et al., 2002). A flow transition occurred with an increase in impeller speed, in which unstable behaviour was observed with powder walls collapsing causing large amounts of dust. Knight et al. (2001) observed a sharp decrease in the magnitude of the torque at a critical speed. No further description of the flow profile during this period was reported.

At high impeller speed, a significant movement in the circumferential direction combined with a secondary toroidal roping motion was observed. Numerous researchers have described helical or toroidal flow patterns in the powder flow resulting from a combination of gravitational, centripetal, centrifugal and impact forces (Schaefer et al., 1993, Ramaker et al., 1998, Knight et al., 2001). This regime has been given the term roping regime by Litster et al. (2002) (see Figure 2.2b).

The formation of this stable flow pattern promoted high vertical bed turnover which is essential for controlled granulation (Hapgood, 2000). While it is desirable to operate within the roping regime, optimal roping patterns are achieved at a critical transition point and any further increase in impeller speed after this point has little effect on the powder flow profile (Knight et al., 2001). Operations in the bumping flow is highly undesirable as the poor bed turnover rate allowed only a small region of the powder to experience high shear rates, leading to high attrition and non-uniform particles. Binder addition in the bumping regime would lead to caking and pooling on the powder surface, resulting in non-uniform size distributions (Hapgood et al., 2003).

For the reasons stated above, the transition from the gravity dominated bumping regime to the rotational inertia dominated roping regime is a matter of keen interest for many researchers as it has practical implications for industry.

Formulation properties such as cohesion and particle size distributions complicate matters
to a greater extent with few studies focused on the effect of these variables on the most simple systems (Litster et al., 2002, McCarthy, 2003). In understanding the dynamic kinetics of mixer granulators, it is important to also consider the complex cyclical interaction of powder bed with a series of impeller blades, particularly at high speeds. This concept is rarely considered in previous studies of mixers.

4.1.1 Powder flow dimensionless groups

In the pharmaceutical industry, operators have used a range of dimensionless groups including Froude number, swept volumes, power number, reynolds number (Faure et al., 1999) to characterise the geometric and dynamic similarities of the large variety of granulation mixers available to varying degrees of success. The recent advancement in the granular flow theory to develop an overarching theory to encompass a wide variety of granular flow experienced in a range of geometries had lead to the introduction of dimensionless shear rate $I$ (GRD Midi, 2004). Froude number and the dimensionless shear rate are discussed below in relation to vertical-axis bladed mixers.

Froude number

The Froude number was first used in naval architecture to compare geometrically similar hulls that were also dynamically similar in terms of wave resistance Froude (1874). Nowadays, it has found widespread application in mixing applications. Rumpf and Müller (1962) used the Froude number $Fr_I$ as a dimensionless rotating frequency to compare different operating conditions of various mixing equipment. The dimensionless number represents the ratio between centrifugal and gravitational acceleration given by:

$$ Fr_I = \frac{\omega_I^2 R}{g} $$  \hspace{1cm} (4.1)

where $\omega_I$ denotes the angular rotational speed of the impeller (rad.s$^{-1}$), $R$ is the mixer radius (m) and $g$ is the gravitational acceleration constant (9.81m.s$^{-2}$). Another common variation of the Froude equation used in the granulation literature is:

$$ Fr_I = \frac{DN^2}{g} $$  \hspace{1cm} (4.2)

where $D$ is the impeller diameter and $N$ is the impeller rotational speed (rpm). Both Eqn.4.1 and 4.2 centre on variables relating to impeller speed. Researchers have attempted to predict the transitional impeller speed for scale-up based on this method of balancing the rotational inertia of the impeller with gravity with mixed results (Ramaker et al., 1998,
Litster et al., 2001). The limitations of using the Froude number to scale up is that it only considers the impeller speed and mixer radius and not material properties.

Knight et al. (2001) introduced the *powder Froude number*, a modified version of the impeller Froude number (Eqn. 4.1) that takes into account powder bed behaviour and is the ratio of powder centripetal acceleration to gravity given in terms of powder angular rotational speed $\omega_p$ or averaged characteristic powder velocity $v$:

$$Fr_p = \frac{\omega_p^2 R}{g} = \frac{v^2}{gR}$$  \hspace{1cm} (4.3)

where $R$ is the mixer radius and $g$ is the gravitational constant. To take into account mixer fill levels and the changing bed shape, the mixer radius $R$ has been replaced with toroid centroid radial position $R_c = (R - R_o)/2 + R_0$ where $R_0$ is the inner radius of the toroid and the characteristic velocity is defined as the averaged powder tangential velocity $v_\theta$ to give:

$$Fr_p = \frac{v_\theta^2}{gR_c}$$  \hspace{1cm} (4.4)

### 4.2 Proposed Powder Flow Regime Map

To develop the regime map, consider the powder flow behaviour in a cylindrical mixer at the particle or powder element level. Figure 4.1 shows the trajectory of a tracer particle or powder element relative to the passage of impeller blades beneath the bed. The process of momentum transfer from rotating impeller blades to the powder bed is predominately controlled by impeller speed.

At low impeller speeds (a-d) the particle motion is dominated by gravitational forces. It is defined here as the *bumping regime*. In the particle simplest motion, the particle is lifted to form a “bump” in front of the approaching blade (a). This bump is eventually pushed over the blade, forming a complete rise and fall trajectory under the influence of gravity. A rest period exists between each blade pass producing the characteristic bumping behaviour at the powder bed level. When the impeller speed is increased somewhat (b), the trajectory of the first blade pass will overlap with the rising heap of the next approaching blade, eliminating the rest period between the interacting bumps. These multiple bumps interactions reduce the significance of the bumping behaviour and promote vertical circulation patterns in the bed, an *apparent roping* behaviour. However, if the impeller speed continues to increase (c), the bed collapses dramatically as the powder falls to the base of the mixer causing a shift back to the simplest bumping motion. The sequence of the blade passes relative to
the powder bed produces a “phantom”, non-interacting blade that passes beneath the bed, enabling the particle to complete a rise and fall trajectory with a rest period in between each blade pass. Once again, the characteristic bumping behaviour is exhibited in the powder flow. This interesting resonance in the mixer is a product of a characteristic timescale for the bed to settle. If the impeller speed is increased even further, the powder flow returns to the apparent roping behaviour with bumps forming multiple interactions (d).

At a critical speed (e), the steadily increasing effect of rotational forces overcomes gravitational forces and the particle motion is no longer dominated by the bumping motion. Instead, the dominant flow behaviour of the powder is a second order roping motion, exhibiting large rise and fall recirculation trajectories at the powder bed scale. This is defined as the roping regime.

The transitions between these regimes are governed by two key dimensionless groups, the Bed Resonance number and the powder Froude number which takes into account the effects of material properties in the impeller zone, the region of most significance to the powder flow behaviour.
4.2 Proposed Powder Flow Regime Map

The transition to roping regime (e) corresponds to the critical speed at which the powder Froude number is equal to unity. This term has been denoted as the critical powder Froude number given by:

\[ Fr_c = \frac{v_c^2}{R_c g} = 1 \] (4.5)

The significance of this parameter is that it demarcates the transition from bumping to roping. Physically, it marks the point at which centripetal acceleration exceeds gravity within the powder bed. Ideally for granulation, operators should operate beyond critical Froude number to achieve stable roping flow behaviour.

The multiple transitional states in the bumping regime (Fig. 4.1a-d) are demarcated by the dimensionless Bed Resonance number \( \beta \), which is the relationship between the bulk powder motion in the vertical direction and blade interactions. \( \beta \) is defined as:

\[ \beta = \frac{t_a}{t_b} = t_a \omega_b \] (4.6)

where \( t_a \) denotes the powder bump period - the time required for a complete rise and fall trajectory from a blade passing beneath the powder bed, \( t_b \) the period between blade intervals relative to the powder and \( \omega_b \) is the blade frequency relative to the powder (see Figure 4.2).

![Figure 4.2](image)

**Figure 4.2:** Schematic diagram of the powder bump height \( h_a \), powder bump period \( t_a \) and the blade interval period \( t_b \) measurements used to calculate Bed Resonance number.

The powder flight time \( t_a \) was estimated from PEPT data. It could also be derived from fundamental principles of the unhindered rise and free fall of a particle under the influence of gravity. As the vertical displacement of the powder \( h_a \) at the unconfined surface can be obtained experimentally using PEPT, flight time is given by:
Powder Flow Regime Map

\[ t_a = 2t_{\text{rise}} = 2 \sqrt{\frac{2h_a}{g}} \] (4.7)

The critical bumping transition point occurs when \( \beta \) equals unity.

\[ \beta_c = \frac{t_a}{t_b} = t_a \omega_b = 1 \] (4.8)

Based on these concepts, a regime map is proposed for the flow behaviour in a bladed, vertical high shear mixer (see Figure 4.3). The powder Froude number lies on the horizontal axis and describes the balance between gravitational and rotational forces while the Bed Resonance number lies on the vertical axis and describes the relationship between the blade sequence and bulk powder behaviour.

![Regime Map Diagram](image)

**Figure 4.3:** Proposed powder flow map for bladed, vertical high shear mixers.

In the following sections, the validity of the proposed powder flow regime map was tested with PEPT data that can directly measure \( t_a, t_b \) and \( v_\theta \) in powder flow experiments and compare the observed flow regime with that predicted by Figure 4.3.
4.3 Experimental methodology

The experiments were carried out in a 0.21 m diameter, vertical cylinder mixer with a variable control speed drive ranging from 0-670 rpm. A 90° two-bladed rectangular impeller was used with design parameters of 50 mm width, 10 mm height, 1 mm clearance between the end of the blade and the wall and 3 mm between the bottom edge of the blade and base of the bowl. The mixer shaft was mounted to the motor and control box at the bottom of the mixer (refer to Figure 3.8).

A cohesive sand mixture was produced by mixing 2 kg of dry sand (size distribution of 150-250 µm) with 50 mL of silicone oil with a viscosity of 0.1 Pa.s (Fluka Products, Sigma-Aldrich, Australia). The cohesive sand was prepared by kneading the mixture in a plastic bag for 5 minutes to obtain a uniform and reproducible starting mass. A summary of the material properties is found in Table 3.1.

4.3.1 PEPT experiments

Direct measurements of three-dimensional motion of powder within the mixer were carried out using the PEPT technique at the University of Birmingham. This technique has been described in detailed in Section 3.5. In short, PEPT involves the detection of gamma-rays produced by the result of positron decay of a radioactive tracer particle. The tracer emits positrons that annihilate with electrons to produce back-to-back gamma-rays. Two camera detectors each 50 x 40 cm$^2$ and situated 54 cm apart, were used to detect the tracer using the location of several intersecting gamma-rays. The camera can follow a tracer moving at 1 m/s to within 0.5 mm at a data acquisition rate of over 250 times per second (Bridgwater et al., 2004). This enables detailed studies of powder flow at speeds of industrial significance.

For the tracer particle, a resin bead of 212 µm diameter and density of 1050 kg/m$^3$ was used. It was activated by an ion exchange method with radioactive water produced in a cyclotron (Bridgwater et al., 2004). For a typical PEPT experiment, the powder was loaded with the tracer and allowed to mix for 2 minutes in order to reach steady state flow before the one hour data acquisition commenced.

4.3.2 Torque experiments

Torque experiments were carried out using the same material formulation, mixer bowl and impeller described earlier. However, a different motor was used with a computer controlled variable speed drive ranging from 0-1000 rpm. The detailed description of the torque measurement set-up was described in Section 3.4.
4.4 Results and discussion

Figure 4.4: Tracer axial trajectory at varying impeller speeds for cohesive sand using the rectangular impeller.

For this study, the height of the horizontal static bed before mixing was $H = 0.07$ m. Cohesive sand was used for all experiments. Impeller speed was varied between 100 and 600 rpm (1.67 and 10 Hz) to cover operating conditions of industrial significance. For all experiments, the blades imparted momentum to the powder bed both axially in the form of vertical displacement and horizontally, in the form of radial and angular displacement. The momentum of the rotating impeller blades resulted in a clockwise rotation of the powder bed in the same direction and also, vertical turnover as observed in the mixer.

Figure 4.4 shows the qualitative results of PEPT powder flow experiments following the
axial motion of a tracer particle as a function of time. At low impeller speeds of 100 and 200 rpm, the tracer trajectory exhibited unimodal peaks corresponding to the blade frequency relative to the powder. This was clearly bumping behaviour. When the speed was increased to 250 rpm, there was a change in the signature vertical motion of the tracer particle to full vertical turnover with a double bumping peak overlaid. This is characteristic of the apparent bumping regime. As the impeller speed continued to increase there are further transitions to bumping flow and back to apparent roping. By 600 rpm, fully developed roping flow is achieved. These results qualitatively support the proposed powder flow regime map for bladed impellers.

From Figure 4.4, dominating frequencies for the particle flow could not be determined visually from the complex trajectory of particle. Signal analysis was therefore required to extract this information from the PEPT experiments.

### 4.4.1 Fourier Analysis

Fourier analysis was used on the PEPT data to examine the sinusoidal components of the particle trajectory over time. As the acquisition of the 3D coordinates $P_{xyz}(t)$ was determined by the statistical triangulation method, the PEPT data was inherently non-equidistant. Each data set was re-sampled in order to obtain equidistant samples before FFT analysis. High sampling rates of PEPT data (e.g. 20 point per second at 600 rpm), reduced any potential errors from aliasing. The fast Fourier transform (FFT) was multiplied by its complex conjugate to obtain a power spectral density. The resulting plot of magnitude of power as a function of frequency allowed easy detection of any dominant periodic components of the data.

The power spectral density in the axial plane at varying impeller speeds are shown in Figure 4.5. Two distinct spectral signature groups are observed that quantitatively support the two types of motion, bumping and roping. Above an approximate frequency of 2 Hz, frequency groups are shown that correspond to the first order bumping motion and below this frequency, a broad signature group correlates to the second order roping motion.

The bumping peaks corresponded to the relative bumping frequency of the powder. Generally, the particle frequency increased linearly with increasing impeller speeds. The power spectrum of axial position shows dominant particle frequencies at 2.89 Hz and 5.68 Hz for blade frequencies of 3.33 Hz (100 rpm) and 6.67 Hz (200 rpm) respectively. The difference in the frequency is the circumferential displacement of the tracer as it moved in a clockwise direction around the mixer.

At an impeller speed of 250 rpm (4.17 Hz) apparent roping behaviour is observed. The
Figure 4.5: Power spectral density of particle axial trajectory.

Bimodal peaks of the tracer trajectory are quantified by the two bumping groups at particle frequencies of 3.35 and 7 Hz. For a two-bladed impeller, these bumping groups corresponded to the relative impeller frequency of 4.17 Hz and the relative blade frequency of 8.34 Hz respectively. At even higher speeds, the trend in the experimental data was as expected with the bumping peaks decreasing in significance.

In contrast, the intensity of low frequency signatures of the roping motion increased with increasing impeller speed. The power spectrum shows the broadening of the spread of frequencies with increasing impeller speed except at 5 Hz. This results was as expected as visual observations noted the powder bed reverting back to the bumping regime. Overall, the power spectrum analysis of the tracer’s axial position distinctly shows the mechanisms
4.4 Results and discussion

of the two types of flow regimes and clearly supports the transitional behaviour proposed.

4.4.2 Motion of particles

Streamline diagrams of the trajectories of these behaviours are depicted in Figure 4.6.

![Streamline diagrams of particle trajectories of the three powder flow regimes.](image)

**Figure 4.6:** Streamline diagrams of particle trajectories of the three powder flow regimes.

![Snapshots of powder flow regime using 2 kg of cohesive sand and the rectangular blade in the Hobart mixer (R = 0.15 m) at a) 235 rpm, bumping regime, b) 360 rpm, bumping regime (apparent roping behaviour), c) 370 rpm, bumping regime and d) 560 rpm, roping regime.](image)

**Figure 4.7:** Snapshots of powder flow regime using 2 kg of cohesive sand and the rectangular blade in the Hobart mixer (R = 0.15 m) at a) 235 rpm, bumping regime, b) 360 rpm, bumping regime (apparent roping behaviour), c) 370 rpm, bumping regime and d) 560 rpm, roping regime.

Figure 4.8 shows the trajectory of tracer particles in cohesive sand using the rectangular impeller. These plots were obtained by collapsing the two-dimensional radial-axial plane to captured internal circulations beneath the bed surface. These snapshots at seven different impeller speeds cover all three powder flow regimes.

The motion of particles is driven by the rotating impeller blades. As expected, the trajectories have two distinct motions: a vertical “bumping” motion at the powder element level (A) caused by the blades passage beneath the bed, and a larger “roping” motion at the powder bed level (B) developed by the impeller imparting rotational inertia to the powder bed to overcome gravitational forces. This roping motion is caused when particles
are impacted by the impeller at the bottom of the mixer and then, pushed out towards the wall, up the wall and back around to the centre. Both bumping and roping motions are observed for impeller speeds from 100-500 rpm to varying degrees however, at 600 rpm only roping motion is observed.

For the rectangular impeller experiments, bumping motion is significant because firstly, the blade angle is perpendicular to the direction of the rotating blade, secondly the ratio of immersion height to impeller height is low, and thirdly the blade height relative to particle size is high. The most significant vertical bumping motion is observed at 300 rpm as the bed revert back to the bumping regime phase due to interactions of the bed with the blade sequencing. This interesting reversal back to bumping regime is highly dependent on impeller design, fill level and formulation properties. Most systems will not experience this distinct second phase bumping motion but rather a more subtle version of this transitional bumping behaviour in which random chaotic motion is seen as reported by Litster et al. (2001).

The tracer trajectory moved in one direction for all impeller speeds (200-600 rpm) except for 100 rpm in which it moved in two directions. At low impeller speeds, a diagonal shear plane developed. Here, the particle either falls under the influence of gravity towards the
centre of the mixer or is pushed towards the mixer wall to be recirculated back into the bed again. Stewart et al. (2001a) observed similar bi-directional flow patterns at very low impeller speeds. The development of the diagonal shear plane was explained by low rotational inertia which was unable to overcome the wall friction. While focusing on understanding powder flow behaviour at low speeds is useful to validate simulation models, these results do not necessarily apply to systems operating at industrially significant speeds.

At impeller speeds greater than critical powder Froude number (600 rpm), roping motion dominates. Bumping is no longer significant as seen by the lack of oscillations. Here, significant roping motion is observed allowing a continual renewal of fresh powder at the surface as well as regular interaction with shear forces in the impeller zone. The circulation frequencies (as defined in Section 3.6.2) are shown in Figure 4.9. The measured circumferential and vertical circulation frequencies are a magnitude smaller than the impeller rotation, demonstrating that the powder bed is moving much slower than the impeller. Generally, the circulation increases with increasing impeller speed except for a significant decrease at 300 rpm (5 Hz). These results are consistent with the proposed powder flow regimes in which the powder regime transitionally shifts from apparent roping at 250 rpm, back to bumping at approximately 300 rpm, where only one impeller is interacting with the powder bed. Hence, the reduced rotation result.

For all experiments at the bulk powder level, the motion in the circumferential direction was clockwise which not surprisingly, was also the same direction as the impeller. At a particle level, there are occasions where the particle appears to move in opposite directions of the impeller blade. This occurs as the particle is falling after the blade has moved from beneath it and is more frequent at low speeds because the particle has time to fall to the base of the mixer.

### 4.4.3 Bump height

The bump height $h_a$ is measured from the axial trajectory of the tracer particle. Fig. 4.10 shows an example of the axial position and corresponding measured bump height in the bumping and roping regime. Bump height varies with axial and radial position, and powder flow behaviour. In the bumping regime, bump height increases slightly as the tracer particle moves up the powder bed. This increase is caused by a decrease in the bed density closer to the bed surface, allowing the particles to move more freely in the vertical direction. In contrast, in the roping regime, the bump height decreases with increasing axial height in the more tightly packed bed. As the tracer particle moves further away from the impeller, the less influence on the vertical displacement from each blade pass.
**Figure 4.9**: Average circulation frequency of cohesive sand using rectangular impeller.

**Figure 4.10**: Axial position of tracer particle of cohesive sand and rectangular blade at a) 300 rpm and c) 600 rpm. The red cross represents the intermediate minimums and maximums of the axial trajectory. Values of bump heights at b) 300 rpm and d) 600 rpm.
4.4 Results and discussion

4.4.4 Velocity profile

Velocities were calculated from discrete 3D spatial and temporal data obtained from PEPT experiments using a least squares fitted (LSF) algorithm. The LSF method estimated the tracer velocity along its trajectory by fitting a number of data points $i$ to an $n$th-order polynomial in a least-squares sense, provided that $i > n + 1$. For this analysis, a velocity vector was fitted to four positional data points along the tracer trajectory, with the average of the points used as the vector origin. The selection of $i$ optimised the sampling rate at the highest impeller speed, hence reducing smoothing effects of the LSF method that could potentially masked important bumping motion.

The flight height $h_a$ was measured from powder located at the free surface, unconfined by the weight of the bed. The flight trajectory measured by PEPT is observed to be approximately uniform in terms of rise and fall trajectory, allowing flight time to be measured by Equation 4.7.

Velocities were made dimensionless by dividing by the impeller tip speed:

$$v^* = \frac{v}{v_{\text{tip}}} = \frac{v}{\omega_i \pi D_i / 60}$$

(4.9)

where $v$ is the powder velocity ($v_r$:radial, $v_\theta$:tangential, $v_z$:axial or $v_p$:overall), $N$ is the rotation speed of the impeller (rpm) and $D_i$ is the impeller diameter.

| Experiment   | $N$ (rpm) | $v_{\text{tip}}$ (m/s) | $v_\theta$ ± $\sigma$ (m/s) | $v_p$ ± $\sigma$ (m/s) | $v^*_\theta$ (-) | $v^*_p$ (-) | $|v_r|$ (m/s) | $|v_z|$ (m/s) |
|--------------|-----------|-------------------------|-----------------------------|------------------------|----------------|-------------|-------------|-------------|
| Rectangular  | 100       | 1.10                    | 0.14 ± 0.05                 | 0.19 ± 0.05            | 0.13           | 0.17        | 0.01        | 0.01        |
| Cohesive sand| 200       | 2.20                    | 0.35 ± 0.07                 | 0.42 ± 0.08            | 0.16           | 0.19        | 0.02        | 0.02        |
|              | 250       | 2.75                    | 0.46 ± 0.12                 | 0.53 ± 0.13            | 0.17           | 0.19        | 0.03        | 0.02        |
|              | 300       | 3.30                    | 0.42 ± 0.11                 | 0.55 ± 0.13            | 0.13           | 0.17        | 0.02        | 0.02        |
|              | 400       | 4.40                    | 0.60 ± 0.19                 | 0.68 ± 0.22            | 0.14           | 0.15        | 0.04        | 0.04        |
|              | 500       | 5.50                    | 0.68 ± 0.19                 | 0.73 ± 0.21            | 0.12           | 0.13        | 0.05        | 0.05        |
|              | 600       | 6.60                    | 0.91 ± 0.52                 | 0.97 ± 0.63            | 0.14           | 0.15        | 0.07        | 0.09        |
| Triangular   | 300       | 3.30                    | 0.50 ± 0.14                 | 0.53 ± 0.14            | 0.15           | 0.16        | 0.01        | 0.01        |
| Cohesive sand| 350       | 3.85                    | 0.26 ± 0.09                 | 0.35 ± 0.09            | 0.07           | 0.09        | 0.02        | 0.01        |
|              | 500       | 5.50                    | 0.49 ± 0.15                 | 0.54 ± 0.16            | 0.09           | 0.10        | 0.02        | 0.02        |

Fig. 4.11 shows the influence of impeller tip speed on the average particle velocities (averaged, radial, tangential and axial) for the cohesive sand and rectangular bladed experiments. The averaged and radial velocities increased smoothly with increasing impeller speeds. The average particle velocities measured in the mixer granulator were approximately 10-20% of
Table 4.2: Surface velocities of cohesive sand with rectangular and triangular impeller blades at various rotating speeds.

<table>
<thead>
<tr>
<th>Experiment</th>
<th>$N$ (rpm)</th>
<th>$v_{\text{tip}}$ (m/s)</th>
<th>$v_{\theta, \text{sur}}$ (m/s)</th>
<th>$v_{p, \text{sur}}$ (m/s)</th>
<th>$v_{\theta, \text{sur}}^*$ (m/s)</th>
<th>$v_{p, \text{sur}}^*$ (m/s)</th>
<th>$v_{r, \text{sur}}$ (m/s)</th>
<th>$v_{z, \text{sur}}$ (m/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rectangular</td>
<td>100.00</td>
<td>1.10</td>
<td>0.08 ± 0.03</td>
<td>0.17 ± 0.04</td>
<td>0.07</td>
<td>0.16</td>
<td>0.03</td>
<td>0.02</td>
</tr>
<tr>
<td></td>
<td>100</td>
<td>1.10</td>
<td>0.14±0.05</td>
<td>0.19±0.05</td>
<td>0.13</td>
<td>0.17</td>
<td>0.01</td>
<td>0.01</td>
</tr>
<tr>
<td>Cohesive sand</td>
<td>200</td>
<td>2.20</td>
<td>0.35±0.07</td>
<td>0.42±0.08</td>
<td>0.16</td>
<td>0.19</td>
<td>0.02</td>
<td>0.02</td>
</tr>
<tr>
<td></td>
<td>250</td>
<td>2.75</td>
<td>0.46±0.12</td>
<td>0.53±0.13</td>
<td>0.17</td>
<td>0.19</td>
<td>0.03</td>
<td>0.02</td>
</tr>
<tr>
<td></td>
<td>300</td>
<td>3.30</td>
<td>0.42±0.11</td>
<td>0.55±0.13</td>
<td>0.13</td>
<td>0.17</td>
<td>0.02</td>
<td>0.02</td>
</tr>
<tr>
<td></td>
<td>400</td>
<td>4.40</td>
<td>0.60±0.19</td>
<td>0.68±0.22</td>
<td>0.14</td>
<td>0.15</td>
<td>0.04</td>
<td>0.04</td>
</tr>
<tr>
<td></td>
<td>500</td>
<td>5.50</td>
<td>0.68±0.19</td>
<td>0.73±0.21</td>
<td>0.12</td>
<td>0.13</td>
<td>0.05</td>
<td>0.05</td>
</tr>
<tr>
<td></td>
<td>600</td>
<td>6.60</td>
<td>0.91±0.52</td>
<td>0.97±0.63</td>
<td>0.14</td>
<td>0.15</td>
<td>0.07</td>
<td>0.09</td>
</tr>
<tr>
<td>Triangular</td>
<td>300</td>
<td>3.30</td>
<td>0.50±0.14</td>
<td>0.53±0.14</td>
<td>0.15</td>
<td>0.16</td>
<td>0.01</td>
<td>0.01</td>
</tr>
<tr>
<td></td>
<td>350</td>
<td>3.85</td>
<td>0.26±0.09</td>
<td>0.35±0.09</td>
<td>0.07</td>
<td>0.09</td>
<td>0.02</td>
<td>0.01</td>
</tr>
<tr>
<td></td>
<td>500</td>
<td>5.50</td>
<td>0.49±0.15</td>
<td>0.54±0.16</td>
<td>0.09</td>
<td>0.10</td>
<td>0.02</td>
<td>0.01</td>
</tr>
</tbody>
</table>

the of impeller tip speed. This is consistent with other data in the literature (Wellm, 1997, Litster et al., 2002). The axial and tangential velocities show more complex behaviours with a kink in the plot. As expected, tangential velocity contributes significantly to the averaged velocity of the bed with axial and radial velocities having more of an influence speeds less than critical $Fr_p$.

Operating at low impeller speeds ($< 200$ rpm), the velocities varied linearly with impeller tip speed. This is consistent with PEPT measurements obtained by Stewart et al. (2001a) which also studied flows at low speeds in a vertical-axis mixer. With increasing tip speed, the powder bed moved into apparent roping. In this regime, an increase in the tangential and radial velocities are observed however the axial velocity remained unchanged. This supported the observed dominance of roping behaviour over bumping behaviour.

With further increases in impeller speed, the powder bed reverted from apparent roping back to bumping regime. Correspondingly, a drop in the tangential velocity was observed with a complementary rise in the axial velocity. Increasing the impeller speed even more, the behaviour returns to apparent roping. As the maximum impeller speed of the experiments of 600 rpm still lies within the bumping regime, it can only be extrapolated that the system will move towards roping behaviour. The overall powder velocities generally follow the same linearly increasing trend in the bumping regime. Other researchers have reported that in the roping regime, surface powder velocities are generally less sensitive to impeller tip speed (Litster et al., 2002). The merging of axial and radial velocities at higher speeds supports the observation that fully developed roping motion dominates at higher impeller speeds.

Perhaps even more informative than a single average value for characterising powder
Results and discussion

Figure 4.11: Average overall powder velocities and velocity components (tangential, axial and radial) of cohesive sand using the rectangular impeller at various rotation speeds.

velocity, is the particle velocity distribution. Fig. 4.12 shows the effect of changing impeller speeds on the particle velocity distributions for cohesive sand with the rectangular bladed impeller. The results confirmed that particle velocity can vary widely across the mixer. At the lowest speed, the particle velocity distribution was narrow. Most particle velocities were less than 0.5 m/s. This was reflected in the relatively static bed motion of the bumping regime. As the impeller speed is increased, the distributions are broader. Generally, there was very little difference between the average particle velocity and the tangential velocity distributions. Only at 300 rpm was small changes was observed, in line with the bed reverting back to the bumping regime.

For this mixer geometry, when powder Froude number equates to unity, the critical particle velocity is 1.01 m/s. A comparison of average particle velocity for all experiments is shown in Fig. 4.13. Free-flowing dry sand exhibited higher velocities than cohesive sand for both impeller types. With powder flow regime transitions, the triangular impeller transitioned to the second bumping phase later as demonstrated by a significant drop in particle velocity at 350 rpm instead of 300 rpm for the rectangular impeller. This could be explained by the triangular impeller being less able to impart forces on the particles due to its slanted front edge.

It would be useful to have simple method to calculate particle velocity from known operating parameters like impeller tip speed. Fig. 4.14 shows the dimensionless particle velocity for all bladed experiments. Values across the same mixing system are not constant. They
vary significantly, almost doubling in minimum and maximum values 0.13 to 0.19 respectively for cohesive sand using the rectangular blade. It is clear that predicting averaged powder velocities using impeller tip speed would be erroneous.

4.4.5 Velocity vector maps

While a single trajectory of the tracer gives information about one particle, time-averaged analysis allows the entire powder bed to be characterised. Maps of time-averaged velocity fields for cohesive sand using the rectangular blade at different impeller speeds are shown in Fig. 4.15. The vectors represent the radial and axial components of the velocity and the colours correspond to the magnitude of the average particle velocity.

The maps highlight that particle velocities vary spatially in the mixer. For instance, around the impeller zone particle velocities were high typically 50% greater than the rest
4.4 Results and discussion

Figure 4.13: Comparison of average particle velocity of all bladed experiments. The shaded symbols denote overall particle velocity $v_p$ values and the hollow symbols are the tangential velocity $v_\theta$ values.

of the bed with lower velocities the further the particles are from the impeller. Surface velocities are generally a magnitude less than the impeller tip speed which consistent with observations made by other researchers (Ramaker et al., 1998, Litster et al., 2002).

The bed shape also changes from a relatively flat horizontal surface to a curved vertical surface against the mixer wall with larger rotational inertia from the increased impeller speeds. This has also been observed by other researchers (Hapgood, 2000, Knight et al., 2001, Sato et al., 2005).

To further analyse this spatial non-uniformity, PEPT measurements of particle tangential velocities were averaged across vertical and horizontal cross sections (slices) of the vessel and displayed in Fig. 4.16. The axial profile demonstrate considerable variations across the regimes, with a high degree of uniformity in the bumping regime and a low degree in the roping regime. With different particle velocity in different regions of the bed, it is clear that the highest velocities occur around the impeller zone. These plots are typical of systems with rotating impellers that produce regions of highly localised velocities (Guida et al., 2009). It is interesting to note that the roping regime (600 rpm) is the only system with axially averaged values greater than $v_{crit}$. Here, the centrifugal forces dominate over gravity and suggest that the flow behaviour is dictated by what is occurring in the impeller zone. This supports the use of the powder Froude number in which the characteristic particle velocity is estimated from particle velocities in the impeller zone. With increasing height, the particle velocity is reduced due to wall friction and the compressive stress of the bed. At the top of the bed, a small overhang is observed in the profile as particle velocities are no longer hindered by the weight of the bed and a free to bump under the influence of only gravity.
The radial profiles of the bumping regime generally follow a curved line with low particle velocities near the centre of the impeller, increasing with increased radial position, reaching a maximum in the mid-region of the mixer and then dipping back slightly due to the effects of wall friction. In the roping regime (600 rpm), impacts of avalanching particles with the impeller blade resulted in abnormally high particle velocities in the region close to the mixer centre. The low density of particles in the central region of the mixer allowed these particles to move unhindered which is not the case for particles in the bulk of the bed which typically collide with other particles, transmitting the force to the rest of the bed instead. A significant dip is also observed in the radial profile at 600 rpm which was caused by the tracer becoming trapped in the gap between the blade and the base of the mixer for approximately a minute. Apart from the regions of low particle density, average tangential velocity of the radial annuli did not exceed the critical velocity. This is lower than the axial slices due to the friction of the wall and the orientation of the particle flow in relation to the motion of the blade. Only the roping regime came close to the critical level showing that the influence of the impeller blade is stronger in the region close to the bottom of the mixer and that interaction of the particles with the wall and other particles have a significant impact on the overall flow behaviour.

4.4.6 Occupancy maps

- Discussion about bed volume: The bed volume varied considerably depending on the powder flow behaviour. An initial increase was seen reaching the maximum bed dilation at the height of the bumping behaviour (300 rpm) before decreasing as condense roping flow dominated. This is a somewhat surprising result as one would expect the bed volume to
4.4 Results and discussion

Figure 4.15: Azimuthally averaged normalised tangential velocity $v_\theta/v_{tip}$ (shading) and radial-axial $v_{rz}$ velocity vectors (arrows) maps for cohesive sand and rectangular impeller at a) 100 rpm, b) 200 rpm, c) 250 rpm, d) 300 rpm, e) 400 rpm, f) 500 rpm, and g) 600 rpm.
Figure 4.16: Average powder tangential velocity as a function of a) height and b) radial distance of cohesive sand with the rectangular impeller. —— marks the critical particle tangential velocity for this mixer geometry. The impeller boundary is indicated by ···.
steadily increase as impeller speed increased. An explanation for this result is the dilation of the powder bed from the two different powder flow behaviour observed. Bumping behaviour dominants at low speeds and causes intermittent dilation of the powder bed as the blade passes beneath while at higher speeds, roping behaviour dominants produces a stable dense toroidal flow. Also, this result shows there are some issues with the accuracy of the PEPT tracer, even at lower speeds. The annular cell situated 2nd from the bottom left corner of the mixer, should be void of particles as the impeller blade physically occupies that space. However, it should be noted that the mixer did vibrate considerable during operations, even with the wheels of the trolley that the mixer was situated on was locked which may have resulted in the inaccuracies of the tracer particle.

The two-dimensional occupancy distributions plots of cohesive sand with the rectangular impeller at different impeller speeds are shown in Fig. 4.17.

Analysing the occupancy plots, rough estimates of the bed shape can be inferred from the cross-sectional profile. Changing the impeller speed has a significant effect on the shape of the bed in the mixer. At low impeller speeds, particles are detected in the lower region of mixer, forming a relatively horizontal surface. At higher speeds, centrifugal forces pushed particles out towards the wall resulting in a curvature of the bed surface. The inner radius \( r_i \) of the bed torus increased from no torus \( (r_i = 0) \) in the bumping regime, to a torus shape that is approximately half the radius \( R \) of the mixer \( (r_i = \frac{1}{2}R) \) in the roping regime. A corresponding change in maximum bed height is also observed with increasing impeller speed from 6 cm in the bumping regime to 10 cm in the roping regime.

The impeller speed has a significant effect on the occupancy distribution of particles in the powder bed. High occupancy of particles is observed above the impeller close to the mixer wall and in the core of the powder bed. As one might expect, as the impeller speed increased, centrifugal forces also increased forcing the powder bed out towards the wall and finally, with no where else to go, up the wall. This is demonstrated by observing the movement of the region of highest occupancy (the darkest coloured cells) which moved from the bottom of the mixer to half-way up the powder bed.

Generally, low occupancies are seen in the path of the impeller, near the centre of the mixer and at the bed surface. At 100 rpm, high levels of occupancy are seen at the outer edge of the impeller path suggesting stagnant particles in the rest period between blade passes. With increasing impeller speeds, this dead mixing zone is eliminated and only particles stuck in between the gap of the blade and the mixer are observed. These occupancy plots also demonstrate the interaction of the bed with the impeller blade and support the proposed powder flow regime.

The powder bed density at different impeller speed was estimated from occupancy data
Figure 4.17: Occupancy plots of cohesive sand using rectangular impeller at a) 100 rpm, b) 200 rpm, c) 250 rpm, d) 300 rpm, e) 400 rpm, f) 500 rpm, and g) 600 rpm.
(see Fig. 4.18) as described in section 3.6.6. Initially, the bed density decreased with increasing impeller speed, reaching a local minimum at 300 rpm and then increasing slightly at 500 rpm, before dropping off significantly at 600 rpm. This result corresponded to visual observations of the state of the powder bed as it transitioned through the powder regimes. Generally, the state of the bed in the roping regime is highly fluidised, flowing in a fluid-like motion. In comparison, the apparent roping behaviour is moderately fluidised demonstrating a jolting mixture of bumping and roping flow. In the bumping regime, the particles either slid over the blade (100 rpm) or was hit with some force (200 and 300 rpm), resulting bumping flow of varying degree of fluidisation.

![Image of bed density graph]

**Figure 4.18:** Bed density of the powder bed at various impeller speeds.

### 4.4.7 Mixing

Mixing is measured using relative standard deviation (RSD) described in section 3.6.10. In general, the convective-shear mixing performance circumferentially around the mixer increased with increasing impeller speeds except at 100 rpm which actually showed the best mixing (Fig. 4.19a). This can be explained by the combination of convective-shear and diffusive mixing which both occur at low speeds. Above 100 rpm, the powder bed moved in a solid rotating body reducing the effectiveness of diffusive mixing. In the vertical direction, effect of solid body rotation are pronounced. At higher speeds (500 and 600 rpm), the oscillations in RSD are counter-intuitive but can be explained by the recirculation within the roping behaviour.
Figure 4.19: Effect of impeller speed on degree of mixing of cohesive sand using a rectangular blade in the a) horizontal and b) vertical azimuthal plane.
4.4.8 Shear rates

Shear rates are obtained using the data analysis technique described previously in section 3.6.9. Fig. 4.20 shows the effect of impeller speed on the shear rate for cohesive sand with the rectangular impeller. Shear rates vary spatially within the mixer granulator with the highest shear rates located around the impeller. Negligibility shear is observed in the path of the impeller where particles are hit by the flat, front face of the impeller and only experience small differences in velocity between the adjacent cells. Moving vertically from the impeller, regions of low shear rate exists as the influence of the moving blade decreases. At the powder bed surface, shear rate increases again as particles are free to flow unhindered. Radially, shear rates also varies from a relatively horizontal layer at low speeds to a curved profile at high speeds. This shift is caused by two factors, the increasing influence of rotational inertia over gravity and the increased frictional effect of particle-wall interaction.

4.5 Validation of regime map

The value of $Fr_p$ was calculated from the characteristic powder velocity, taken to be the average powder tangential velocity. Fig. 4.21 and Table 4.3 shows the flow regime classification. There is remarkably good agreement between the visually observed flow behaviour and the powder flow classification.

<table>
<thead>
<tr>
<th>Exp</th>
<th>$N$ (rpm)</th>
<th>$h_a$ (mm)</th>
<th>$t_a$ (s)</th>
<th>$\omega_b$ (/s)</th>
<th>$t_b$ (s)</th>
<th>$\beta$ (-)</th>
<th>$R_c$ (m)</th>
<th>$v_\theta$ (m/s)</th>
<th>$Fr_p$ (-)</th>
<th>Regime</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>100</td>
<td>23.90 ± 0.140</td>
<td>2.89</td>
<td>0.346</td>
<td>0.40</td>
<td>0.0525</td>
<td>0.14</td>
<td>0.04</td>
<td>Bumping</td>
<td></td>
</tr>
<tr>
<td>Coh</td>
<td>200</td>
<td>25.10 ± 0.143</td>
<td>5.68</td>
<td>0.176</td>
<td>0.81</td>
<td>0.0643</td>
<td>0.35</td>
<td>0.19</td>
<td>Bumping</td>
<td></td>
</tr>
<tr>
<td></td>
<td>250</td>
<td>26.19 ± 0.146</td>
<td>7.00</td>
<td>0.142</td>
<td>1.02</td>
<td>0.0643</td>
<td>0.46</td>
<td>0.34</td>
<td>Bumping (AR)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>300</td>
<td>41.66 ± 0.184</td>
<td>4.27</td>
<td>0.234</td>
<td>0.81</td>
<td>0.0643</td>
<td>0.42</td>
<td>0.27</td>
<td>Bumping</td>
<td></td>
</tr>
<tr>
<td></td>
<td>400</td>
<td>41.75 ± 0.185</td>
<td>5.44</td>
<td>0.184</td>
<td>1.00</td>
<td>0.0643</td>
<td>0.60</td>
<td>0.58</td>
<td>Bumping (AR)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>500</td>
<td>28.60 ± 0.153</td>
<td>6.82</td>
<td>0.147</td>
<td>1.04</td>
<td>0.0643</td>
<td>0.68</td>
<td>0.72</td>
<td>Bumping (AR)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>600</td>
<td>31.95 ± 0.161</td>
<td>8.17</td>
<td>0.122</td>
<td>1.32</td>
<td>0.0729</td>
<td>0.91</td>
<td>1.15</td>
<td>Roping</td>
<td></td>
</tr>
<tr>
<td>Tri-</td>
<td>300</td>
<td>17.30 ± 0.119</td>
<td>8.37</td>
<td>0.119</td>
<td>1.00</td>
<td>0.0525</td>
<td>0.50</td>
<td>0.49</td>
<td>Bumping (AR)</td>
<td></td>
</tr>
<tr>
<td>Coh</td>
<td>350</td>
<td>23.00 ± 0.137</td>
<td>5.59</td>
<td>0.178</td>
<td>0.77</td>
<td>0.0525</td>
<td>0.26</td>
<td>0.13</td>
<td>Bumping</td>
<td></td>
</tr>
<tr>
<td></td>
<td>500</td>
<td>25.00 ± 0.143</td>
<td>7.73</td>
<td>0.129</td>
<td>1.11</td>
<td>0.0525</td>
<td>0.49</td>
<td>0.47</td>
<td>Bumping (AR)</td>
<td></td>
</tr>
</tbody>
</table>

"Bumping (AR)" refers to powder flow in the Bumping regime exhibiting apparent roping behaviour (see Section 4.4.2).
Figure 4.20: Contour maps of shear rates of cohesive sand and rectangular impeller at a) 100 rpm, b) 200 rpm, c) 250 rpm, d) 300 rpm, e) 400 rpm, f) 500 rpm, and g) 600 rpm. Note the different shear rate (s$^{-1}$) colour scales. The location of the impeller was shown by - -.
4.5 Validation of regime map

4.5.1 Torque measurements of bumping regime

The effect of the bumping regime on impeller torque for various cohesion and blade-rake angle combination are shown in Fig. 4.22. Interesting oscillations in torque measurements are observed with increasing impeller speeds for all experiments except dry sand with triangular blade. Other researchers have reported this phenomena in their result without effectively explaining the cause (Wellm, 1997, Knight et al., 2001). Here, an attempt is made to offer an explanation for these observations based on the proposed flow regimes.

With increasing impeller speed, the bed is steadily increasing in torque as momentum is transferred to the bed through the impact of the leading edge of both impeller blades. At a critical transition point the torque decreases, 250 rpm for the rectangular and 320 rpm for the triangular blade. This gradual decrease is caused by the bed interacting with mostly the top surface of both blade. At the intermediate torque minima, 300 rpm for the rectangular and 350 rpm for the triangular blade, the powder bed is now only interacting with one impeller blade as the second blade rotates within the void created by the first blade position. The torque measurement supports this explanation as the intermediate minima is half the torque (relative to the initial starting point) of the intermediate maxima, i.e. only half the force is...
FIGURE 4.22: Effect of cohesion and blade-rake angle on impeller torque at various impeller rotating speeds for fill load of H/D = 0.29. Bed resonance is observed for all experiments except for dry sand with triangular blade.

...transferred as only half the impeller (one blade) is providing the work.

Interestingly, no significant drop in torque caused by bed resonance was observed for the dry sand and triangular blade experiment. Considering the competing sinusoidal wave theory proposed earlier in Section 4.2, the combination of low bump amplitude from the low blade-rake angle (short $t_a$) and the lack of cohesivity in the powder bed (long $t_b$) reduced the likelihood that a bed collapse would occur before the roping regime becomes dominant. This supports the argument that only under certain conditions does this complex bumping regime resonance have significant impact on powder flow behaviour. Work by Knight et al. (2001) using the same mixing bowl as this current study (as well as a variety of others) supports this observation however more work is required to elucidate the effect of fill level, number of impeller blade, radius of mixing bowl, cohesivity of materials and blade-rake angle has on bumping regime resonance.

From these results, it is evident that torque measurements can be used to validate bed resonance behaviour as predicted from the powder flow regime map. However, it cannot be...
used to directly detect the critical transition speed from *bumping to roping* ($Fr_{p,crit}$) which is of interest for effective scale up of stable roping motion.

**4.5.2 Effective shear thickness on roping regime**

![Figure 4.23: Effective shear thickness for a) gravity dominated bumping regime and b) inertia dominated roping regime. Figure from Mort (2009).](image)

One method to validate the transition from bumping to roping regime is to measure the effective shear layer $\delta_{shear}$. The height (thickness) of the effective sheared layer plays an important role in the transmission of stress from the impeller to the powder bed (see Fig. 4.23 from Mort (2009)). Mort (2009) proposed that a difference exist between the effective shear layer in a centripetal mixer-granulator of different flow regimes. At flow where centripetal acceleration is less than gravity (low $Fr_I$), the shear stress from the impeller decays over a short distance $\delta$ relative to the mixer scale. At flow where centripetal acceleration is greater than gravity (high $Fr_I$), the shear stress is transmitted through the entire powder bed resulting in torodial flow.

Table 4.4 shows the height of the shear layer estimated from Fig. 4.20, at the maximum height at which the shear rate averaged across the mixer axis in horizontal slices reached the minimum shear rate. In the bumping regime, the thickness of the layer varied with impeller speed. In the roping regime, the effective shear layer consisted of the entire powder bed. There is excellent agreement between predicted transition between the flow regimes and the shear layer thickness.

**4.6 Relationship for measurable regime map parameter**

The powder flow regime map while useful in understanding flow phenomena, the practicality of the map for an operator of a vHSM is limited as it depends on the actual powder parameters ($v_\theta$) and not impeller parameters ($v_{tip}$). To overcome this limitation, the dimensionless numbers of the powder flow regime map can be related to easily measured parameters. The
average tangential velocity of the powder bed can be approximated from the impeller tip speed by:

\[ v^* = \frac{v^*}{v_{tip}} \]  

(4.10)

where \( v^* \) is the dimensionless tangential velocity corresponding to \( \approx 0.14 \) and \( 0.09 \) for the rectangular and triangular blade of wetted material respectively (see Table 4.1). Dry materials increases \( v^* \) by approximately 10%. As experiments were only carried out to 600 rpm (at the boundary of the roping behaviour), it cannot be confirmed that powder velocities become insensitive to impeller speeds at high velocity as observed in previous studies (Litster et al., 2002). Another option would be to relate tangential velocity to empirically derived powder surface velocity. The surface velocity measurements of the powder bed using PEPT or the less expensive particle image velocimetry (PIV) method can be used. For this particular system using the rectangular blade, in the bumping regime the tangential velocity is approximately equal to the surface velocity. In the roping regime, the tangential velocity is: \( v^* \approx 1.15v_{sur} \).

In terms of the bed resonance number, simple experiments or simulations can be carried out to ascertain the theoretical bump period \( t_a \) and the period between two blade passes relative to the powder \( t_b \). A single (or double) blade moving through a box of powders with a glass window could be conducted similar to experiments carried out by numerous researchers (Bagster and Bridgwater, 1967, Bagster, 1969, Bridgwater et al., 1969b, Rad, 2012). Alternative, DEM simulations can be conducted of the moving blade through a rectangular box of granular material similar to Sir (2011).

The radial distance of the bed centre from the mixer central axis can be estimated from

### Table 4.4: Shear layer thickness.

<table>
<thead>
<tr>
<th>Experiment</th>
<th>( N ) (rpm)</th>
<th>( \delta_{shear} ) (m)</th>
<th>( \delta_{shear}/R )</th>
<th>Flow</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rectangular blade</td>
<td>100</td>
<td>0.015</td>
<td>0.14</td>
<td>Bumping</td>
</tr>
<tr>
<td>Cohesive sand</td>
<td>200</td>
<td>0.015</td>
<td>0.14</td>
<td>Bumping</td>
</tr>
<tr>
<td></td>
<td>250</td>
<td>0.040</td>
<td>0.38</td>
<td>Bumping (AR)</td>
</tr>
<tr>
<td></td>
<td>300</td>
<td>0.030</td>
<td>0.29</td>
<td>Bumping</td>
</tr>
<tr>
<td></td>
<td>400</td>
<td>0.060</td>
<td>0.57</td>
<td>Bumping (AR)</td>
</tr>
<tr>
<td></td>
<td>500</td>
<td>0.070</td>
<td>0.67</td>
<td>Bumping (AR)</td>
</tr>
<tr>
<td></td>
<td>600</td>
<td>0.100</td>
<td>0.95</td>
<td>Roping</td>
</tr>
<tr>
<td>Triangular blade</td>
<td>300</td>
<td>0.020</td>
<td>0.19</td>
<td>Bumping (AR)</td>
</tr>
<tr>
<td>Cohesive sand</td>
<td>350</td>
<td>0.030</td>
<td>0.29</td>
<td>Bumping</td>
</tr>
<tr>
<td></td>
<td>500</td>
<td>0.035</td>
<td>0.33</td>
<td>Bumping (AR)</td>
</tr>
</tbody>
</table>
the powder bed shape. Real-time cameras/laser devices or visual inspection can assess the bed shape to estimate inner radius of the toroid. The minimum value of $r_c = R/2$, half the the mixer radius occurring at low speeds in the bumping behaviour when the powder bed surface remains horizontal.

4.7 Potential application of regime map to granulation

For bladed vHSM granulators, the powder flow regime map has great potential for developing an understanding of granulation processes, design and scale up. This regime map was the first to map the complex flow behaviour observed in a bladed vertical high shear mixer from low to industrial significant impeller speeds.

At $Fr_p$ values below unity, increasing $\beta$ caused a shift towards the apparent roping behaviour and increased vertical bed turnover rates. Low $\beta$ led to low turnover rates as momentum from the blade is transferred predominately to vertical displacement of the bed instead of rotation momentum. In contrast, at $Fr_p$ values above unity, rotational inertia forces dominant and the influence of $\beta$ on the flow is insignificant.

As for designing granulation processes, ideally one would operate in the roping regime which is characterised by high vertical bed turnover resulting in effective mixing. Operating in the bumping regime may lead to poor mixing, pooling of binder on the bed surface and broad particle size distributions.

Visual observation, or measurement of powder surface velocity alone may lead to the classification of a flow as roping when apparent roping is occurring. This is very dangerous from an operational standpoint as a small decrease OR increase in impeller speed (other parameter) could lead to a return to the highly undesirable bumping regime.

4.8 Conclusion

This study has shown that complex powder flow behaviour can be characterised from blade-powder interactions and the powder Froude number. The flow behaviour of cohesive sand and rectangular blade was observed to have not one transition but four transitions as the impeller speed was increased from 100 to 600 rpm (1.67 to 10 Hz). The system transitioned from bumping to apparent roping, back to bumping, then to apparent roping again and finally, to the roping regime. The apparent roping behaviour was introduced to define observed flow behaviour for this particular system which has not been fully characterised in the literature before. Cohesive sand and triangular blade was also observed have multiple transitional
A powder flow regime map was proposed and verified using PEPT and torque experiments. Further research is required to elucidate the boundaries for a variety of formulation and mixer designs. A fundamental understanding of the physical interaction between the material properties and impeller blades at high speeds is also lacking, which would be useful for predictive models.

The interesting flow phenomena highlighted by the powder flow regime map has implications for industries, especially for granulation processes where the identification of the roping motion is essential for controlled agglomeration. The map simplified the complexity of powder flow behaviour into two dimensionless groups which enabled a rational approach to devising appropriate operational parameters for a given formulation and mixer system.

**Nomenclature**

\[
\begin{align*}
D & \quad \text{mixer diameter (m)} \\
 d_p & \quad \text{particle diameter (m)} \\
 Fr_c & \quad \text{critical powder Froude number (–)} \\
 Fr_I & \quad \text{impeller Froude number (–)} \\
 Fr_p & \quad \text{powder Froude number (–)} \\
 g & \quad \text{gravitational constant (m/s}^2\text{)} \\
 I & \quad \text{dimensionless shear rate (–)} \\
 N & \quad \text{impeller rotational speed (rpm)} \\
 P & \quad \text{characteristic normal stress (kPa)} \\
 R & \quad \text{mixer radius (m)} \\
 R_c & \quad \text{radial position of the powder bed centroid (m)} \\
 R_0 & \quad \text{inner radius of the toroid (m)} \\
 h_a & \quad \text{powder vertical displacement height (m)} \\
 h_{bed} & \quad \text{static bed height (m)} \\
 t_a & \quad \text{powder bump period (s)} \\
 t_b & \quad \text{blade interval period relative to the powder (s)} \\
 t_{rise} & \quad \text{powder bump rise period (s)} \\
 v_c & \quad \text{critical powder velocity (m/s)} \\
 v_{tip} & \quad \text{impeller tip speed (m/s)} \\
 v_\theta & \quad \text{average powder tangential velocity (m/s)}
\end{align*}
\]
Greek symbols

\( \beta \) dimensionless bed resonance number (\(-\))

\( \beta_c \) dimensionless critical bed resonance number (\(-\))

\( \rho \) powder bulk density \( g/mL \)

\( \omega_b \) blade frequency relative to the powder (\( rps or Hz \))

\( \omega_I \) impeller angular rotational speed (\( rad.s^{-1} or Hz \))

\( \omega_p \) powder angular rotational speed (\( rad.s^{-1} or Hz \))

\( \dot{\gamma} \) shear rate (\( s^{-1} \))
The effects of cohesion and blade-rake angle on powder flow and mixing

Abstract

Powder flow and mixing patterns beneath the powder surface at industrially relevant speeds are still relatively unknown. Innovations in imaging technology have allowed real-time, non-invasive studies of particulate flow in opaque systems using Positron Emission Particle Tracking (PEPT). The aim of Chapter 5 is to investigate the effect of cohesion and blade-rake angle on the powder flow and mixing behaviour in a vertical high shear mixer (vHSM) at high impeller speeds. Experiments with dry and cohesive (oil-coated) sand were carried out in a 0.21 m diameter cylindrical mixer using a two-bladed rectangular (90°) impeller and a two-bladed triangular (10°) impeller at constant impeller rotating speed, (500 rpm). The results provided detailed information of localised powder trajectories, velocities, occupancies (bed densities), shear rates, solid body rotation frequencies and mixing patterns useful for understanding and optimisation of mixer granulators performances.
5.1 Introduction

Granulation of powders is commonly processed in vessels using either passive or mechanical means to agitate the mixture. One such device is the vHSM which utilises mechanically rotating impellers to move granular solids from regions of low to high shear. A high level of regulatory demand in the pharmaceutical industry has lead to significant research in understanding and controlling granulation processes. Key to the quality and uniformity of products is the predictive characterisation of powder flow behaviour.

Powder flow patterns beneath the powder surface at industrial relevant operating conditions where rotation inertia exceeds gravity ($Fr_p > 1$ refer to Eqn. 4.4) are still relatively unknown. Most of the work published on powder flow behaviour in vertical high speed mixers deals with results obtained in the bumping regime at low powder Froude numbers (Stewart et al., 2001b, Conway et al., 2005, Lekhal et al., 2006). In this regime, the flow is dominated by the “bumping” passage of the impeller blades beneath the bed. Much less has been published about stable roping regime flow at high Froude numbers. Over the past two decades, PEPT has become a well-established non-invasive experimental technique that enable the study of three-dimensional powder flow patterns beneath the surface. While the technique is expensive and has resolution limitations for tracer speeds beyond 1 m/s, it still provides the best approach to study dynamic granular systems.

5.1.1 Effect of cohesion on powder flow and mixing

The study of the effect of cohesion on powder flow is important in granulation, as the addition of moisture to the dry powder bed during wetting and nucleation can have a dramatic effect on flow and mixing behaviour. Muguruma et al. (2000) observed experimentally using a high speed camera that the powder velocities of powders containing up to 12% water decreases by 10% compared to the dry powder velocity. This trend was also supported by DEM simulations (Muguruma et al., 2000). Radl et al. (2010) also studied the effect of cohesion in a four bladed mixer using DEM using glass spheres and a liquid bridge model. At low impeller speeds where the maximum tip speed tested was 0.3 m/s, results showed that the roughness (amplitude) of the heap formation as the blade moved beneath the bed was higher for the wet mass. The analysis of mixing quality as measured by velocity fluctuations shows that the spatial distribution of mixing intensity is influenced by the moisture content. Higher mixing rates was observed for wet particles both locally and globally compared to dry granular matter.
5.1.2 Effect of blade-rake angle on powder flow and mixing

Numerous studies have been conducted to investigate the effect of blade-rake angle on powder flow using a simple experimental system of a single blade moving through a dry bed of particles in a rectangular box Bagster and Bridgwater (1967), Bagster (1969), Bridgwater et al. (1969a), Bagster and Bridgwater (1970). Bagster and Bridgwater (1970) showed that the blade-rake angle can have a significant effect both on the force on the blade and the volume of material conveyed by a blade. For example, a vertical blade has a high energy input for transporting powder material. Particle motion over single blade have also been studied using PEPT (Stewart et al., 2001a) and DEM (Stewart et al., 2001b) showing good qualitatively agreement. Sir (2011) DEM studies of blade-rake angle on powder mixing in a rectangular box showed that the mixing performance was highest for blade-rake angles that offered a maximal surface area or maximal resistance to the flow of particles, which occurred for blade-rake angles from 70° to 90°. Chandratilleke et al. (2009) studied the effects of blade-rake angle on particle mixing in a cylindrical mixer using DEM and found that the maximum mixing rate was achieved with the 90° impeller at tip speeds of 0.26 m/s. Chandratilleke et al. (2010) carried out further simulations at higher tip speeds (1.3 m/s) however, it still only reached 1/10 the blade tip speed required for a so-called high speed mixer (Knight et al., 2001).

This chapter reports on the results of PEPT experiments investigating cohesion and impeller blade-rake angle effects on flow behaviour and mixing patterns in a vHSM at constant impeller rotating speed of 500 rpm (tip speed = 5.4 m/s). The study of dry and cohesive (oil coated) sand with 90° rectangular and 10° triangular impeller blades describes qualitatively the three-dimensional motion of powder material to provide experimental data for validation of flow simulations. Spatial averages of axial, radial and tangential velocities, shear rates and solid body rotation have been used to characterise the flow. A comparison of mixing is conducted using the relative standard deviation of particle concentration. The findings presented are relevant to the operation of vHSM as they provide insight on parameters that may affect bulk powder flow, mixing, segregation, surface wetting, particle breakage and agglomeration. Finally, recommendations are provided for optimising granulation processes in relation to powder flow and mixing.

5.2 Experimental procedure

The experiments were carried out in a 0.21 m diameter custom built vHSM at the University of Birmingham. Two types of impeller designed were investigated, a two-bladed rectangular
(90°) impeller and a two-bladed triangular (10°) impeller. Detailed description of the equipment set up is given in Section 3.3. Two types of powders were investigated, dry sand and cohesive sand. The cohesive sand mixture was prepared by placing dry sand and silicone oil into a plastic bag in a ratio of 2 kg of dry sand to 50 mL of silicone oil (Sigma Aldrich DC 200, $\mu = 0.1$ Pa.s) and then hand kneaded for 5 minutes to obtain a uniform and reproducible starting mass of 2 kg of dry sand and silicone oil. A summary of the material properties is found in Table 3.1 and 3.6. The PEPT technique discussed in Section 3.5 was used to analyse powder flow behaviour and for brevity was not repeated here. The horizontal surface of the loaded powder gave a fill level of $H/D=0.29$ with the impeller blade completely immersed and approximately 70 mm of mixture above the mixer base. The impeller rotating speed was held constant at 500 rpm and a summary of the four experiments is given in Table 5.1.

<table>
<thead>
<tr>
<th>Impeller design</th>
<th>Material</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Dry sand</td>
</tr>
<tr>
<td>90° Rectangular</td>
<td>▲</td>
</tr>
<tr>
<td>10° Triangular</td>
<td>▲</td>
</tr>
</tbody>
</table>

### 5.3 Results and discussion

The PEPT raw data was process and analysed using the newly developed Matlab code described in Section 3.6. All experiments fall within the bumping regime ($Fr_p < 1$ refer to Eqn. 4.4) with $Fr_p$ between 0.23-0.55 (the lowest $v_\theta = 0.49$ for the cohesive sand and triangular blade, and the highest $v_\theta = 0.76$ for dry sand and rectangular blade). In this regime, the bumping behaviour controlled by gravity dominates however, rotation inertia forces leading to roping behaviour has an increasing influence with higher powder Froude number.

#### 5.3.1 Powder flow

Figure 5.1 shows 10 seconds snapshots of the axial motion of the tracer particle as a function of time. All experiments exhibit two distinct sinusoidal frequency indicative of bumping regime flow: a high frequency *bumping* signature corresponding to blade passes beneath the powder bed, and a low frequency *roping* signature, a result of the toroidal motion of the powder bed. Despite identical impeller rotating speed (impeller tip speed), the trajectory of the tracer is distinctly different for each material - blade design combination. Both
The effects of cohesion and blade-rake angle on powder flow and mixing

Figure 5.1: Tracer axial trajectory for a) dry sand and rectangular impeller b) cohesive sand and rectangular blade c) dry sand and triangular blade and d) cohesive sand and triangular blade at 500 rpm ($v_{tip} = 5.4 \text{ m/s}$).

The rectangular impeller experiments show more regular roping behaviour, evidence that the higher the blade-rake angle, the more efficient the blade is at transferring rotational inertia forces to the powder bed which is as expected. A physical explanation is that the 90° blade presents a larger surface area of maximal resistance than the 10°, transferring more momentum to each particle in contact with the blade. This observation has also been reported by other researchers (Kuo et al., 2003, Chandratilleke et al., 2009, Sir, 2011).

The increase in cohesion has pronounced effect on the flow behaviour, reducing the influence of inertia forces and hence, increasing the bumping behaviour. A possible explanation for this observation is that liquid bridges formed by adding moisture, acts as a lubricant. Hapgood (2000) describes that the wetting of the particles lubricates the contact points and reduces the interparticle friction.
5.3.2 Velocities

The average tangential velocity profiles at the centroid and wall are shown for comparison in Fig. 5.2. Clear spatial differences are observed in the mixer with the velocity profiles at the wall showing significant wall effects across the different runs. The typically “pot-belly” profile at the centroid decreases in significance at the wall. At the centroid, increasing the cohesion resulted in a decrease in tangential velocity for both blade-rake angle however, at the wall, the reverse is observed. Beyond the impeller region, cohesive sand is observed to have higher tangential velocity than dry sand at the wall. One possible explanation for the higher velocity values of wet sand is that the liquid binder acts as a lubricant between the powder bed and the mixer wall, similar to the lubrication between particle-particle interaction described above.

Interestingly, the velocity results are contradictory to frictional properties measured by the Schulze ring shear tester in which the wall adhesion for cohesive sand was greater than dry sand (Table 3.6). Experiments by Darelius et al. (2007a) with microcrystalline cellulose and water also supported this trend in which the material properties measured by a Jenike shear tester actually increased with the addition of a binder. Darelius et al. (2007a) offered an explanation for this observation in which the liquid binder in the wet powder acted like a “glue” under static conditions (shear testing) and as a “lubricant” under dynamic motion. This highlights the importance of cohesion and the need for understanding and characterisation of powder materials to enable predictive scale-up based on first principles.

Fig. 5.3 shows the azimuthal averaged, normalised powder tangential velocity overlaying the radial-axial velocity vectors maps and the effect of varying the impeller type and cohesion of the powder at the same rotational speed. Dry powders are observed to have more erratic motion at the bed surface with particles moving freely, potentially colliding with other particles mid-air. It was observed that more dust and fine particles are created for dry sand experiments. In contrast, cohesive sand tend to form clumps and stick within the bulk of the powder bed. Some cells near the mixer centre exhibited negative tangential velocities which physically means that the particle moves in opposite direction of the blade. This can occur if the particles cascading down the face of the powder bed impacts on the blade at an angle that pushes it in the opposite direction to the movement of the blade.

The frequency distribution of normalised velocity components for the four runs are shown in Fig. 5.4 and the resulting averaged velocity values given in Table 5.2. Increasing the blade-rake angle increases the average powder velocity of the bed. This finding is in agreement with other workers (Kuo et al., 2003, Chandratilleke et al., 2009, Sir, 2011). With the addition of moisture (increase cohesion) the average powder velocity in the bed decreases. This trend
Figure 5.2: Velocity profiles in the tangential direction at the a) centroid and b) wall. The location of the impeller is shown by - - .
5.3 Results and discussion

Figure 5.3: Azimuthally averaged normalised tangential velocity $v_\theta/v_{\text{tip}}$ (shading) and radial-axial $v_r$ velocity vectors (arrows) maps at 500 rpm ($v_{\text{tip}} = 5.4 \text{ m/s}$) for a) dry sand and rectangular impeller b) cohesive sand and rectangular blade c) dry sand and triangular blade and d) cohesive sand and triangular blade.

follows similar observations by other researchers (Muguruma et al., 2000).

| Blade | Sand  | $vol$ $(\text{cm}^3)$ | $p_{\text{bed}}$ $(\text{kg/m}^3)$ | $v_\theta \pm \sigma$ (m/s) | $v_p \pm \sigma$ (m/s) | $v_\theta^*$ (-) | $v_p^*$ (-) | $|v_r|$ (m/s) | $|v_z|$ (m/s) |
|-------|-------|----------------------|-----------------------------|------------------------|------------------------|----------------|----------------|----------------|----------------|
| Rect  | Dry   | 3342                 | 598                         | 0.76 ± 0.30            | 0.82 ± 0.32            | 0.14           | 0.15           | 0.07           | 0.08           |
| Rect  | Coh   | 3143                 | 636                         | 0.68 ± 0.19            | 0.73 ± 0.21            | 0.12           | 0.13           | 0.05           | 0.05           |
| Tri   | Dry   | 2901                 | 689                         | 0.55 ± 0.22            | 0.60 ± 0.25            | 0.10           | 0.11           | 0.04           | 0.03           |
| Tri   | Coh   | 2857                 | 700                         | 0.49 ± 0.15            | 0.54 ± 0.16            | 0.09           | 0.10           | 0.02           | 0.01           |

5.3.3 Bed occupancy

Fig. 5.5 shows the occupancy plots of the four experiments. Qualitatively, the angle of the cascading surface of the powder bed is almost vertical for the dry sand, while the cohesive sand exhibit less steep angles. The bed is more dilated with cohesive sand which can be
The effects of cohesion and blade-rake angle on powder flow and mixing

<table>
<thead>
<tr>
<th>Material</th>
<th>Blade-Rake Angle</th>
<th>Impeller Shape</th>
<th>Axial</th>
<th>Radial</th>
<th>Tangential</th>
<th>Overall</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dry sand</td>
<td>90°</td>
<td>Rectangular</td>
<td><img src="image1" alt="Axial" /></td>
<td><img src="image2" alt="Radial" /></td>
<td><img src="image3" alt="Tangential" /></td>
<td><img src="image4" alt="Overall" /></td>
</tr>
<tr>
<td>Cohesive sand</td>
<td>90°</td>
<td>Rectangular</td>
<td><img src="image5" alt="Axial" /></td>
<td><img src="image6" alt="Radial" /></td>
<td><img src="image7" alt="Tangential" /></td>
<td><img src="image8" alt="Overall" /></td>
</tr>
<tr>
<td>Dry sand</td>
<td>10°</td>
<td>Triangular</td>
<td><img src="image9" alt="Axial" /></td>
<td><img src="image10" alt="Radial" /></td>
<td><img src="image11" alt="Tangential" /></td>
<td><img src="image12" alt="Overall" /></td>
</tr>
<tr>
<td>Cohesive sand</td>
<td>10°</td>
<td>Triangular</td>
<td><img src="image13" alt="Axial" /></td>
<td><img src="image14" alt="Radial" /></td>
<td><img src="image15" alt="Tangential" /></td>
<td><img src="image16" alt="Overall" /></td>
</tr>
</tbody>
</table>

Figure 5.4: Normalised frequency distribution for Lagrangian velocity components at 500 rpm for various materials and blade-rake angles. Velocities have been normalised by dividing by impeller tip speed. Frequency bin width are 0.02 m.s\(^{-1}\).

explained by the greater influence bumping behaviour has on the overall flow behaviour.

5.3.4 Shear rates

Shear rates are obtained using the data analysis technique described previously in section 3.6.9. Figure 5.6 and 5.7 shows the effect of cohesion and blade-rake angle on the shear rates of the tangential velocity component. Shear rates vary spatially within the mixer granulator with the highest shear rates located around the impeller and at the free surface as expected. Increasing blade-rake angle did lead to higher overall shear rates in the bed. Lower shear is observed in the direct path of the rectangular impeller where particles are hit by impact on the flat, front face of the impeller. Interestingly though, the triangular blade exhibited higher localised shear rates in the impeller region however, these shear forces did not transmit
5.3 Results and discussion

5.3.5 Mixing

Statistical analysis is used to determine the homogeneity of the mixture which can be represented by the relative standard deviation (RSD) as described previously in Section 3.6.10.

---

**Figure 5.5:** Occupancy plots at 500 rpm \( (v_{tip} = 5.4 \text{ m/s}) \) for a) dry sand and rectangular impeller b) cohesive sand and rectangular blade c) dry sand and triangular blade and d) cohesive sand and triangular blade.

Evenly to the rest of the bed in comparison to the higher blade-rake angle of the rectangular blade. This leads to the conclusion that for better flow conditions, the impeller blades need to impart greater impact than shear forces. Perhaps high shear mixers should also be known as high shear-impact mixer instead.

The shear rates decrease for increasing cohesivity. The statically measured material properties however failed to detect any differences in the internal angle of friction and anti-correlatedly, cohesion was significantly higher for cohesive sand than dry sand (Table 3.1). A similar explanation to the wall friction described above is that particle-particle friction of cohesive sand was lowered as the liquid provided a lubricant between the frictional contact points under dynamic conditions.
Convective-shear mixing performance around the mixer axis for the four experiments varying material cohesion and blade-rake angle at constant impeller speed showed similar mixing results (Fig. 5.8a). Circumferentially, the RSD mixing value initially decreased till a value of 0.5 and then showed only small improvements in the mixing as it moved towards a mixing equilibrium. The rectangular blades showed only slightly better mixing results than the triangular blade while there were no discernible differences between the dry and cohesive sand in the horizontal direction.

For vertical mixing all experiments except for the triangular blade and cohesive sand showed similar results in which mixing initially becomes slightly more ordered as the impeller sweeps all the particles in the impeller zone together towards the wall, and then improves as the dispersive action of the blade comes into play, reaching a steady-state mixedness at RSD = 1.5. For the triangular blade and cohesive sand, the dominant bumping flow behaviour resulted in poorer mixing.
5.4 Implications for granulation

An understanding of powder flow behaviour can provide practical strategies for operators for the optimal operating conditions in a vHSM. Consider the granulation process over time. Initially in the wetting and nucleation stage, high powder turnover at the bed surface is important to enable operations in the drop controlled nucleation regime. Ideally in this regime, the powder flux through the spray zone should be sufficiently high so that one drop produces one nucleus granule resulting in a narrow size distribution of granules (Litster et al., 2001). This can be accomplished by either high powder surface velocity (high impeller speed) or large spray zone width (larger or multiple nozzles). With increases in moisture content of the powder bed, bed cohesion increases and the surface powder velocity decreases, potentially leading to pooling of liquid at the surface which is undesirable. To compensate for the effects of cohesion, impeller speeds could be ramped up during the binder addition phase. Once binder addition is completed, the impeller speed could be reduced during the
Figure 5.8: Effects of cohesion and blade-rake angle on degree of mixing in the a) horizontal and b) vertical azimuthal plane.
consolidation and growth stage to minimise spatial shear gradients within the mixer, reducing the likelihood of binomial granule size distribution from breakage and attrition.

In terms of design parameters, the transmission of high shear-impact forces within these mixer is largely dependent on impeller design. Large blade-rake angle produces better flow and mixing results with more evenly distributed shear rates throughout the mixer.

### 5.5 Conclusions

This study has shown that:

- The average tangential and overall velocity of the powder decreases by 10% with the addition of moisture (cohesion) to the dry powder.

- Increasing blade-rake angle from $10^\circ$ to $90^\circ$ increases the average tangential and overall powder velocity by approximately 38%.

- The frictional properties of powders as measured by static methods cannot be used to explain the dynamic flow behaviour along the mixer wall. Instead of adhering to the wall, the cohesive (oil) sand is observed to lubricate the walls, decreasing the frictional retarding forces along the wall.

- The mixing performances as measured by RSD, shows that cohesion and blade-rake angle had no influence on circumferential mixing however, vertical mixing improves with higher blade-rake angle due to an increased thickness of the shear transmission layer.
Nomenclature

\( \dot{A} \) area flux of powder through the spray zone \( (m^2.s^{-1}) \)

\( d_d \) average liquid drop size \( (m) \)

\( \dot{V} \) volumetric spray rate \( (m^3.s^{-1}) \)

\( U_c \) representative collision velocity in the granulator \( (m.s^{-1}) \)

\( w \) mass ratio of liquid to solid \((-)\)

\( Y_g \) granule dynamic yield stress \( (kPa) \)

Greek symbols

\( \varepsilon_{\text{min}} \) minimum porosity the formulation \((-)\)

\( \rho_g \) granule density \( (g/mL) \)

\( \rho_l \) liquid density \( (g/mL) \)

\( \rho_s \) solid particles density \( (g/mL) \)

\( \Psi_a \) dimensionless spray flux \((-)\)
Abstract

This chapter investigates the use of a simplified network of idealised mixed regions called compartments to characterise and simulate particulate flow behaviour in a laboratory-scale vertical high shear mixer granulator. Compartment models (CMs) reduce the highly complex, non-ideal flow field by aggregating spatially similar flow properties, reducing computational time and offering an easier option for determining mixing performance. A general framework was derived for the development of CMs to accurately and efficiently capture the system behaviour. The framework describes methodologies to identify compartments and define the compartment structure and connectivity. Residence time distributions (RTDs) and particle fluxes between adjoining regions in the granulator were extracted from Positron Emission Particle Tracking (PEPT) experiments using cohesive sand and a two-bladed rectangular impeller.

Initially, a two-compartment model (two-CM) was developed consisting of two regions: an impeller and a circulation compartment. The RTD results showed that the mixing behaviour in the impeller compartment was well-mixed while the circulating compartment was a combination of a well-mixed volume interspersed with short-circuiting and internal circulation. The mixing pattern in the impeller compartment showed good agreement with a single, ideal
well-mixed continuously stirred tank reactor (CSTR) model. The circulation compartment was modelled as a combination of a single well-mixed CSTR for the short-circuiting paths in parallel with a cascade of well-mixed reactors for the internal circulation. The model showed reasonable fits to the complex mixing behaviour in the circulation compartment impeller at rotating speeds of 100-600 rpm.

A multi-compartment model (multi-CM) was developed to represent the key granulation processes of wetting and nucleation, growth and breakage. The mixer was partitioned into three regions: surface, circulation and impeller compartments. The increased partitioning created compartments with spatially similar properties enabling better fits to the ideal mixing models. The surface compartment showed reasonable agreement with the single well-mixed model while the circulation showed better agreement with the combination model at higher impeller speeds. The results also showed that spatial differences exist within the powder bed, for example the average velocity in the impeller compartment was five times greater than in either the circulation or the surface compartments at the highest impeller speed tested. The proposed framework and mixing compartment model can potentially be used in combination with population balance models, to better predict granulation processes through more realistic approaches to simulating powder flow.

6.1 Introduction

A major challenge in developing a model to predict the performance of granulators is capturing the kinetic processes and flow behaviour in an appropriate mathematical description. Traditionally, a general model has been used to lump physical kinetic processes inside a granulating vessel. The fundamental granulation rate processes of wetting and nucleation, consolidation and growth, and breakage and attrition, are assumed to all occur simultaneously in all regions of a granulator mixer.

Complex powder flow was experimentally observed to vary spatially in the mixer (Tran et al., 2005). As a consequence, it is inappropriate to assume all points inside the mixer have the same flow characteristic and kinetic properties. Compartment models (CMs) can be used to accurately model spatial differences in the mixer. This simplification of the behaviour of a spatially distributed physical system was also referred to as a lumped parameters model or a zonal model. A generalised approach to characterise powder flow behaviour using ideal systems could solve the problem of equipment dependent parameters (Michaels, 2003).

CMs have been applied extensively to characterise hydrodynamics in fluid processes (Mann et al., 1997, Alexopoulos et al., 2002, Howes et al., 2003, Bezzo et al., 2004, Bezzo and Macchietto, 2004, Kresta et al., 2004) and recently, to heterogeneous particulate mixing.
systems in V-blenders (Portillo et al., 2006a,b) and horizontal-axis mixers (Portillo et al., 2007, 2008). CMs’ main advantage over a full Discrete Element Models (DEM) is its ability to significantly reduce computational time (Portillo et al., 2007). Each spatial volume or compartment was assumed to be perfectly mixed locally, so that any variables being modelled was constant in each compartment. The connections between compartments were expressed by some form of transfer coefficient or flow rate. Standard differential-algebraic equation (DAE) solvers can easily tackle these dynamic models (Bermingham et al., 1998). Chemical engineering has long used two ideal reactor types, the continuous stirred tank reactor (CSTR) and plug flow reactor (PFR), to mathematically model mixing behaviour as a function of residence time and rates of reactions (Levenspiel, 1999). A similar approach would progress the study of powder flow towards quantitative characterisation of key transport properties (e.g. powder “viscosity”) and transport kinetics (e.g. growth and breakage kernels) (Michaels, 2003).

Freireich (2010) used particulate flow results from DEM simulation to develop compartment models for a vertical-axis mixer. Residence time distributions (RTD) were obtained from DEM for each compartment and fitted parameters were found for the simulated results. For the vertical-axis mixer, two distinct regions were identified as significant zones: a spray and a bed zone. The spray zone was modelled as a well-mixed tank in parallel with a series of tanks. The bed zone model was identical to the spray zone model with the addition of a short-cut path capturing a large fraction of particles with extremely short bed zone residence times. The spray zone corresponded well with the model however the bed zone largely ignored the oscillatory behaviour observed. For horizontal paddle mixers, Freireich et al. (2011) incorporated a recycle stream into the bed zone model capturing the observed oscillatory behaviour.

In this chapter, a general framework to develop CMs to characterise non-homogeneous flow in vertical axis mixers at high impeller speeds is presented. PEPT experiments will be used to capture the dynamics of particulate flow in different regions of the mixer. The resulting RTD plots will provide fitted parameters for the flow models. The effects of partitioning methods on the CM structure for both a two-CM and multi-CM will be investigated.

6.2 Model development and theory

6.2.1 General framework

In this section, a framework to develop CMs for high shear mixer granulators is proposed. The first step in setting up the model is to represent the mixing system through a set
Compartment models

Figure 6.1: General framework to develop compartment models for mixing systems.

6.2.2 Defining compartments

One key consideration in compartment modelling is the definition of zones that represent critical physical and chemical phenomena of the process. The methodology of defining compartments representing homogeneous and well-mixed regions in a mixing system is not trivial. There is often uncertainty concerning the boundary of compartments and the number of model compartments to include in the model. The larger the number of compartments, the greater the network complexity (inter-compartment flows) but the mixing dynamics of the compartments are simpler. The smaller the number of compartments, the greater the complexity of the internal mixing structure. Another limitation to the number of compartments is the computation cost, which is sufficiently large that it was desirable to begin with a small value and assess the adequacy of the fit to determine whether the current number
Figure 6.2: a) Schematic representations of granulation processes occurring in the mixer and the simplified compartment model diagrams of the b) two-CM and c) multi-CM where $C_{imp}$, $C_{cir}$ and $C_{sur}$ are the impeller, circulation and surface compartments respectively and $F$ are particle fluxes between the compartment.

of compartments describes the physical and chemical phenomena of the observed data. In this study, the boundary was chosen on the basis of mixer geometry and/or spatially similar powder properties.

Two-compartment model (two-CM)

Consider a simple CM made up of two compartments, namely the impeller and circulation zones (Fig. 6.2a). Several studies have described two distinct flow regions in mixers with rotating impellers: a high particle speed region close to the impeller and a low particle speed region elsewhere (Stewart et al., 2001a, Ng et al., 2007a). PEPT measurements have shown that the particle velocity was two time greater near the impeller than in the rest of the powder bed (Tran et al., 2005). Geometric arguments was used to demarcate the impeller-circulation boundary with a user-defined cylindrical grid of equal-volume cells. The impeller compartment consists of the cells encompassing the impeller and the clearance volume between the impeller bottom edge and the mixer. The remainder of the mixer was
designated as the circulation compartment. Relating the CM to granulation rate processes, the high shear impeller compartment could correspond to breakage and attrition processes and the low shear circulation region to growth and consolidation.

Multi-compartment model (multi-CM)

![Cumulative distribution plot of occupancy of cohesive sand and rectangular blade at 600 rpm. Only cells in which the tracer was observed have been considered in the distribution plots and compartments. The plateau in $F(x)$ demarcates a region of low occupancy, providing the threshold value for the surface compartment ($\text{occ} < 0.0009$).](image)

While the two-CM can give an adequate description of the flow field, a multi-CM would significantly increase the detail of mixing without considerable increases in the computational time relative to a DEM solution. A multi-CM is proposed consisting of three compartments, taking into account the granulation process of wetting and nucleation in the surface compartment, growth in the circulation compartment and breakage in the impeller compartment (Fig. 6.2b). The approach used to partition the powder bed was based on mixer geometry and internal occupancy gradients. The surface compartment of the powder bed is defined by a time occupancy criterion and all connecting cells are included in the surface zone. The value of the occupancy threshold was obtained from the observed plateau in the cumulative distribution plot of the occupancy at the highest impeller speed tested (600 rpm) (Fig. 6.3). In the 2D azimuthal-averaged gridded projection, the surface boundary was defined as the annular cells with an occupancy less than or equal to a threshold value of 0.09% of the total occupancy time (Fig. 6.4).
Figure 6.4: Schematic diagram to define the surface compartment in the azimuthal projection. Using a particle occupancy criterion \( 0 < \text{occ}_{\text{cell}} < 0.009 \), the shaded green region are cells in which the tracer was observed shows all the cells which in at least one particle (yellow circles) is present and falls between the criterion values. All internal interconnected cells are also included in the surface compartment.

Obtaining these properties experimentally with PEPT is both time consuming and expensive. An alternative is DEM modelling which has been used to simulate and obtain internal parameters for CMs (Portillo et al., 2008). A limitation of defining compartment boundaries empirically of both experimental and DEM simulations is that internal properties are specific to each material, mixer geometry and operating condition combination. Nevertheless, DEM coupled with CM shows great potential in the near future with rapid advancement in computational processing power (Freireich, 2010).

6.2.3 Global mixing properties

Global mixing and flow properties (i.e. RTDs and particle fluxes) were obtained from a single particle trajectory by extracting it from the time series of a virtual cluster of particles. Underlying the use of time averaged data is the theoretical principle of ergodicity, which states that a time average is equal to a population average. For this theorem to hold true, it is necessary to assume that experiments were run for a sufficiently long time for the tracer to visit all regions of the mixer.

Assuming ergodicity, consider the volume \( V \) of a compartment. The start of each new particle begins each time the trajectory crosses the boundary of the compartment volume (Fig.3.15a). The number of crossings is the particle flux \( F_{ij} \) from compartment \( C_{ij} \) to \( C_{i'j'} \).
This method generates a cluster of particles with different trajectory lengths (residence times $\tau$) from discrete passes through the volume. The RTD in each compartment can then be obtained and used to describe the mixing behaviour within the vessel or in this case, within each compartment.

### 6.2.4 Sub-compartment model generation

![Diagram](image)

**Figure 6.5:** Schematic of sub-compartment model for a) the single well-mixed CSTR model and b) the combination model consisting of a single well-mixed CSTR in parallel with a series of well-mixed CSTR. $\tau_1$ is the average time spent in the single well-mixed tank, $\lambda$ is the proportion of flow entering the single mixed tank, $\tau_3$ is the average time spent in the single well-mixed tank, $\tau_4$ is the average time spent in the series of well-mixed tanks and $N_2$ is the number of tanks in the series for Eqn. 6.1 and 6.3.

**Theory of ideal mixing systems**

Traditional approaches used to model flow behaviour in mixer granulators relied on the assumption of idealised well-mixed tanks. As demonstrated by Freireich (2010), if the compartments were characterised with only a well-mixed tank model, then significant errors would result. The general concept behind compartment models is the representation of
complex, non-ideal flows of real systems with a simple configuration of idealised and well
specified systems. This approach has commonly been used by chemical engineers to charac-
terise macro-mixing of the material flow using RTDs (Levenspiel, 1999). At one end of
the mixing model spectrum is the continuously stirred tank reactor (CSTR) in which all
particles are perfectly mixed with a RTD given by:

\[ E(t) = \frac{1}{\tau_1} e^{-t/\tau_1} k_1 \]  (6.1)

where \( \tau_1 \) is the average time spent in the single well-mixed tank and \( k_1 \) is a scaling
constant.

At the other end is the perfect plug flow reactor (PFR). All the particles in a PFR
exhibit identical behaviour with the same velocity and residence time. Under real industrial
conditions, the mixing behaviour lie between these two extremes. An intermediate model is
a cascade of perfectly mixed tanks given by:

\[ E(t) = \frac{1}{(N_1 - 1)! \tau_2} \left( \frac{t}{\tau_2} \right)^{N_1-1} e^{-t/\tau_2} \]  (6.2)

where \( N_1 \) is the number of perfectly mixed equal volume tanks in the series and \( \tau_2 \) is the
average residence time in each tank. It should be noted that a cascade of an infinite series
of well-mixed tanks models a PFR.

The non-ideal mixer approach considers a network of idealised well-mixed tanks to model
the flow behaviour in different compartments. The CM proposed by Freireich (2010) identi-
fied here as the combination model, was generated by connecting a single well-mixed CSTR
in parallel with a series of well-mixed CSTR (Fig. 6.5a), its RTD is given by:

\[ E(t) = (\lambda e^{-t/\tau_3} + (1 - \lambda)) \frac{N_2}{\tau_4} \left( \frac{1}{(N_2 - 1)!} \right) \left( \frac{N_2 t}{\tau_4} \right)^{N_2-1} e^{-N_2 t/\tau_4} k_2 \]  (6.3)

where \( \lambda \) is the proportion of flow entering the single mixed tank, \( \tau_3 \) is the average time
spent in the single well-mixed tank, \( \tau_4 \) is the average time spent in the series of well-mixed
tanks, \( N_2 \) is the number of tanks in the series and \( k_2 \) is a scaling constant.

### 6.2.5 Deriving model parameters

The mixing models were fitted to the extracted RTD to obtain values of the model pa-
rameters. The goodness of fit was expressed as the coefficient of determination \( (R^2) \) given
by:
Compartment models

\[ R^2 = 1 - \frac{SS_{resid}}{SS_{total}} \]  \hspace{1cm} (6.4)

where \( SS_{total} \) is the total sum of squares (the sum of the squared differences from the mean of the residence time) and \( SS_{resid} \) is the sum of the squared residuals from the regression (the sum of the square of the differences between the experimental values and the outcomes of the model divided by the experimental value) given by:

\[ SS_{resid} = \sum \left( \frac{\tau_{experiment} - \tau_{model}}{\tau_{experiment}} \right)^2 \]  \hspace{1cm} (6.5)

The closer \( R^2 \) is to 1, the better the fit. Optimisation of the model parameters was performed towards a maximum coefficient of determination.

\section{6.3 Experimental method}

The powder flow experiments were carried out in a custom-made vHSM granulator. The bottom-driven motor shaft was controlled by a variable speed control box that enabled the impellers to rotate at speeds of 0-667 rpm. The connecting mixer shaft assembly is of the same internal dimensions used by Wellm (1997) to take advantage of the interchangeable bowls. A 0.21 m diameter mixing bowl with a volume capacity of 6.9 L was used for this investigation (Fig. 3.6). The mixer bowl was constructed from stainless steel pipes. For the 2 kg powder load, the ratio of powder bed height to bowl diameter \( H/D \) was approximately 0.29.

A 90\(^\circ\) rectangular two-bladed impeller was used for all experiments. The impeller was designed to be similar to impellers found in commercially available mixers (e.g. Aeromatic-Fielder) with a height and width of 10 mm and 50 mm, respectively. This impeller produces both impact and shear forces within the mixer. No chopper blades or baffles were present in the mixer to simplify the flow field. The powder material under investigation was cohesive sand, a mixture of sand and silicone oil. A summary of the material properties is found in Table 3.1. Impeller speeds varied between 100 - 600 rpm.

\subsection{6.3.1 Positron Emission Particle Tracking (PEPT) experiments}

Direct measurements of three-dimensional motion of powder within the mixer were carried out using the PEPT technique. This technique has been described in detailed by other workers (Parker et al., 2002, Bridgwater et al., 2004). PEPT involves the detection of gamma-rays produced by the result of positron decay of a radioactive tracer particle. The
tracer emits positrons that annihilate with electrons to produce back-to-back gamma-rays. Two camera detectors each 50 x 40 cm$^2$ and situated 54 cm apart, were used to detect the tracer using the location of several intersecting gamma-rays. The camera can follow a tracer moving at 1 m/s to within 0.5 mm at a data acquisition rate of over 250 times per second (Bridgwater et al., 2004). This enables detailed studies of powder flow at speeds of industrial significance.

For the tracer particle, a resin bead of 212 $\mu$m diameter and density of 1050 kg/m$^3$ was used. It was activated by an ion exchange method with radioactive water produced in a cyclotron (Bridgwater et al., 2004). For a typical PEPT experiment, the powder was loaded with the tracer and allowed to mix for 2 minutes in order to reach steady state flow before the one hour data acquisition commenced.

Velocities were calculated from discrete 3D spatial and temporal data obtained from PEPT experiments using a least squares fitted (LSF) algorithm. The LSF method estimated the tracer velocity along its trajectory by fitting a number of data points $i$ to an $n$th-order polynomial. For this analysis, a velocity vector was fitted to five positional data points along the tracer trajectory, with the average of the points used as the vector origin.

### Cell properties

Eulerian maps were obtained by spatially discretising the mixing vessel into equal volume cells projected onto the azimuthal plane. For the purpose of identifying mixing behaviour, it was assumed that the vertical mixer was axial-symmetric and a 2D azimuthally-averaged view was sufficient to capture the flow field and the mixing pattern in the mixer. Using a user-defined cylindrical grid of equal-volume annular cells, the occupancy of each cell $occ_{cell}$ is defined as the fraction of time the tracer spent in each cell of the overall experimental time $t_T$ given by:

$$occ_{cell} = \frac{\sum \tau_{cell}}{t_T} \approx \frac{n_p}{n_T}$$

(6.6)

where $\tau_{cell}$ is the residence time of each pass through the cell. Note that only cells in which the tracer is observed is considered in the compartments. Alternatively, occupancy can be evaluated from a virtual bed of discrete particles generated by dividing the Lagrangian trajectory into strings at constant time intervals (Fig. 3.15b). Occupancy is then the ratio of the number of particles (strings) contained in a cell $n_p$, to the total number of particles in the mixer $n_T$ (Eqn. 6.6).

The average residence time of a cell $\tau_{cell}$ is previously given by:
\[ \bar{\tau}_{\text{cell}} = \frac{\sum n_{\text{pass}} \tau_{\text{cell}}}{n_{\text{pass}}} = \frac{n_p k}{n_{\text{pass}}} \]  

where \( n_{\text{pass}} \) is the number of tracer visits through the cell and \( k \) is the constant time interval between each particle. Internal properties of the cells (e.g. particle velocities in each component, overall 3D and 2D (radial-axial) particle velocity, standard deviation of the velocity, tracer acceleration, number and frequency of visits are also obtained.

**Compartment properties**

The volume of a compartment is the total volume of annular cells with particles present, regardless of if the cells are only partially filled as in the case with surface cells. The occupancy of each compartment \( \text{occ}_{\text{comp}} \) is the sum of \( \text{occ}_{\text{cell}} \) within the compartment. The average velocities in each compartment were obtained by multiplied the cell properties by \( \text{occ}_{\text{cell}} \) and summing the values of that compartment.

### 6.4 Results and discussion

#### 6.4.1 Case study: Two-compartment model (two-CM)

The spatial distributions of the two-CM for impeller speeds from 100-600 rpm are shown in Fig. 6.6a and the resulting volume and occupancy values are displayed in Table 6.1. As the impeller compartment was defined using a geometric criterion, the volume varied between 511-520 \( cm^3 \) depending on if all cells were occupied around the impeller shaft. The bed volume of the circulation compartment varied considerably based on the powder flow behaviour and the degree of bed dilation. An initial increase was observed reaching the maximum bed dilation when the bumping behaviour dominated (300 rpm) before decreasing as stable, compact roping flow dominated. This shift in the shape of the powder bed as speed increased also caused a decrease in the occupancy of the impeller compartment.

**Residence time distribution**

Fig. 6.7a,b shows the experimentally obtained RTDs for the two-CM for 100-600 rpm. In the impeller compartments, the RTD profiles shows simple exponential decay functions with extended tails, an indication of a well-mixed compartment for all cases except at 100 rpm. At the lowest speed, an offset was observed resulting from longer periods between intermittent blade passes. The circulation compartment was more complex, an initial peak \( (\tau < 0.4 \text{ s}) \) was present at all speeds resulting from a significant portion of particles traversing
6.4 Results and discussion

Figure 6.6: Results of the spatial distribution of compartments in the a) two-CM and b) multi-CM. The red squares ■, blue circles ● and green triangles ▲ represent cells within the impeller, circulation and surface compartment, respectively.
Figure 6.7: Results of the residence time distribution and fitted mixing model for the a) impeller compartment (identical for both two-CM and multi-CM), b) two-CM circulation compartment, c) multi-CM circulation model and d) multi-CM surface compartment. The plots are truncated to highlight interesting behaviours however Table 6.1 shows maximum residence time values.
### Table 6.1: Properties of each compartment from experimental PEPT data for the two-CM and multi-CM: occupied volume \( \text{vol} \), occupancy \( \text{occ} \), average particle velocity \( v_p \), average dimensionless particle velocity \( v_p^* \), average residence time \( \tau \), one standard deviation of average residence time \( \sigma_\tau \) (68% percentile) and maximum residence time \( \tau_{\text{max}} \).

<table>
<thead>
<tr>
<th>N (rpm)</th>
<th>CM Type</th>
<th>Comp</th>
<th>( \text{vol} ) (cm(^3))</th>
<th>( \text{occ} ) (%)</th>
<th>( v_p ) (m/s)</th>
<th>( v_p^* ) (-)</th>
<th>( \tau ) (s)</th>
<th>( \sigma_\tau ) (s)</th>
<th>( \tau_{\text{max}} ) (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>Both</td>
<td>( C_{\text{imp}} )</td>
<td>511</td>
<td>24.2</td>
<td>0.14</td>
<td>0.13</td>
<td>0.29</td>
<td>1.18</td>
<td>37.13</td>
</tr>
<tr>
<td></td>
<td>Two</td>
<td>( C_{\text{cir}} )</td>
<td>1896</td>
<td>75.80</td>
<td>0.20</td>
<td>0.18</td>
<td>0.92</td>
<td>3.82</td>
<td>70.33</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>( C_{\text{cir}} )</td>
<td>1541</td>
<td>74.29</td>
<td>0.20</td>
<td>0.18</td>
<td>0.70</td>
<td>2.11</td>
<td>33.35</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>( C_{\text{sur}} )</td>
<td>355</td>
<td>1.52</td>
<td>0.17</td>
<td>0.15</td>
<td>0.06</td>
<td>0.04</td>
<td>0.24</td>
</tr>
<tr>
<td>200</td>
<td>Both</td>
<td>( C_{\text{imp}} )</td>
<td>520</td>
<td>16.46</td>
<td>0.31</td>
<td>0.14</td>
<td>0.13</td>
<td>0.54</td>
<td>12.89</td>
</tr>
<tr>
<td></td>
<td>Two</td>
<td>( C_{\text{cir}} )</td>
<td>2208</td>
<td>83.54</td>
<td>0.45</td>
<td>0.20</td>
<td>0.66</td>
<td>1.54</td>
<td>17.57</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>( C_{\text{cir}} )</td>
<td>1801</td>
<td>81.94</td>
<td>0.45</td>
<td>0.20</td>
<td>0.49</td>
<td>1.03</td>
<td>8.64</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>( C_{\text{sur}} )</td>
<td>407</td>
<td>1.60</td>
<td>0.33</td>
<td>0.15</td>
<td>0.04</td>
<td>0.03</td>
<td>0.33</td>
</tr>
<tr>
<td>250</td>
<td>Both</td>
<td>( C_{\text{imp}} )</td>
<td>520</td>
<td>16.62</td>
<td>0.49</td>
<td>0.18</td>
<td>0.13</td>
<td>0.41</td>
<td>12.83</td>
</tr>
<tr>
<td></td>
<td>Two</td>
<td>( C_{\text{cir}} )</td>
<td>2442</td>
<td>83.38</td>
<td>0.54</td>
<td>0.20</td>
<td>0.67</td>
<td>1.13</td>
<td>12.98</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>( C_{\text{cir}} )</td>
<td>1844</td>
<td>81.20</td>
<td>0.54</td>
<td>0.20</td>
<td>0.48</td>
<td>0.75</td>
<td>5.28</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>( C_{\text{sur}} )</td>
<td>597</td>
<td>2.18</td>
<td>0.40</td>
<td>0.15</td>
<td>0.05</td>
<td>0.03</td>
<td>0.31</td>
</tr>
<tr>
<td>300</td>
<td>Both</td>
<td>( C_{\text{imp}} )</td>
<td>520</td>
<td>13.80</td>
<td>0.34</td>
<td>0.10</td>
<td>0.12</td>
<td>0.52</td>
<td>12.80</td>
</tr>
<tr>
<td></td>
<td>Two</td>
<td>( C_{\text{cir}} )</td>
<td>2892</td>
<td>86.20</td>
<td>0.58</td>
<td>0.18</td>
<td>0.74</td>
<td>1.43</td>
<td>21.64</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>( C_{\text{cir}} )</td>
<td>2260</td>
<td>83.67</td>
<td>0.59</td>
<td>0.18</td>
<td>0.50</td>
<td>0.85</td>
<td>7.62</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>( C_{\text{sur}} )</td>
<td>632</td>
<td>2.53</td>
<td>0.40</td>
<td>0.12</td>
<td>0.05</td>
<td>0.03</td>
<td>0.18</td>
</tr>
<tr>
<td>400</td>
<td>Both</td>
<td>( C_{\text{imp}} )</td>
<td>511</td>
<td>13.10</td>
<td>0.74</td>
<td>0.17</td>
<td>0.10</td>
<td>0.32</td>
<td>7.91</td>
</tr>
<tr>
<td></td>
<td>Two</td>
<td>( C_{\text{cir}} )</td>
<td>2719</td>
<td>86.90</td>
<td>0.68</td>
<td>0.15</td>
<td>0.67</td>
<td>0.89</td>
<td>11.07</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>( C_{\text{cir}} )</td>
<td>1992</td>
<td>84.22</td>
<td>0.68</td>
<td>0.15</td>
<td>0.46</td>
<td>0.58</td>
<td>4.03</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>( C_{\text{sur}} )</td>
<td>727</td>
<td>2.68</td>
<td>0.62</td>
<td>0.14</td>
<td>0.05</td>
<td>0.03</td>
<td>0.22</td>
</tr>
<tr>
<td>500</td>
<td>Both</td>
<td>( C_{\text{imp}} )</td>
<td>520</td>
<td>7.19</td>
<td>0.89</td>
<td>0.16</td>
<td>0.18</td>
<td>0.32</td>
<td>3.55</td>
</tr>
<tr>
<td></td>
<td>Two</td>
<td>( C_{\text{cir}} )</td>
<td>2624</td>
<td>92.81</td>
<td>0.72</td>
<td>0.13</td>
<td>2.35</td>
<td>1.99</td>
<td>14.98</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>( C_{\text{cir}} )</td>
<td>2052</td>
<td>90.86</td>
<td>0.72</td>
<td>0.13</td>
<td>0.99</td>
<td>1.33</td>
<td>11.58</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>( C_{\text{sur}} )</td>
<td>571</td>
<td>1.96</td>
<td>0.70</td>
<td>0.13</td>
<td>0.04</td>
<td>0.03</td>
<td>0.27</td>
</tr>
<tr>
<td>600</td>
<td>Both</td>
<td>( C_{\text{imp}} )</td>
<td>520</td>
<td>9.83</td>
<td>1.66</td>
<td>0.25</td>
<td>0.08</td>
<td>0.16</td>
<td>4.07</td>
</tr>
<tr>
<td></td>
<td>Two</td>
<td>( C_{\text{cir}} )</td>
<td>2295</td>
<td>90.17</td>
<td>0.90</td>
<td>0.14</td>
<td>0.71</td>
<td>0.84</td>
<td>18.05</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>( C_{\text{cir}} )</td>
<td>1775</td>
<td>88.73</td>
<td>0.90</td>
<td>0.14</td>
<td>0.43</td>
<td>0.61</td>
<td>10.44</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>( C_{\text{sur}} )</td>
<td>520</td>
<td>1.44</td>
<td>0.74</td>
<td>0.11</td>
<td>0.02</td>
<td>0.03</td>
<td>0.56</td>
</tr>
</tbody>
</table>

As the boundary of the impeller compartment \( (C_{\text{imp}}) \) for both the two-CM and multi-CM are identical, the results are displayed as “Both”.
Compartment models

Table 6.2: Model parameters fitted to PEPT experimental data for the two-CM and multi-CM.

<table>
<thead>
<tr>
<th>RPM</th>
<th>Comp</th>
<th>Type</th>
<th>$N$</th>
<th>$t_1$</th>
<th>$k_1$</th>
<th>$R^2$</th>
<th>$t_2$</th>
<th>$t_3$</th>
<th>$\lambda$</th>
<th>$k_2$</th>
<th>$R^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>Both</td>
<td>C_{imp}</td>
<td>0.293</td>
<td>0.117</td>
<td>0.690</td>
<td></td>
<td>0.214</td>
<td>0.208</td>
<td>0.014</td>
<td>3.919</td>
<td>22</td>
</tr>
<tr>
<td></td>
<td>Two</td>
<td>C_{cir}</td>
<td>0.304</td>
<td>0.113</td>
<td>0.649</td>
<td></td>
<td>0.018</td>
<td>0.201</td>
<td>0.047</td>
<td>1.573</td>
<td>15</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>C_{cir}</td>
<td>0.330</td>
<td>0.111</td>
<td>0.660</td>
<td></td>
<td>0.020</td>
<td>0.214</td>
<td>0.036</td>
<td>1.973</td>
<td>16</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>C_{sur}</td>
<td>0.079</td>
<td>0.012</td>
<td>0.666</td>
<td></td>
<td>0.079</td>
<td>6.178</td>
<td>0.395</td>
<td>0.030</td>
<td>7</td>
</tr>
<tr>
<td>200</td>
<td>Both</td>
<td>C_{imp}</td>
<td>0.052</td>
<td>0.145</td>
<td>1.000</td>
<td></td>
<td>0.052</td>
<td>50.000</td>
<td>0.014</td>
<td>10.379</td>
<td>57</td>
</tr>
<tr>
<td></td>
<td>Two</td>
<td>C_{cir}</td>
<td>0.239</td>
<td>0.110</td>
<td>0.471</td>
<td></td>
<td>0.095</td>
<td>0.163</td>
<td>0.034</td>
<td>0.985</td>
<td>57</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>C_{cir}</td>
<td>0.261</td>
<td>0.115</td>
<td>0.463</td>
<td></td>
<td>44.491</td>
<td>0.136</td>
<td>0.170</td>
<td>0.802</td>
<td>11</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>C_{sur}</td>
<td>0.042</td>
<td>0.013</td>
<td>0.826</td>
<td></td>
<td>0.042</td>
<td>50.739</td>
<td>0.010</td>
<td>1.257</td>
<td>67</td>
</tr>
<tr>
<td>250</td>
<td>Both</td>
<td>C_{imp}</td>
<td>0.049</td>
<td>0.145</td>
<td>0.999</td>
<td></td>
<td>0.049</td>
<td>3.044</td>
<td>0.983</td>
<td>0.147</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td>Two</td>
<td>C_{cir}</td>
<td>0.144</td>
<td>0.092</td>
<td>0.961</td>
<td></td>
<td>0.088</td>
<td>0.163</td>
<td>0.003</td>
<td>12.531</td>
<td>3</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>C_{cir}</td>
<td>0.188</td>
<td>0.098</td>
<td>0.933</td>
<td></td>
<td>0.176</td>
<td>0.121</td>
<td>0.066</td>
<td>1.375</td>
<td>60</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>C_{sur}</td>
<td>0.053</td>
<td>0.013</td>
<td>0.775</td>
<td></td>
<td>0.053</td>
<td>50.719</td>
<td>0.008</td>
<td>1.529</td>
<td>64</td>
</tr>
<tr>
<td>300</td>
<td>Both</td>
<td>C_{imp}</td>
<td>0.027</td>
<td>0.256</td>
<td>1.000</td>
<td></td>
<td>0.027</td>
<td>50.000</td>
<td>0.012</td>
<td>21.283</td>
<td>70</td>
</tr>
<tr>
<td></td>
<td>Two</td>
<td>C_{cir}</td>
<td>0.260</td>
<td>0.098</td>
<td>0.560</td>
<td></td>
<td>23.726</td>
<td>0.151</td>
<td>0.001</td>
<td>66.444</td>
<td>7</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>C_{cir}</td>
<td>0.262</td>
<td>0.112</td>
<td>0.483</td>
<td></td>
<td>25.796</td>
<td>0.140</td>
<td>0.003</td>
<td>32.377</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>C_{sur}</td>
<td>0.060</td>
<td>0.012</td>
<td>0.697</td>
<td></td>
<td>0.060</td>
<td>51.450</td>
<td>0.051</td>
<td>0.243</td>
<td>11</td>
</tr>
<tr>
<td>400</td>
<td>Both</td>
<td>C_{imp}</td>
<td>0.035</td>
<td>0.203</td>
<td>1.000</td>
<td></td>
<td>0.035</td>
<td>50.000</td>
<td>0.013</td>
<td>15.124</td>
<td>60</td>
</tr>
<tr>
<td></td>
<td>Two</td>
<td>C_{cir}</td>
<td>0.173</td>
<td>0.068</td>
<td>0.806</td>
<td></td>
<td>0.136</td>
<td>0.649</td>
<td>0.634</td>
<td>0.097</td>
<td>33</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>C_{cir}</td>
<td>0.125</td>
<td>0.077</td>
<td>0.922</td>
<td></td>
<td>0.114</td>
<td>0.635</td>
<td>0.760</td>
<td>0.098</td>
<td>30</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>C_{sur}</td>
<td>0.050</td>
<td>0.012</td>
<td>0.701</td>
<td></td>
<td>0.050</td>
<td>50.532</td>
<td>0.013</td>
<td>0.924</td>
<td>68</td>
</tr>
<tr>
<td>500</td>
<td>Both</td>
<td>C_{imp}</td>
<td>0.063</td>
<td>0.127</td>
<td>0.988</td>
<td></td>
<td>0.046</td>
<td>0.466</td>
<td>0.792</td>
<td>0.172</td>
<td>3</td>
</tr>
<tr>
<td></td>
<td>Two</td>
<td>C_{cir}</td>
<td>3.092</td>
<td>0.117</td>
<td>0.590</td>
<td></td>
<td>3.128</td>
<td>1.654</td>
<td>0.564</td>
<td>0.136</td>
<td>7</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>C_{cir}</td>
<td>0.127</td>
<td>0.064</td>
<td>0.916</td>
<td></td>
<td>0.123</td>
<td>1.544</td>
<td>0.315</td>
<td>0.201</td>
<td>6</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>C_{sur}</td>
<td>0.029</td>
<td>0.013</td>
<td>0.899</td>
<td></td>
<td>0.019</td>
<td>0.062</td>
<td>0.841</td>
<td>0.014</td>
<td>68</td>
</tr>
<tr>
<td>600</td>
<td>Both</td>
<td>C_{imp}</td>
<td>0.048</td>
<td>0.117</td>
<td>0.996</td>
<td></td>
<td>0.025</td>
<td>0.221</td>
<td>0.874</td>
<td>0.184</td>
<td>3</td>
</tr>
<tr>
<td></td>
<td>Two</td>
<td>C_{cir}</td>
<td>0.691</td>
<td>0.100</td>
<td>0.490</td>
<td></td>
<td>0.028</td>
<td>0.545</td>
<td>0.071</td>
<td>0.696</td>
<td>32</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>C_{cir}</td>
<td>0.030</td>
<td>0.094</td>
<td>0.901</td>
<td></td>
<td>0.030</td>
<td>0.547</td>
<td>0.676</td>
<td>0.140</td>
<td>30</td>
</tr>
<tr>
<td></td>
<td>Multi</td>
<td>C_{sur}</td>
<td>0.009</td>
<td>0.012</td>
<td>0.994</td>
<td></td>
<td>0.009</td>
<td>50.512</td>
<td>0.007</td>
<td>1.640</td>
<td>57</td>
</tr>
</tbody>
</table>

through the compartment in short passes, an indication of possible short-circuiting paths within the compartment. Following the initial peak, there were minor oscillations that increased in prominence as speeds increased, a potential sign of strong internal circulation or parallel paths to the exit. In addition, the extended exponentially decaying tail indicates an underlying well-mixed tank or a series of tanks.

The average residence time, standard deviation and maximum residence time for the two-CM are shown in Table 6.1. Poor mixing and the existence of longer circulation paths at the lowest speed resulted in a wide range of residence times ($\sigma > 1$) and maximums that were two magnitudes greater than the average. Above $100$ rpm, the average residence time was
generally insensitive to impeller speeds in the range tested for both compartments as short-circuiting skewed the calculations of the mean residence time. The spread of the residence times narrowed with increasing speed as the powder flow moved towards more stable roping behaviour. The decrease in short-circuiting paths as observed by the low prominence of a peak at $\tau < 0.4\, \text{s}$ for 500 rpm (Fig. 6.7b) resulted in a higher average residence time. This can be explained by a “sweet spot” in the resonance of the powder bed with the impeller blades in minimal contact.

The spatial location of individual passes (grouped by residence time) within the powder bed azimuthal profile provide further explanations for the observed results (Fig. 6.8). The spatial RTD plots through the impeller and circulation compartments over 100 s or 1000 s periods show that very short passes ($\tau < 0.4\, \text{s}$) were found near the compartment boundary corresponding to short-circuiting paths. Physically, these paths can be attributed to bumping behaviour as the impeller blades passed beneath the powder bed. The height of these paths at low speeds is equivalent to the height of the impeller blade. Intermediate passes between ($0.4\, \text{s} < \tau < 4\, \text{s}$) move through the circulation compartment in concentric loops about an internally circulating core. Long passes ($\tau > 4\, \text{s}$) occurred when the particles were in a slow flowing stream moving alongside the mixer wall due to particle-wall friction. These longer circulation path particles were pushed circumferentially around the mixer, rising steadily to the top of the bed before cascading down the face of the powder bed.

**Model fitting**

Both the well-mixed and combination models show excellent agreement in the impeller compartment for all impeller speeds except the lowest (100 rpm) (Fig. 6.7a) with resulting model parameters shown in Table 6.2. At the lowest speed the well-mixed model failed to model the offset of the exponential decay curve however, the simplicity of this model with only two fitted parameters makes it the preferred choice over the combination model for all speeds except for the lowest. In the circulation compartment the combination model was more robust with five fitted parameters, enabling it to capture the initial large peak in the single well-mixed tank in addition to the oscillatory peaks within the parallel series of well-mixed tanks. However, at 400 and 500 rpm the model failed to simulate the complex flow pattern. One way to solve this issue is to develop a more complex mixing model or another option is to partition the circulation compartment further into potential simpler ideal compartment models (e.g. multi-CM).
Figure 6.8: Lagrangian particle trajectories grouped by residence times of individual passes through the a) impeller and b) circulation compartments over 100 s or 1000 s period for cohesive sand using the rectangular impeller at 100 - 600 rpm. The location of the impeller was shown by - - - .
6.4.2 Case study: Multi-compartment mixing model (multi-CM)

The spatial distribution of the mixer to include an additional surface compartment for the multi-CM is presented in Fig 6.7b. The occupancy criterion created a surface compartment with a depth ranging from 1-5 cells (≈ 20-100 particles) depending on the flow behaviour. The greater the dominance of bumping behaviour, the larger the surface compartment volume. Occupancy was less than 3% in the surface compartment across all impeller speeds tested (Table 6.1). This can be explained by the low occupancy criterion used to partition the surface compartment resulting in partially filled cells. The powder flow within this region was generally chaotic, interacting mostly through collisions with other particles as they cascaded down the face of the powder bed in a dilute, chaotic phase.

The RTD plots and corresponding ideal mixing model fits of the multi-CM are shown in Fig. 6.7c,d. As the impeller compartment mixing model (Eqn. 6.1) was identical to the two-CM, refer to Section 6.4.1 for the results. For the circulation compartment, the combination mixing model (Eqn. 6.3) showed improved fits for 400 and 500 rpm as the increased partitioning of the powder bed into spatially similar properties allowed the regions to resemble more closely ideal mixing systems. In the surface compartment, the RTD plots resembled an exponential decay curve with multiple peaks caused by the presence of bumping behaviour and longer circulation paths near the mixer wall. With increasing impeller speeds, the peaks decreased until at 600 rpm the peaks were completely eliminated with fully developed roping behaviour and the curve resembles a well-mixed system. The impeller compartment showed comparable results using both the combination and well-mixed sub-compartment model and the resulting fitted parameters are shown in Table 6.2. In general, the combination model showed very good agreements with the RTDs in the circulation compartments and only adequate fits of both models in the impeller compartment across the operation conditions of interest. The fitted model parameters are displayed in Table 6.2.

Powder fluxes

Accurate dynamic simulation of the mixing process requires knowledge of powder fluxes between compartments to describe the convective-dispersive flow. The time domain was discretised using the constant time interval method (Doucet et al., 2008). Following the approach described by Portillo et al. (2008), the particle flux $F_{ij}$ was defined as the ratio of number of particles $n_{ij}$ transferred from one compartment $C_i$ to a neighbouring compartment $C_j$ to the total number of particles within the compartment $N_i$:

$$F_{ij} = \frac{n_{ij}}{N_i}$$  \hspace{1cm} (6.7)
The higher the flux value, the better the flow between the interconnected compartments. At each time step $k$, the change in the number of particles in compartment $C_i$, was denoted as $\Delta F_{ij}$. In the closed mixing system, the change in each species $i$ in all compartments at every time step must equal zero given by:

$$\sum_{i=0}^{N} \sum_{j=0}^{N} \Delta F_{i\neq j} = 0$$

(6.8)

where $w$ is the sum of all interconnected compartments.

The effect of impeller speed on vertical fluxes at steady state for the multi-CM is shown in Fig. 6.9. The particle flux from the impeller to the circulation compartment remained relatively insensitive to impeller speeds until 600 rpm, at which point it increased. This result demonstrates the transition from bumping to roping flow behaviour described previously (refer to Chapter 4). The reverse particle flux from the circulation back to the impeller compartment showed a steady decrease from 100 - 500 rpm and then a slight increase at 600 rpm, which is as expected as bumping behaviour diminished under the more dominant stable roping flow. The fluxes between the other compartments were less significant to the overall mixing of the powder bed.

**Figure 6.9:** Average particle fluxes between interconnecting compartments of the multi-CM.
6.5 Implications for granulation processes

The proposed framework has several implications for granulation processes. Consider a multi-CM where the three compartments: impeller, circulation and surface, represented the three key granulation mechanisms occurring in the mixer: wetting and nucleation, growth and coalescence, and breakage and attrition, respectively. Take wetting and nucleation for example. Particle dynamics influences the time particles spend in the surface zone and their interactions with surrounding particles. From observing only the surface layer, it has been assumed that particles at the surface operating in the roping regime would exhibit good bed turnover rates, providing fresh powder to the spray zone (Litster et al., 2002). This is true for the majority of the powder at the surface. However, the RTD measurements from PEPT experiments have revealed an extended tail where surface powder near the wall has a tendency to recirculate in longer circulation path powder while still moving circumferentially around the mixer. During spraying, liquid binder would tend to pool in these regions allowing longer penetration times and hence, inhomogeneity of the liquid distribution among the particles. This leads to wet, oversized agglomerates and a broad range of granule sizes, undesirable for controlled granulation. One way to avoid this would be to have sprays angled parallel to the falling surface instead of directly above the mixer. Another option would be to design mixers with tapered edges to encourage bed turnover. Baffles or choppers could be added to encourage the break-up of the flow however, this adds another dimension of complication in modelling the flow behaviour.

6.6 Limitations

Although it is possible to use RTD data from PEPT experiments, the expense and time-consuming process of obtaining PEPT data makes this technique rather limiting for modelling of powder flows. In addition, constraints of equipment size within the PEPT scanner makes this technique restricted to laboratory scale equipment, though new advancements in portable PEPT could make this a viable option for larger industrial sized equipment (Ingram et al., 2007). The development of DEM models validated by PEPT experimental data will provide a more useful and less time expensive tool for validating simulated particulate flow models.
6.7 Conclusion

The single well-mixed CSTR model showed good agreement in the impeller compartment and reasonable agreement in the surface compartment. The combination model consisting of a single well-mixed CSTR in parallel with a series of well-mixed CSTR was used successfully to model the complex behaviour in the circulation compartment. Spatial differences in volume, occupancies and velocities were observed within different regions of the mixer across the impeller rotating speeds tested. Further work to incorporate compartment mixing models with a general multi-dimensional population balance model has the potential to improved mixing performances and improve predictive granulation models.

Nomenclature

\( C_{\text{cir}}, C_{\text{imp}}, C_{\text{sur}} \) circulation, impeller, surface compartment

\( D \) bowl diameter

\( d_{10}, d_{50}, d_{90} \) 10th, 50th, 90th diameter percentile \( \mu m \)

\( E_t \) residence time distribution (s\(^{-1}\))

\( F_{ijk} \) particle flux from compartment \( i \) to \( j \) (–)

\( F_z \) cumulative distribution (–)

\( k \) constant time interval (s)

\( k_1 \) scaling constant (–)

\( H \) powder bed height (m)

\( N \) impeller rotational speed (rpm)

\( N_1, N_2 \) number of tanks in series (–)

\( N_i \) total number of particles in compartment (#)

\( n_p \) number of particles (#)

\( n_{\text{pass}} \) number of tracer passes through the cell (#)

\( n_T \) total number of particles in the mixer

\( \text{occ}_{\text{cell}} \) occupancy in each cell (–)

\( R^2 \) coefficient of determination (–)

\( SS_{\text{resid}} \) sum of the squared residuals (s\(^2\))

\( SS_{\text{total}} \) total sum of squares (s\(^2\))

\( t \) time (s)

\( t_T \) overall experimental time (s)

\( v_p \) average powder velocity (ms\(^{-1}\))

\( v_p^* \) dimensionless average powder velocity (–)

\( v_{\text{vol}} \) volume of compartment cm\(^3\)

\( \overline{\pi} \) overall mean particle concentration

\( w \) sum of all interconnected compartment

\( z \) height of impeller compartment (m)
### Greek symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \lambda )</td>
<td>proportion of flow entering the single tank ((-))</td>
</tr>
<tr>
<td>( \sigma )</td>
<td>standard deviation ((-))</td>
</tr>
<tr>
<td>( \tau, \bar{\tau} )</td>
<td>residence time, average residence time ((s))</td>
</tr>
<tr>
<td>( \tau_{max} )</td>
<td>maximum residence time ((s))</td>
</tr>
</tbody>
</table>
Conclusions and Recommendations

7.1 Summary

This thesis has investigated the flow behaviour of powder material in a vertical-axis high shear mixer granulator. The objectives of the study were to develop a powder flow regime map to characterise the flow field, to investigate the effect of impeller speed, powder cohesion and blade-rake angle on powder flow and mixing behaviour and to outline a framework for compartmental models.

In Chapter 4, two distinct powder flow regimes were identified. The bumping regime was observed to have complex flow behaviour with either single or double internal vertical circulations. Low levels of bed rotation and vertical turnover in addition to a small shear layer resulted in undesirable conditions for controlled granulation. In contrast, the roping regime is characterised by a high degree of bed rotation, good vertical bed turnover and a shear layer that extends to the entire height of the bed with high powder velocities in the impeller zone, well above the critical Froude powder velocity. Clearly, granulator mixers should operate in the roping regime for controlled granulation. Transitional states caused by bed resonance show apparent roping behaviour which visually appears to be similar to roping regime however, powder velocity results show that these states still fall within the gravity dominated bumping regime. Operating in this transitional states should be avoided.
The flow behaviour of cohesive sand and rectangular blade was observed to have not one transition but four transitional states as the impeller speed was increased from 100 to 600 rpm (1.67 to 10 Hz). The system transitioned from bumping to apparent roping, back to bumping, then to apparent roping again and finally, to the roping regime.

The transition between the two regimes is governed by the dimensionless powder Froude number the ratio of powder centripetal acceleration to gravity given by:

\[ Fr_p = \frac{v^2}{gR_c} \]  

(4.4)

The transitional states within the bumping regime can be differentiated by the dimensionless Bed Resonance number given by:

\[ \beta = \frac{t_a}{t_b} \]  

(4.6)

The powder flow regime map was verified using PEPT and torque experiments. The interesting flow phenomena highlighted by the powder flow regime map has implications for industries, especially for granulation processes where the identification of the roping motion is essential for controlled agglomeration.

The three-dimensional flow behaviour of the powder flow regimes has been characterised for dry and cohesive sand with two impeller geometries using PEPT in Chapter 5. These experiments demonstrate the existence of different particle parameters (e.g. powder velocity, shear rates) in different locations within the mixer. With the addition of moisture (cohesion) to the dry powder, the average velocity of the powder decreases by 10%. Increasing blade-rake angle from 10 to 90 degrees increased the average powder velocity by approximately 38%. Vertical mixing improved with higher blade-rake angle due to an increased thickness of the shear transmission layer however no significant improvements were observed with changes in bed cohesion. PEPT has proved to be a powerful tool for studying flow behaviour below the powder surface without disturbing the flow. The detailed quantitative information obtained from PEPT can be used with DEM to validate current particulate flow models for better design of mixer granulators.

An understanding of the magnitude and location of these parameters can be used to develop multi-dimensional microscopic population balance models that reflect what is physically occurring in the mixer granulator (i.e. growth and breakage in the impeller zone, nucleation in the spray zone). While PEPT cannot directly measure collisional velocities critical for growth and breakage, this tool does provide a quantitative approach from which order of magnitude estimations are possible.

A general framework has been developed to generate compartment models of non-homogeneous
mixing behaviour in vertical mixers using PEPT data in Chapter 6. Flow characteristics obtained from these experiments were used to generate RTD for different regions in the mixer. These RTD provided the basis for fitted compartment models.

The two-compartment model consisted of an impeller and circulation zone. This model was further developed into a multi-compartment model with the addition of a surface zone to incorporate the key granulation process of wetting and nucleation. Combinations of perfectly mixed tanks, cascades of perfectly mixed tanks, short-cuts and recirculation were used to reproduce the non-ideal mixing observed in the PEPT experiments.

The multi-CM derived from PEPT experiments should prove to be a useful tool for the validation of DEM simulations. Further work to incorporate multi-CM with a general multi-dimensional population balance model shows potential to produce improved mixing performances and predictive granulation models.

7.2 Original contributions

This thesis has resulted in several original contributions. The most significant being the development of a powder flow regime map that demonstrated the relationship between powder Froude number and the newly proposed Bed Resonance number from empirical measurements in Chapter 4.

The identification and characterisation of the apparent roping regime for bladed vHSM in Section 4.2 has provided a new explanation for the mechanisms behind the intermediate regime reported by previous researchers.

A number of experiments were carried out using PEPT that directly measured powder velocities below the surface that have not been presented in the literature before. Also, the methodology developed for in depth data analysis of PEPT experiments are also new, specifically, the Fourier transfer analysis method and the two-dimensional shear rates estimations.

The use of existing compartmental models in combination with experimental PEPT data for vHSM in Chapter 6 as far as the author is aware, is also new.

7.3 Future work

Further research is required to elucidate the boundaries for a variety of formulation including more cohesive powders and mixer designs involving different impeller designs. A fundamental understanding of the physical interaction between the material properties and impeller blades at high speeds is also lacking, which would be useful for predictive models.
The future work in this area is concerned with extending the CM framework to include granulation kinetic information using population balance models. The combination of these two complementary models can provide valuable information on the detailed interaction between the powder flow and granulation processes. The resulting models would be a step closer towards an *a priori* model that predict the performance of granulation processes.
References


References


References

Discrete Element Methods The 4th International Conference on Discrete Element Methods, Brisbane, August 2007.


References


References


