

Copyright is owned by the Author of the thesis. Permission is given for a copy to be downloaded by an individual for the purpose of research and private study only. The thesis may not be reproduced elsewhere without the permission of the Author.

**AIR-SUSPENSION COATING OF DAIRY POWDERS:  
A MICRO-LEVEL PROCESS APPROACH**

A thesis presented in partial fulfilment of the requirements for the degree of  
Doctor of Philosophy  
in  
Technology  
at Massey University, Palmerston North, New Zealand

Stephen R L Werner

2005

## ABSTRACT

Air-suspension particle coating is a process by which thin coatings are applied to powder particles. The coatings can be formulated to act as permeable barriers to increase powder shelf-life or to impart controlled release character. The ultimate objective of a coating operation is to produce individual particles, each with a well-controlled, even coating.

This project was focused on the air-suspension coating of fine powders of  $\sim 100\ \mu\text{m}$  in diameter for the dairy industry. Despite the widespread use of the technology in the pharmaceutical industry, its use in the food industry has been limited. Little is known about the fundamental mechanisms, and so published work to date is product and equipment specific and is statistical in the way the experimental design and analysis has been approached. This 'black box' approach is time consuming and costly. Better methods based on an understanding of the physical and chemical mechanisms are needed to deal with the numerous products and constantly changing formulations typical of the dairy industry.

This thesis proposes a new approach to air-suspension particle coating research. The basis of this 'micro-level process approach', is to deconvolute the complex coating process into smaller manageable parts based on classical physical phenomena for which descriptions already exist. The thesis identifies and develops an understanding of the key micro-level processes controlling coated product quality and process performance. Four were selected for further study: drying, droplet impact and spreading, and stickiness which encompasses the two key micro-level processes of droplet impact and adherence and inter-particle agglomeration. They were studied separately to deconvolute the variable effects and interactions.

Kinetic data were collected for the drying droplets containing maltodextrins, whey protein isolate and gum arabic. A mathematical model, based on 'ideal shrinkage' was developed to predict the drying kinetics of single droplets with particular interest in the development of the surface glass transition temperature. The model accurately predicted the kinetics until significant morphological changes occurred in the droplet. To better predict the kinetics late in the drying process, the droplet radius was set to be constant at a time based on the surface proximity to the surface glass transition temperature (critical  $X$  concept). This was done to arrest droplet shrinkage in line with experimental observations and to more accurately depict the drying of high molecular weight, amorphous glass forming polymers. After this point, a new flexible calculation scheme was used to better predict the variation in internal droplet structure as either a dense, 'collapsed shell' structure or a 'dense skin-porous crumb' structure. Further study should focus on the surface and internal droplet structure (porosity and mechanical integrity) development during drying, particularly the conditions leading to the arresting of the droplet radius and the subsequent rate of skin thickness progression.

The critical  $X$  concept was used to make industrial-scale predictions of the optimum drying conditions that ensure maximum droplet impact and adherence efficiency and minimum inter-particle agglomeration in a Würster-style coating operation. This enabled the prediction of two key design parameters, the nozzle distance from the powder impact point and the Würster

insert height. The span in design parameters showed that there is significant opportunity for design optimisation based on the critical  $X$  concept.

A probe tack test was used to map the level of stickiness of droplets of different coating materials as they dried. As skin formation progressed, the stickiness passed through a maximum, in most cases to arrive at a point at which the droplet was no longer sticky at all (non-adhesive state). The maximum point of stickiness represents the ideal state to ensure successful droplet–substrate impact and adherence. The minimum point of stickiness represents the ideal state to prevent unwanted inter-particle agglomeration. The time interval between the onset of stickiness and the non-adhesive state was particularly dependent on the addition of plasticisers, but also on the formulation and the drying air conditions. Future work should look to establish a possible relationship between the surface glass transition temperature and the probe tack test stickiness measurements.

The impact and spreading of droplets containing maltodextrin DE5 on to solid anhydrous milkfat was studied using a high speed video camera. It was found that the final spread diameter was able to be fixed close to the maximum spread diameter by using surfactants, thus avoiding significant recoil. Because existing literature focuses on predicting the maximum spread diameter, this work defines a need for adequate prediction methods for the final spread diameter, as this is the significant parameter in coating applications.

Formulation and operating guidelines were established to independently optimise each micro-level process. These were used in a series of population based coating experiments in a pilot-scale Würster coater. This study highlighted the limited flexibility of the standard ‘off-the-shelf’ Würster coating apparatus for the coating of fine sized dairy powders. Because of this, the validation of the guidelines were inconclusive and optimisation could not be carried out. Further validation work is required on a custom-built apparatus for dairy powders.

This work has advanced the fundamental knowledge of the coating process and is independent of material, equipment and scale. This knowledge, based on physical and chemical mechanisms, can be used to develop coating formulations and identify optimum process conditions for successful coating in less time and at less expense than is current practice.

The next step is to put the guidelines into practice and craft the engineering of a continuous coating apparatus for dairy powder applications.

## ACKNOWLEDGEMENTS

I have always struggled to explain to people what I have been doing for the last three years (alright three and a half!). I almost have it now. A wise person once said, 'If it's green or wiggles it's biology, if it stinks it's chemistry, if it has numbers it's math, if it doesn't work it's technology'. I received a Bachelor in Technology (Chemical) so I know about stink and broken. I then decided to become an 'expert' in this field. Yet another wise, but not necessarily the same person defined an 'expert' as, 'someone who learns more and more about less and less, until he knows everything about nothing'. So there you have it. I am qualified for stink and broken, and what's more, I know heaps about not much. My future looks brite and there are a few people I must thank for helping me on to this bountiful path.

First, I thank my principal supervisor, Dr Jim Jones, for his endless energy for this project which kept me motivated. You gave me all the slack I needed to guide this work and subtly pushed me back on track when I strayed too far.

Thanks also to my secondary supervisor, Associate Professor Tony Paterson for his support 'on and off the field'. Always remember that winning one set off me at tennis does not constitute winning a match. Anyway, that day I had a head cold and the balls were not ATP standard pressure!

I am grateful to my industry supervisors, Professor Richard Archer and Dr David Pearce, for the financial support, entertaining project meetings and creative ideas.

Thanks to Dr Tony Howes and Dr Besh Bhandari at The University of Queensland for welcoming me to their departments during my three-month stay.

Dr John Bronlund has been my Yoda of mathematical modelling, and I would also like to thank him for the great chats on wine, cheese, million dollar ideas and introducing me to 'Acid Jazz'.

Many thanks must go to the unsung heroes in the Massey University and Fonterra, Palmerston North laboratories and workshops who have lent a hand. Particular mention has to go to Bruce Colins and Colin Knight for helping me with some 'not-so-standard requests!'

Thanks to all the postgrads that have come and gone during my time here: both the 'old school' postrads Ratty, Bogan, Gollum, Steam, Cheese, Kylie and Krishna, who departed many years ago but left a lasting impression, and the 'new school' postgrads Rachel, Craig, Adi, Petja, Mossop, Robert W, Jeremy, Antje and Anna. Thanks also to the foreign crowd for showing this boy a little bit of culture: Juan and Patty for the great Chilean barbeques and the Dutch contingent, Franz, Corine, Mariska, Hans, Mike and Sandfly for their papernotens and general all round drunken parties. I will see you kaaskops at Carnaval! Special thanks also to Sam and Lauren, a couple of great friends for their patience, support and timely sanity checks! Love you girls!

Thanks also to the Stubbings whanau who have been supportive over these and many years before. Finally, a special thank you to my mother, for her love, support, and food parcels over these years. I'm off for a haircut and a shave.

## TABLE OF CONTENTS

<b>ABSTRACT.....</b>	<b>iii</b>
<b>ACKNOWLEDGEMENTS.....</b>	<b>v</b>
<b>TABLE OF CONTENTS.....</b>	<b>vii</b>
<b>LIST OF FIGURES.....</b>	<b>xv</b>
<b>LIST OF TABLES.....</b>	<b>xxi</b>

### CHAPTER 1 THESIS OVERVIEW

<b>1.1</b>	<b>PROBLEM DEFINITION .....</b>	<b>1</b>
<b>1.2</b>	<b>PROPOSED SOLUTION.....</b>	<b>1</b>
<b>1.3</b>	<b>THESIS OBJECTIVES.....</b>	<b>1</b>
	1.3.1 Overall thesis objective .....	1
	1.3.2 Specific thesis objectives .....	2
<b>1.4</b>	<b>THESIS SYNOPSIS .....</b>	<b>2</b>
<b>1.5</b>	<b>THESIS SCOPE.....</b>	<b>4</b>

### CHAPTER 2 AIR-SUSPENSION PARTICLE COATING TECHNOLOGY – 'STATE OF THE ART'

<b>2.1</b>	<b>INTRODUCTION.....</b>	<b>5</b>
<b>2.2</b>	<b>OPERATING PRINCIPLE .....</b>	<b>5</b>
<b>2.3</b>	<b>INDUSTRIAL APPLICATIONS .....</b>	<b>6</b>
<b>2.4</b>	<b>COATING OBJECTIVES .....</b>	<b>7</b>
	2.4.1 Ultimate objective.....	7
	2.4.2 Product quality objectives .....	7
	2.4.3 Coater performance objectives .....	7
<b>2.5</b>	<b>COATING MATERIALS .....</b>	<b>8</b>
<b>2.6</b>	<b>COATING EQUIPMENT.....</b>	<b>8</b>
<b>2.7</b>	<b>KEY PROCESS VARIABLES.....</b>	<b>9</b>
<b>2.8</b>	<b>COATING QUALITY .....</b>	<b>11</b>
	2.8.1 Coating mass uniformity.....	11
	2.8.2 Coating morphology.....	12
<b>2.9</b>	<b>MODELLING THE COATING PROCESS.....</b>	<b>16</b>
<b>2.10</b>	<b>CURRENT 'STATE OF THE ART' .....</b>	<b>16</b>
	2.10.1 Direction of past researchers .....	16
	2.10.2 The development and optimisation process.....	17
	2.10.3 Application in the dairy industry .....	18
<b>2.11</b>	<b>CONCLUSIONS AND RECOMMENDATIONS .....</b>	<b>18</b>

## CHAPTER 3 'MICRO-LEVEL PROCESS APPROACH' TO AIR-SUSPENSION PARTICLE COATING RESEARCH

3.1	INTRODUCTION .....	21
3.2	PROPOSED NEW APPROACH TO COATING RESEARCH BASED ON EXAMINATION OF MICRO-LEVEL PROCESSES .....	21
3.3	MICRO-LEVEL PROCESS IDENTIFICATION .....	22
3.4	PARTICLE MOTION.....	24
3.5	ATOMISATION.....	25
3.6	DROPLET-PARTICLE COLLISION .....	26
3.7	DROPLET IMPACT AND ADHERENCE.....	27
3.8	DROPLET IMPACT AND SPREADING .....	28
3.9	INFILTRATION .....	30
3.10	DRYING.....	32
3.11	FILM FORMATION.....	34
3.12	LAYERING .....	35
3.13	INTER-PARTICLE AGGLOMERATION.....	36
3.14	CONCLUSIONS AND RECOMMENDATIONS .....	39

## CHAPTER 4 MATERIALS CHARACTERISATION AND METHODS

4.1	INTRODUCTION .....	41
4.2	MATERIALS SELECTION .....	41
4.2.1	Introduction.....	41
4.2.2	Potential applications.....	41
4.2.3	Maltodextrins and corn syrup solids .....	42
4.2.4	Gum arabic.....	43
4.2.5	Whey protein isolate .....	44
4.2.6	Plasticisers .....	46
4.2.7	Surfactants/Wetting agents .....	46
4.2.8	Anti-tack agents.....	47
4.2.9	Conclusions.....	47
4.3	OXYGEN DEPLETION STUDIES.....	48
4.4	BINARY MOISTURE DIFFUSION COEFFICIENT .....	49
4.5	MOISTURE SORPTION ISOTHERMS .....	50
4.6	GLASS TRANSITION TEMPERATURE .....	53
4.7	SOLUTION VISCOSITY .....	55
4.8	SOLUTION SURFACE TENSION.....	56
4.9	DROPLET-SURFACE CONTACT ANGLE .....	57
4.10	PARTICLE SIZE ANALYSIS.....	58
4.11	COATING MASS.....	59
4.12	COATING MORPHOLOGY .....	60
4.13	CONCLUSIONS AND RECOMMENDATIONS .....	60

## CHAPTER 5 SINGLE DROPLET DRYING KINETICS AND MODELLING

<b>5.1</b>	<b>INTRODUCTION</b> .....	<b>61</b>
<b>5.2</b>	<b>CONCEPTUAL MODELS OF THE DROPLET DRYING PROCESS</b> .....	<b>61</b>
<b>5.3</b>	<b>CONTROLLING DRYING MECHANISMS</b> .....	<b>66</b>
<b>5.4</b>	<b>MATHEMATICAL MODEL SUMMARY</b> .....	<b>69</b>
	5.4.1 Assumptions .....	70
	5.4.2 Summary of key equations.....	70
	5.4.3 Numerical solution .....	71
<b>5.5</b>	<b>EXPERIMENTAL METHOD SELECTION</b> .....	<b>72</b>
<b>5.6</b>	<b>EXPERIMENTAL SET-UP</b> .....	<b>72</b>
	5.6.1 Set-up overview .....	72
	5.6.2 Air delivery and heating system .....	73
	5.6.3 Main drying chamber .....	73
	5.6.4 Data and image acquisition system .....	74
	5.6.5 Moisture content history monitoring system.....	74
	5.6.6 Temperature history monitoring system.....	74
<b>5.7</b>	<b>EXPERIMENTAL PROTOCOL</b> .....	<b>75</b>
	5.7.1 Key variable and level selection.....	75
	5.7.2 Solution preparation .....	75
	5.7.3 Moisture content and temperature history data collection.....	75
<b>5.8</b>	<b>PRELIMINARY RIG TESTING</b> .....	<b>76</b>
	5.8.1 Air velocity.....	76
	5.8.2 Relative humidity.....	76
	5.8.3 Temperature .....	76
	5.8.4 Droplet suspension system.....	77
	5.8.5 Prediction of the drying of a pure water droplet.....	78
	5.8.6 Validation of Ranz and Marshall's (1952) correlations for Nusselt and Sherwood numbers.....	79
<b>5.9</b>	<b>DRYING KINETICS AND MODELLING OF MALTODEXTRIN DE5</b> .....	<b>82</b>
	5.9.1 Initial solids concentration and air temperature.....	82
	5.9.2 Air velocity.....	85
	5.9.3 Initial droplet size .....	87
	5.9.4 Effect of wetting agent .....	88
	5.9.5 Relative humidity.....	89
	5.9.6 Deficiencies in the model .....	90
	5.9.7 Conclusions .....	92
<b>5.10</b>	<b>'HYBRID DROPLET DRYING MODEL'</b> .....	<b>92</b>
<b>5.11</b>	<b>INDUSTRIAL-SCALE PREDICTIONS</b> .....	<b>101</b>
<b>5.12</b>	<b>DRYING KINETICS OF MALTODEXTRINS DE5, DE10 AND DE18, WHEY PROTEIN ISOLATE AND GUM ARABIC</b> .....	<b>107</b>
	5.12.1 Drying kinetics at 20% w/w solids and 40 °C .....	108
	5.12.2 Drying kinetics at 20% w/w solids and 78 °C .....	109
	5.12.3 Drying kinetics at 40% w/w solids and 40 °C .....	110
	5.12.4 Drying kinetics at 40% w/w solids and 78 °C .....	111
	5.12.5 Conclusions .....	111
<b>5.13</b>	<b>PLASTICISATION OF MALTODEXTRIN DE5 AND WHEY PROTEIN ISOLATE</b> .....	<b>112</b>
	5.13.1 Maltodextrin DE5 plasticisation .....	112

5.13.2	Whey protein isolate plasticisation .....	114
5.13.3	Conclusions.....	115
<b>5.14</b>	<b>MORPHOLOGICAL DEVELOPMENT .....</b>	<b>116</b>
<b>5.15</b>	<b>CONCLUSIONS AND RECOMMENDATIONS .....</b>	<b>121</b>

## **CHAPTER 6 DEVELOPMENT OF DROPLET STICKINESS DURING DRYING**

<b>6.1</b>	<b>INTRODUCTION .....</b>	<b>123</b>
<b>6.2</b>	<b>SKIN FORMATION.....</b>	<b>123</b>
6.2.1	Skin formation mechanisms .....	123
6.2.2	Controlling skin formation.....	124
6.2.3	Conclusions.....	125
<b>6.3</b>	<b>STICKINESS .....</b>	<b>125</b>
6.3.1	Defining stickiness.....	125
6.3.2	Stickiness theory and characterisation.....	125
6.3.3	Conclusions.....	128
<b>6.4</b>	<b>EXPERIMENTAL METHOD SELECTION.....</b>	<b>128</b>
6.4.1	Glass transition temperature .....	129
6.4.2	Sticky-point temperature and the critical X concept.....	130
6.4.3	Probe tack test .....	131
6.4.4	Relating probe tack measurements to glass transition temperature.....	133
<b>6.5</b>	<b>EXPERIMENTAL SET-UP.....</b>	<b>134</b>
6.5.1	Set-up overview.....	134
6.5.2	Actuator assembly with droplet holder.....	135
6.5.3	Probe.....	135
6.5.4	Moisture content history monitoring system .....	136
6.5.5	Temperature history monitoring system .....	137
<b>6.6</b>	<b>EXPERIMENTAL PROTOCOL .....</b>	<b>137</b>
6.6.1	Key variable and level selection .....	137
6.6.2	Stickiness tests.....	137
6.6.3	Moisture content and temperature history data collection .....	138
<b>6.7</b>	<b>PRELIMINARY RIG TESTING .....</b>	<b>138</b>
6.7.1	Drying air conditions.....	138
6.7.2	Droplet suspension system .....	138
6.7.3	Drying kinetics .....	138
6.7.4	Standardisation of probe-associated variables.....	140
6.7.5	Modes of failure.....	143
6.7.6	Reproducibility of stickiness measurement.....	146
6.7.7	Interpretation of results.....	146
6.7.8	Conclusions.....	146
<b>6.8</b>	<b>STICKINESS OF MALTODEXTRINS.....</b>	<b>147</b>
6.8.1	Maltodextrin DE18.....	147
6.8.2	Maltodextrin DE10.....	148
6.8.3	Maltodextrin DE5.....	149
6.8.4	Discussion.....	149
6.8.5	Conclusions.....	151
<b>6.9</b>	<b>RELATIONSHIP BETWEEN PROBE TACK TEST STICKINESS AND SURFACE GLASS TRANSITION TEMPERATURE .....</b>	<b>151</b>
<b>6.10</b>	<b>STICKINESS OF WHEY PROTEIN ISOLATE .....</b>	<b>152</b>

6.11	STICKINESS OF GUM ARABIC.....	153
6.12	PLASTICISATION OF MALTODEXTRIN DE5 AND WHEY PROTEIN ISOLATE.....	154
6.13	CONCLUSIONS AND RECOMMENDATIONS.....	157

## CHAPTER 7 DROPLET IMPACT AND SPREADING

7.1	INTRODUCTION.....	159
7.2	DROPLET IMPACT AND SPREADING.....	159
7.2.1	Phenomenological explanation of droplet impact and spreading ....	159
7.2.2	Prediction of droplet impact and spreading.....	161
7.2.3	Simplified droplet impact and spreading model.....	162
7.2.4	Controlling droplet impact and spreading.....	163
7.3	METHOD SELECTION.....	165
7.4	EXPERIMENTAL SET-UP.....	165
7.4.1	Set-up overview.....	165
7.4.2	Droplet production and impact velocity determination.....	166
7.4.3	Image acquisition and analysis.....	167
7.5	EXPERIMENTAL PROTOCOL.....	168
7.5.1	Key variable and level selection.....	168
7.5.2	Solution preparation.....	169
7.6	PLATE PREPARATION AND PRELIMINARY TESTING ON AN ANHYDROUS MILKFAT SURFACE.....	171
7.6.1	Test surface selection.....	171
7.6.2	Anhydrous milkfat plate preparation.....	171
7.6.3	Identification of error sources and statistical significance of the data.....	174
7.7	DYNAMICS OF DROPLET SPREADING.....	176
7.8	DROPLET FORMULATION EFFECTS.....	178
7.8.1	Inertia and viscosity effects.....	178
7.8.2	Surface tension effects.....	182
7.8.3	Surface tension and inertia effects.....	190
7.8.4	Guar gum addition.....	193
7.8.5	Xanthan gum addition.....	195
7.9	SUBSTRATE EFFECTS.....	196
7.9.1	Pre-coating with surfactant.....	196
7.9.2	Roughness and porosity effects.....	199
7.10	CONCLUSIONS AND RECOMMENDATIONS.....	201

## CHAPTER 8 KEY PERFORMANCE GUIDELINES, VALIDATION AND INDUSTRIAL PROCESSING

8.1	INTRODUCTION.....	205
8.2	SUMMARY OF IDEAL COATING CONDITIONS.....	205
8.2.1	Droplet drying and droplet stickiness.....	205
8.2.2	Droplet impact and spreading.....	206
8.3	PILOT-SCALE COATING TRIALS.....	207
8.3.1	Scope.....	207
8.3.2	Experimental set-up.....	208

8.3.3	Coating equipment limitations .....	210
8.3.4	Spray droplet adhesion efficiency trials .....	211
8.3.5	Coating morphology .....	215
8.3.6	Conclusions .....	218
<b>8.4</b>	<b>RECOMMENDATIONS FOR INDUSTRIAL-SCALE COATING.....</b>	<b>218</b>
8.4.1	Product development.....	218
8.4.2	Process development.....	219
8.4.3	Industrial processing scenarios .....	219
<b>8.5</b>	<b>CONCLUSIONS AND RECOMMENDATIONS .....</b>	<b>221</b>

## **CHAPTER 9 CONCLUSIONS AND SUGGESTIONS FOR FUTURE WORK**

<b>9.1</b>	<b>CONCLUSIONS .....</b>	<b>223</b>
<b>9.2</b>	<b>SUGGESTED FUTURE WORK .....</b>	<b>225</b>

<b>REFERENCES.....</b>	<b>227</b>
------------------------	------------

<b>APPENDIX A1 NOMENCLATURE.....</b>	<b>A1</b>
--------------------------------------	-----------

## **APPENDIX A2 SINGLE DROPLET DRYING – MATHEMATICAL MODEL DEVELOPMENT, SOLUTION AND INITIAL VALIDATION**

<b>A2.1</b>	<b>INTRODUCTION .....</b>	<b>A9</b>
<b>A2.2</b>	<b>CONCEPTUAL MODEL DEVELOPMENT .....</b>	<b>A9</b>
A2.2.1	Conceptual model .....	A9
A2.2.2	Assumptions.....	A11
A2.2.3	Validity of the assumptions.....	A11
<b>A2.3</b>	<b>MATHEMATICAL FORMULATION.....</b>	<b>A14</b>
A2.3.1	Moisture history .....	A14
A2.3.2	Temperature history .....	A15
A2.3.3	Droplet velocity.....	A16
A2.3.4	Thermophysical properties of air .....	A16
A2.3.5	Thermophysical properties of water .....	A18
A2.3.6	Thermophysical and chemical properties of polymers.....	A19
A2.3.7	Droplet mixture properties .....	A23
A2.3.8	Gas side heat and mass transfer coefficients.....	A25
<b>A2.4</b>	<b>IMPLEMENTATION OF THE DROPLET DRYING MODEL.....</b>	<b>A28</b>
A2.4.1	Numerical solution type and computer program.....	A28
A2.4.2	Definition of finite difference grid .....	A29
A2.4.3	Physical properties .....	A30
A2.4.4	Initial conditions.....	A30
A2.4.5	Boundary conditions.....	A31
A2.4.6	Internal nodes ( $2 < j < J-1$ ).....	A36
A2.4.7	Central boundary node ( $j = 1$ ).....	A37
A2.4.8	Surface boundary node ( $j = J$ ).....	A38
A2.4.9	Numerical solution procedure.....	A39

---

<b>A2.5</b>	<b>MATHEMATICAL MODEL EVALUATION .....</b>	<b>A40</b>
A2.5.1	Maths checking .....	A40
A2.5.2	Internal and external heat transfer side numerical check with analytical solution.....	A40
A2.5.3	Internal moisture diffusion side numerical check with analytical solution.....	A42
A2.5.4	External mass transfer and volume contraction check with analytical solution.....	A44
A2.5.5	Numerical error checking .....	A45
A2.5.6	Maths checking summary .....	A46
A2.5.7	Mathematical model predictions against experimental drying kinetics and sensitivity analysis.....	A47
<b>A2.6</b>	<b>CONCLUSIONS .....</b>	<b>A50</b>
 <b>APPENDIX A3 PROGRAM SOURCE CODE</b>		
<b>A3.1</b>	<b>NOTES FOR USERS.....</b>	<b>A51</b>

## LIST OF FIGURES

Figure 2.1:	Two particle growth mechanisms (inter-particle agglomeration and surface layering) in an air-suspension particle operation .....	6
Figure 2.2:	Schematic drawing of a typical laboratory-scale Würster coater showing auxiliary equipment and typical dimensions .....	9
Figure 2.3:	The complex effect of atomisation pressure applied in pneumatic nozzles on coating efficiency .....	10
Figure 2.4:	Cross-section of coated particles depicting variation in coating mass uniformity. ....	11
Figure 2.5:	Cross-section of coated particles depicting variation in coating morphology.. ..	12
Figure 2.6:	Fractures in coating surfaces. ....	13
Figure 2.7:	Evidence of droplet spreading and coalescence on a PVP-coated particle. ...	14
Figure 2.8:	Various degrees of spreading and coalescence on coated particles. ....	14
Figure 2.9:	Evidence of porosity in a 20% w/w aqueous solution of sodium chloride. ....	15
Figure 2.10:	Evidence of different morphologies during the drying of lactose at different air temperatures. ....	15
Figure 3.1:	Cross-section of a Würster coater showing regions and phenomena occurring during particle coating. ....	22
Figure 3.2:	Geldart (1973) powder classification diagram for fluidisation. ....	25
Figure 3.3:	Droplet-particle collision mechanisms. ....	27
Figure 3.4:	Possible outcomes of the impact of a droplet on a flat, dry surface in relation to air-suspension particle coating applications. ....	29
Figure 3.5:	Spreading and infiltration of a coating droplet on a porous, wettable particle surface. ....	31
Figure 3.6:	Film formation mechanisms from solution. ....	35
Figure 3.7:	Regimes of granulation. ....	38
Figure 4.1:	Oxygen consumption of a natural cheddar cheese flavoured and a natural butter flavoured spray-dried powder produced at Fonterra, Longburn. ....	49
Figure 4.2:	Predicted binary moisture diffusion coefficient for maltodextrin DE5 as a function of moisture content and temperature. ....	50
Figure 4.3:	Moisture sorption isotherms for potential coating materials at 30 °C. ....	51
Figure 4.4:	Comparison of measured maltodextrin isotherms and literature data. ....	52
Figure 4.5:	Selected thermograms from the DSC showing the variability in the temperature range over which the glass transition occurs in the maltodextrins at different water activities. ....	54
Figure 4.6:	Glass transition temperature profiles for maltodextrin DE5, DE10, DE15 and DE20 from Roos and Karel (1991a) and data collected in this study. ....	55
Figure 4.7:	Particle size distribution of an uncoated and 20% w/w maltodextrin DE5 coated glass ballotini® sample and a 20% w/w maltodextrin DE5 spray droplet size distribution at 2 bar atomisation air pressure and 3.5 g.min <sup>-1</sup> coating solution flowrate. ....	59
Figure 5.1:	5 stage drying concept proposed by Dolinsky and Ivanicki (1984). ....	62

Figure 5.2:	Conceptual descriptions for droplet spray drying that the two major mathematical model type categories (receding interface and effective diffusion models) are based upon.....	65
Figure 5.3:	Scanning electron micrographs of spray dried droplets depicting complex morphological features. ....	66
Figure 5.4:	Schematic diagram of the single droplet drying rig. ....	73
Figure 5.5:	Experimental mass and temperature histories along with model predictions for a 6 $\mu\text{l}$ pure water droplet drying at 40 $^{\circ}\text{C}$ and 0.30 $\text{m}\cdot\text{s}^{-1}$ .....	78
Figure 5.6:	Comparison of experimental $Nu$ and $Sh$ numbers with those obtained from the Ranz and Marshall (1952a) correlations.....	81
Figure 5.7:	Effect of drying air temperature on the moisture content and temperature histories of 20% w/w maltodextrin DE5 droplets along with model predictions.....	83
Figure 5.8:	Effect of drying air temperature on the moisture content and temperature histories of 40% w/w maltodextrin DE5 droplets along with model predictions.....	83
Figure 5.9:	Effect of the drying air velocity on the moisture content and temperature histories of 20% w/w maltodextrin DE5 droplets drying at 40 $^{\circ}\text{C}$ along with model predictions. ....	86
Figure 5.10:	Effect of the drying air velocity on the moisture content and temperature histories of 40% w/w maltodextrin DE5 droplets drying at 78 $^{\circ}\text{C}$ along with model predictions. ....	86
Figure 5.11:	Effect of initial droplet size on the moisture content and temperature histories of 40% w/w maltodextrin DE5 droplets drying at 78 $^{\circ}\text{C}$ and 0.30 $\text{m}\cdot\text{s}^{-1}$ along with model predictions. ....	88
Figure 5.12:	Effect of critical $X$ value and the variable density calculation schemes on the droplet temperature on the moisture content predictions for 40% w/w maltodextrin DE5 droplets at 78 $^{\circ}\text{C}$ . ....	94
Figure 5.13:	Possible internal structures during droplet drying.....	96
Figure 5.14:	‘Ideal shrinkage’ structure model: (a) internal node radii and (b) binary moisture diffusion coefficient as a function of these radii for a 40% w/w maltodextrin DE5 droplet drying at 78 $^{\circ}\text{C}$ .....	97
Figure 5.15:	‘Collapsed shell’ structure model: (a) internal node radii and (b) binary moisture diffusion coefficient as a function of these radii for a 40% w/w maltodextrin DE5 droplet drying at 78 $^{\circ}\text{C}$ for a critical $X = (T - T_g) = 20$ $^{\circ}\text{C}$ ..	97
Figure 5.16:	‘Dense skin–porous crumb’ structure model: (a) internal node radii and (b) binary moisture diffusion coefficient as a function of these radii for a 40% w/w maltodextrin DE5 droplet drying at 78 $^{\circ}\text{C}$ for a critical $X = (T - T_g) = 20$ $^{\circ}\text{C}$ .....	97
Figure 5.17:	Porosity development in a 40% w/w maltodextrin DE5 droplet drying at 78 $^{\circ}\text{C}$ predicted by the fixed-radius, $X = (T - T_g) = 20$ $^{\circ}\text{C}$ model (‘dense skin–porous crumb’ structure).....	100
Figure 5.18:	Micro-level examination of the key control variables in a Würster coater. .	102
Figure 5.19:	Predicted moisture content and temperature distribution in an industrial-scale spray droplet under slow drying conditions.....	103
Figure 5.20:	Predicted moisture content and temperature distribution in an industrial-scale spray droplet under fast drying conditions. ....	103
Figure 5.21:	Droplet velocity profile for the slow and fast drying scenario cases. ....	104
Figure 5.22:	Droplet distance profile for the slow and fast drying scenario cases. ....	105

Figure 5.23:	‘Slow drying’ (Table 5.2) prediction of moisture content and temperature distribution in an industrial-scale spray droplet. ....	106
Figure 5.24:	‘Fast drying’ (Table 5.2) prediction of moisture content and temperature distribution in an industrial-scale spray droplet. ....	106
Figure 5.25:	Effect of polymer type on the drying kinetics for 20% w/w initial solids concentration droplets drying at 40 °C. ....	108
Figure 5.26:	Effect of polymer type on the drying kinetics for 20% w/w initial solids concentration droplets drying at 78 °C. ....	109
Figure 5.27:	Effect of polymer type on the drying kinetics for 40% w/w initial solids concentration (30% w/w for gum arabic) droplets drying at 40 °C. ....	110
Figure 5.28:	Effect of polymer type on the drying kinetics for 40% w/w initial solids concentration (30% w/w for gum arabic) droplets drying at 78 °C. ....	111
Figure 5.29:	Effect of plasticisation with glycerol and maltodextrin DE18 on maltodextrin DE5 droplet drying kinetics at the 20% w/w initial solids concentration. ...	113
Figure 5.30:	Effect of plasticisation with glycerol and maltodextrin DE18 on maltodextrin DE5 droplet drying kinetics at the 40% w/w initial solids concentration. ...	114
Figure 5.31:	Effect of plasticisation with glycerol and maltodextrin DE 18 on whey protein isolate droplet drying kinetics at the 40% w/w initial solids concentration. ...	115
Figure 5.32:	Morphological development in selected drying droplets. ....	117
Figure 5.33:	Morphological development in selected drying droplets from the plasticisation trials. ....	120
Figure 6.1:	Interfacial tensions around a liquid droplet on a flat, solid surface. ....	126
Figure 6.2:	Example of a state diagram showing the glass transition temperature and assumed sticky-point temperature for maltodextrin DE5. ....	130
Figure 6.3:	Schematic of a simple probe tack tester. ....	132
Figure 6.4:	Front-end (air outlet) photograph of the single droplet drying chamber along with probe tack test device. ....	134
Figure 6.5:	Schematic of the stickiness probe and droplet holder. ....	136
Figure 6.6:	Reproducibility of mass history data for 100 µl 40% w/w maltodextrin DE5 droplets drying at 78 °C. ....	139
Figure 6.7:	Moisture content and temperature histories of a 100 µl 20% w/w maltodextrin DE5 droplet drying at 78 °C. ....	139
Figure 6.8:	Comparison of moisture content history data for 100 µl 40% w/w maltodextrin DE5, DE10 and DE18 droplets drying at 78 °C. ....	140
Figure 6.9:	Probe tack test pressure curve for a 100 µl pure water droplet at $24 \pm 0.5$ °C and a probe speed of $30 \text{ mm} \cdot \text{min}^{-1}$ . ....	141
Figure 6.10:	Bonding and debonding behaviour identified from the probe tack measurements for the drying of droplets that form an amorphous skin. ....	145
Figure 6.11:	Stickiness of 100 µl maltodextrin DE18 droplets during drying. ....	147
Figure 6.12:	Stickiness of 100 µl maltodextrin DE10 droplets during drying. ....	148
Figure 6.13:	Stickiness of 100 µl maltodextrin DE5 droplets during drying. ....	149
Figure 6.14:	Tensile pressure as a function of maltodextrin viscosity. ....	150
Figure 6.15:	Stickiness of 100 µl whey protein isolate droplets during drying. ....	153
Figure 6.16:	Stickiness of 100 µl gum arabic droplets during drying. ....	154
Figure 6.17:	Effect of plasticiser addition on the stickiness of 100 µl maltodextrin DE5 droplets during drying. ....	155
Figure 6.18:	Effect of plasticiser addition on the stickiness of 100 µl whey protein isolate droplets during drying. ....	156
Figure 7.1:	Schematic of the impact of a droplet on a smooth, flat surface. ....	159

Figure 7.2:	Evolution of the spread factor for a water droplet of diameter 3.4 mm impinging at $1.5 \text{ m.s}^{-1}$ on to a flat, dry, glass surface. ....	160
Figure 7.3:	Maximum spread factor predicted by the Pasandideh-Fard <i>et al.</i> (1996) model for industrial and experimental conditions on to an anhydrous milkfat surface. ....	164
Figure 7.4:	Photograph of the experimental set-up used for the droplet impact and spreading investigations.....	166
Figure 7.5:	Reynolds and Weber numbers under experimental and industrial-scale conditions.....	169
Figure 7.6:	Anhydrous milkfat surface morphologies under various preparation procedures.....	172
Figure 7.7:	Time series of events depicting the impact, spreading and recoil of various water and maltodextrin DE5 droplet formulations on anhydrous milkfat... ..	177
Figure 7.8:	Concentration (viscosity) and inertia (velocity) effects on the final spread factor for water and maltodextrin DE5 droplets impacting on to AMF. ....	179
Figure 7.9:	Evolution of the spread factor (up to 50 ms) as a function of concentration (viscosity) for droplets impacting at $2.6 \text{ m.s}^{-1}$ on to AMF.....	180
Figure 7.10:	Evolution of the spread factor (up to 500 ms) as a function of concentration (viscosity) for droplets impacting at $2.6 \text{ m.s}^{-1}$ on to AMF.....	180
Figure 7.11:	$\zeta_{max}$ and $\zeta_{end}$ as a function of viscosity and initial impact velocity for water droplets and droplets containing 20, 30 and 40% w/w maltodextrin DE5 impacting on to AMF. ....	181
Figure 7.12:	Effect of surfactant addition to a droplet formulation on the final spread factor for droplets impacting at $2.6 \text{ m.s}^{-1}$ on to AMF. ....	185
Figure 7.13:	Final spread factor as a function of surface tension for droplets impacting at $2.6 \text{ m.s}^{-1}$ on to AMF.....	186
Figure 7.14:	Evolution of the spread factor (up to 50 ms) for 20% w/w maltodextrin DE5 droplets containing surfactants impacting at $2.6 \text{ m.s}^{-1}$ on to AMF.....	187
Figure 7.15:	Evolution of the spread factor (up to 500 ms) for 20% w/w maltodextrin DE5 droplets containing surfactants impacting at $2.6 \text{ m.s}^{-1}$ on to AMF.....	187
Figure 7.16:	Surfactant mass transfer mechanisms during droplet expansion. ....	188
Figure 7.17:	Effect of inertia (velocity) on the final spread factor (up to 10 ms) for 20% w/w maltodextrin DE5 droplets with and without 2% w/w UP impacting on to AMF.....	191
Figure 7.18:	Effect of inertia (velocity) on the final spread factor (up to 500 ms) for water and 20% w/w maltodextrin DE5 droplets with and without 2% w/w UP impacting onto AMF. ....	191
Figure 7.19:	Hypothesised surfactant maldistribution on a droplet surface and the appearance of Marangoni stresses.....	192
Figure 7.20:	Evolution of the spread factor for droplets containing guar gum impacting at $2.6 \text{ m.s}^{-1}$ on to AMF.....	194
Figure 7.21:	Evolution of the spread factor for droplets containing xanthan gum impacting at $2.6 \text{ m.s}^{-1}$ on to AMF. ....	195
Figure 7.22:	Comparison between lecithination of the AMF surface with lecithin incorporation into the droplet on the final spread factor for droplets impacting at $2.6 \text{ m.s}^{-1}$ . ....	197
Figure 7.23:	Evolution of the spread factor for 2.8 mm initial diameter water and 20% w/w maltodextrin DE5 droplets containing 2% w/w lecithins impacting at $2.6 \text{ m.s}^{-1}$ on to lecithinated AMF.....	199

Figure 7.24:	Effect of surface hydrophobicity, roughness and porosity on the final spread factor of 2.8 mm initial diameter 20% w/w maltodextrin DE5 droplets impacting at $2.6 \text{ m.s}^{-1}$ .....	200
Figure 8.1:	Summary of key findings for control of droplet stickiness.....	206
Figure 8.2:	Summary of key findings for control of droplet impact and spreading.....	207
Figure 8.3:	Photograph of the Glatt® Würster coater and auxiliary equipment used for the pilot-scale coating trials.....	208
Figure 8.4:	Schematic of the Glatt® Würster coater drying chamber showing the Würster insert and the position of various sensors.....	209
Figure 8.5:	Typical temperature and relative humidity histories for a coating trial.....	210
Figure 8.6:	Evidence of coating layer growth (increasing colour intensity) with increasing coating time.....	212
Figure 8.7:	Coating mass gain of ballotini® as a function of the mass of dry solids sprayed into the system for 20% w/w and 40% w/w maltodextrin DE5 solutions at air temperatures of 55, 70 and 85 °C.....	213
Figure 8.8:	Mass gain of ballotini® for various formulations for 20% w/w and 40% w/w initial solids concentration formulations drying at 70 °C.....	214
Figure 8.9:	Scanning electron micrographs depicting the droplet impact and spreading and coating layering micro-level processes for a 20% w/w maltodextrin DE5 formulation drying at 70 °C.....	215
Figure 8.10:	Scanning electron micrograph magnification series of coating morphological features for various coating formulations drying at 70 °C.....	217
Figure 8.11:	Envisaged custom-built multicell continuous powder coater for dairy industry applications.....	220
Figure A2.1:	Physical phenomena for modelling during droplet drying.....	A10
Figure A2.2:	Schematic of conceptual droplet drying model including experimental set-up equipment.....	A10
Figure A2.3:	Finite difference grid for droplet drying model.....	A29
Figure A2.4:	Finite difference grid for an internal node calculation ( $2 < j < J-1$ ).....	A36
Figure A2.5:	Finite difference grid for a central node calculation ( $j = 1$ ).....	A37
Figure A2.6:	Finite difference grid for the surface node calculation ( $j = J$ ).....	A38
Figure A2.7:	Program flow chart for single droplet drying mathematical model predictions.....	A39
Figure A2.8:	Comparison of the numerical model predictions with the analytical solution for the centre, mass average and surface temperatures of a 2.4 mm diameter droplet of 40% w/w maltodextrin DE5 drying under a heat transfer third-type boundary condition.....	A42
Figure A2.9:	Comparison of the numerical model predictions with the analytical solution for the centre, mass average and surface temperatures of a 2.4 mm diameter droplet of 40% w/w maltodextrin DE5 drying under a mass transfer first-type boundary condition.....	A44
Figure A2.10:	Comparison of the numerical model predictions with the analytical solution for the droplet radius.....	A45
Figure A2.11:	Moisture content and temperature histories of 40% w/w maltodextrin DE5 droplet along with model predictions.....	A47
Figure A2.12:	Moisture content and temperature histories of 40% w/w maltodextrin DE5 droplet along with model predictions.....	A49
Figure A2.13:	Heat and mass transfer Biot numbers for a 40% w/w maltodextrin DE5 droplet drying at 78 °C.....	A50



---

## LIST OF TABLES

Table 2.1:	Key variables for air-suspension particle coating .....	10
Table 5.1:	Heat transfer and mass transfer Biot numbers and Lewis number under various drying conditions .....	69
Table 5.2:	Parameters used in the industrial-scale drying kinetics predictions.....	102
Table 7.1:	Viscosity and surface tension of solutions.....	170
Table A2.1:	Average and maximum percentage errors in moisture content and temperature predictions as a function of the number of nodes .....	A46
Table A2.2:	System input values used in the model sensitivity analysis .....	A48