Granulation of Whole Milk Powder

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Abstract

High-shear granulation is an attractive alternative to spray drying for producing dried milk products. The capital cost of a granulation circuit is likely to be much less than a spray drying circuit which will reduce the manufacturing costs of milk powders. This work investigated the high-shear granulation of milk powder using milk concentrate as a binding agent in order to determine the feasibility of granulation as an alternative to, or and improvement on, the spray drying process. This research has laid the groundwork for further investigation into milk granulation by defining the conditions for which granulation is achieved and describing the effects of processing parameters on granulation for a pilot-scale mixer granulator. The technical feasibility of granulation is shown by proving that granulation does not affect the quality of the milk. Designs for perceived continuous granulation circuits are included to aid in further milk granulation research.

Successful granulation occurs at a total moisture content of approximately 11 % (±1 %). This was found to be suitable using either reconstituted or evaporated milk concentrated binder at between 20 and 50 % total solids. The time of granulation affects the size distribution of the granules and the granule yield at the end of the process. A narrower size distribution with increasing granule sizes and a reduction in the granule yield is observed for longer granulation times.

Granules were found to have better handling qualities than spray dried milk powders. Granules performed better in many functional tests having a higher bulk density, less change in bulk density during handling, better flowability and less fines. Granulation does not affect the chemical quality of the milk providing the granules are dried immediately after granulation. However, it was found that extended exposure of dried milk solids to a moisture content of 11 % results in an unacceptable amount of insoluble material forming. Granules are well suited as a product for reconstitution but did not perform adequately in wettability tests, suggesting that their use as an instantised product would require further study and improvement.

Further research is required to understand the role of lactose crystallisation and the generation of insoluble material to ensure scaling up of granulation will be successful. An investigation into continuous granulation would be useful for further milk granulation work.

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Chapter 1 – Introduction

1.1 Background

The New Zealand dairy industry produces over 1.2 million tonnes of powders manufactured from milk each year (NZMP, 2001). Powders have low transport cost and can be stored for an extended time due to their low water activity. The handleability and flowability are important physical properties for powders as well as the ease of reconstitution when being used by the consumer. These are influenced by the composition of the powder, the particle size distribution and the surface area.

Dairy powders are almost universally produced by spray drying (Caric, 1994) due to the short drying time and the adaptability of the equipment to manufacture different products. The disadvantages are the high capital costs of the spray drying equipment, the high-energy load required for the evaporation and the generation of fines. This necessitates further handling and recycling during manufacture, as well as an increased possibility of product loss during packaging and end-use of the product.

This project looks at granulation as either an alternative or an improvement to the spray drying process for producing whole milk powder. Granulation is a method used to improve the physical properties of powders by increasing the size of the powder particles (Hounslow et al, 1988). Granulation typically improves the flowability and wettability of powders, producing larger, more spherical particles. In addition, fines are eliminated and the bulk density is less likely to change during storage and transport (Jones, 2001). Granulation can be achieved in a batch, semi-batch or continuous process using a high-shear or low-shear system. High-shear mixer granulation was identified as being the most suitable process for whole milk powder due to the capability of the process for handling sticky materials.

1.2 Project Objectives

The objectives of this research were to:

- granulate milk using a high-shear granulator in a batch process
- investigate the physical changes that may occur to the milk during granulation
- design and simulate a continuous granulation process that will be able to granulate milk using milk concentrate as the sole ingredient.

1.3 Thesis Outline

This thesis aims to investigate the high-shear granulation of whole milk powder and seeks to provide an understanding of the subsequent effects of granulation on the functional quality of dried milk.

Chapter 2 reviews the literature and summarises the knowledge relevant to granulation of milk powders. The scope of the project limits the review primarily to

the particle technology aspects of granulation. As the project progressed there was a lot of knowledge outside the field of particle technology that was relevant. The knowledge of other issues, such as milk chemistry and surface science, are extensive but are herein only covered in summary.

Chapter 3 describes the equipment used for this research. Chapter 4 describes the alterations undertaken of a current batch high-shear granulation rig and the development of suitable experimental conditions for achieving milk granulation.

Chapter 5 describes a series of experiments performed to analyse the optimal process conditions to achieve successful granulation. The effects of varying process conditions on granule physical properties and the efficiency of the granulation are also covered.

Chapter 6 looks at the physical and functional effects of granulation on the granules produced in order to test the technical feasibility of milk granulation. A comparison between the original spray dried milk powder used as feed for the granulator and the final granules is given.

Chapter 7 investigates continuous granulation of milk powder. The results of a simulation of a continuous granulation process are presented. This determines whether continuous granulation is technically feasible. Process block diagrams are proposed for likely continuous granulation circuits, and their merits are discussed. The chapter also summarises suggested further work that is recommended.

The thesis finishes with a final chapter describing the major conclusions and recommendations.

Chapter 2 – Literature Review

2.1 Introduction

2.1.1 Granulation

Granulation is a form of size enlargement that is used to agglomerate primary particles from a feed powder into rounded granules by addition of liquid binder to the powder followed by agitation, which may be by mechanical action or induced by turbulence within a fluidised environment. Granulation improves the physical properties of the feed powder such as, reduced dustiness, greater product strength, better flowability, faster dispersion when mixed with liquids, and reduced segregation of different components of the powder mixture. The physical attributes of the granules produced are altered by controlling the formulation of the feed powder, the type of binder, the ratio of binder to powder, and the process design of the granulator.

Granulation is generally separated into high-shear and low-shear granulation, depending on the intensity of the forces used to granulate the powder. In low-shear granulation, gravity is often used to provide energy to force particles together such as in drum or pan granulation. Low-shear granulation is also achieved through spraying liquid binder onto a fluidised bed. High-shear granulation is carried out through high intensity mixing and compaction by mechanical action. Other granulation processes exist, for example using direct spraying (prilling), extrusion or compression to achieve granulation, but these will not be covered in this review.

2.1.2 Granulation of milk powder

Granulation is used to improve the physical properties of powders. The granulation of milk powder would seek to improve flowability, dispersability, storage performance and with low dustiness without adversely affecting the chemistry of the milk.

Size enlargement of milk powder in the dairy industry is called 'agglomeration'. This is done to 'instantise' the powder (Knipschildt, 1993), which means to improve the reconstitution properties of the milk powder without changing its solubility (Caric, 1994). This is achieved by increasing the size of the particles, which allows them to overcome the surface tension of the water and become wetted more easily. Instantised milk also contains a certain volume of air. This porosity also aids wetting, as the air will be replaced with water when reconstituting the milk. The porosity of the milk particle will also affect the dispersion of the powder. A more porous particle will have a greater solid-liquid interface when wet and therefore disperse quicker.

Agglomeration is achieved in New Zealand by recycling fines from the drying phase of the spray drying process. The fines are blown into the top of the spray drier and are contacted with the milk spray, causing agglomeration of wetted fines and droplets. Alternatively, milk powder is agglomerated in a fluidised bed with water or steam used as a binding agent. The moisture content may rise to 6-10 % during the agglomeration and is followed by a re-drying stage using hot air. Knipchildt (1993) suggests that low to medium heat powder (used in the spray drying process) should be

used for agglomeration of milk powder, as using powder already exposed to a relatively high amount of heat may result in a cooked flavour of the reconstituted milk. The mean size of the 'instantised' agglomerated milk powder particles produced in the dairy industry is around 300-400 μm (Masters, 1991).

2.2 Granulation Processes

2.2.1 Low-shear granulation

Low-shear granulation involves low intensity mixing of the powder with the binder. Processes using low-shear granulation will take longer to reach the same extent of granulation when compared with high-shear granulation, as the slower energy input will result in slower granule growth and generally have a coarser and broader particle size distribution. A further disadvantage is that more binder is required to achieve the same degree of granulation when compared to high-shear systems (Parikh, 1997). The advantage of low-shear over high-shear granulation is that the slow energy input of the low-shear environment results in less heating of the granules during the process. Consequently, the process equipment does not require cooling facilities.

Low-shear granulators often use a tumbling action such as provided in a rotating drum or pan. The tumbling action can be further improved by adding baffles that help to mix and break up large (soft) lumps in the powder. Tumbling granulators require that the pan or drum rotate at a speed less than the critical speed at which a particle will remain at the wall due to centrifugal forces. Tumbling granulators produce spherical granules of moderate density. Modern processes are able to handle up to 800 tons per hour (Ennis and Litster, 1997). As an example, granulation using low-shear systems is used in the fertiliser industry to improve the strength and durability of fertiliser granules to ensure adequate product resistance to attrition during handling, to reduce particle drift when distributed by air, or to provide a slow release of the fertilising chemicals (Gervin, 2002).

Fluidised bed granulation is also a form of low-shear granulation. It uses a turbulent flow of air to fluidise the powder. The binder is added as spray onto the top of the fluidised bed, with larger particles forming and sinking down through the bed (Ennis and Litster, 1997). The surface of the fluidised bed is constantly renewed by the smaller particles. The particles grow as the binder is added until the maximum moisture content is achieved, resulting in de-fluidisation of the bed. The maximum granule size is therefore determined by the ability of the bed to be fluidised (Schaafsma et al, 1999).

2.2.2 High-shear granulation

High-shear granulation uses an impeller blade to mix a particle bed. The intensity of collisions between particles is high enough so that both coalescence and breakage occur. Larger particles are more likely to be broken up during agitation so high-shear granulation limits particle size. The high intensity of mixing means that granule growth can occur rapidly, generally producing more rounded granules than other granulation processes. High-shear granulation is particularly useful for granulating cohesive powders, as the shearing forces are high enough to avoid uncontrolled

granule growth. The pharmaceutical industry is an example where high-shear granulation is used to increase product quality by blending important components in an excipient matrix to avoid segregation and to improve handling and tabletting performance.

A typical mixer-granulator contains a base mounted impeller in a cylindrical bowl. Wellm (1997) found that the design of the impeller had little influence on the granule size distribution. However, Schaefer et al (1993a) found that the size and shape of the impeller is important to ensure that a favourable powder flow pattern is formed in the granulator during melt-granulation.

A chopper is often included to increase the energy input. It is generally smaller and spins at a much higher speed than the impeller, with the axis commonly being perpendicular to the axis of the impeller. The chopper design can be altered to provide a size limiting function or add collisional energy that will aid in the growth of granules. The role of the chopper will depend on the blade design and the speed of the chopper.

2.2.3 Binder addition

A binding substance is usually added to aid the inter-particle attractive forces. The binding substance is a liquid that forms liquid bridges between the primary particles, which increases the inter-particle bond strength and therefore increasing the overall granule strength. The binder is usually added by one of three techniques,

(i) pouring it into the granulator at the start of the granulation

(ii) melting a low-melting point substance

(iii) spraying the binder onto the powder surface.

Each method of binder addition has specific benefits, which are discussed below.

2.2.3.1 Pour-on addition of binder

Pour-on of binder at the start of the granulation is the simplest method of binder addition. It is only used for combinations of product and binder that wet easily, as the initial distribution of binder in the powder will be poor. This can affect the quality of the final granules formed (Ennis and Litster, 1997).

An additional advantage for pour-on granulation is that any liquid binder can theoretically be used for this method, providing binder distributes well during the granulation. Scott *et al.* (2000) found that using a pour on technique for granulating calcium carbonate with Polyethylene Glycol (PEG) in a high shear mixer granulator resulted in an initial bimodal distribution of particle sizes that evolved into a uniform size distribution by the end of the experiment. The distribution of binder throughout the powder became more homogenous as the granulation proceeded because the initial large lumps of granules were broken into smaller granules that were more even in size.

2.2.3.2 Melt granulation

Melt granulation uses a solid binder that melts by heating (Schaefer et al, 1990). It is also a simple operation and controlling the temperature of the process can influence some parameters of the granulation. The binder is added as a powder or flakes and is mixed evenly throughout the powder. The temperature of the granulation is then increased to the melting point of the binder, either through external heating of the granulator or heat addition through agitation. As the binder melts, the powder particles will begin to coalesce and granulation will begin. When the granulation is complete the granules are allowed to cool, solidifying the binder liquid bridges within the granules (Schaefer et al, 1993a, 1993b).

Melt granulation is generally restricted to the use of binders with melting points above ambient temperatures, such as Polyethylene Glycol or waxes. Melt granulation is also restricted to high shear granulation, as the binders used are generally of a high viscosity once melted and a high intensity mixing environment is required to ensure continued blending of the binder-powder mixture.

2.2.3.3 Spray-on addition of binder

Spraying-on is the most common method of binder addition for granulation. It is necessary in cases where powder and binder do not wet well, or for strongly hydrophillic powders which adsorb and immobilise the binder fluid in a gel. The binder is atomised into fine droplets by directing the fluid through a spraying device, the design of which determines the size of the droplets. A spray is produced by introducing energy into the liquid through pressure, centrifugal, kinetic, sonic or vibrational means (Masters, 1991). The simplest method to achieve this is through the use of a pressure spray nozzle in which the liquid flow is forced through a small orifice under pressure. The pressure energy of the liquid flow is converted into kinetic energy of thin moving liquid sheets after exiting the orifice, which then disintegrate into droplets.

One of the most effective methods for atomising the binder is by using a twin-fluid atomiser, which combines a pressure nozzle with a high velocity gas flow. The gas provides extra energy for breaking up the fluid into droplets. The flow of liquid coming out of the spray nozzle forms a hollow cone sheet due to the mixture of the liquid with an air vortex in the spray nozzle. This breaks the liquid into droplets due to turbulent rotational forces (Grant, 1997). Kufferath *et al.* (1999) found that turbulent flow produced a more even drop size distribution than laminar flow in a twin fluid internal-mixing atomiser. Increasing the flowrate through the nozzle so that the turbulent regime began to cavitate resulted in a less uniform droplet size distribution, suggesting that for any nozzle, there is an optimal flowrate that will provide the best droplet size distribution.

2.3 Granulation Theory

2.3.1 Granule growth mechanisms

Mechanistic understanding of granulation has developed over the past 30 years with varying descriptions of the particulate interactions that can occur within the granulation process. The mechanistic approach has identified growth regimes and

processes such as nucleation, coalescence and attrition break-up, and from this a better understanding of granulation has evolved (Tardos, 2001). Unfortunately, granulation theory has not developed to a stage where the control and operation of full scale granulation processes can be fully understood, and a great deal of empiricism still exists (Iveson *et al.*, 2001).

Early research into granulation tended to concentrate on drum and pan granulation whereas research during the past 15 years has focused more on complex processes such as high-shear granulation and fluidised-bed granulation. Vonk *et al.* (1997) argues that the earlier work defining mechanisms of growth of granulation cannot be directly translated to high-shear granulation, as the physical circumstances are different. It is likely that the consolidation and breakage rates are affected by the intensity of the mixing forces during granulation although no mechanistic explanation of the difference between low-shear and high-shear granulation has been investigated. The three mechanisms of granulation described by the Ennis and Litster (1997) approach are shown in figure 2A.

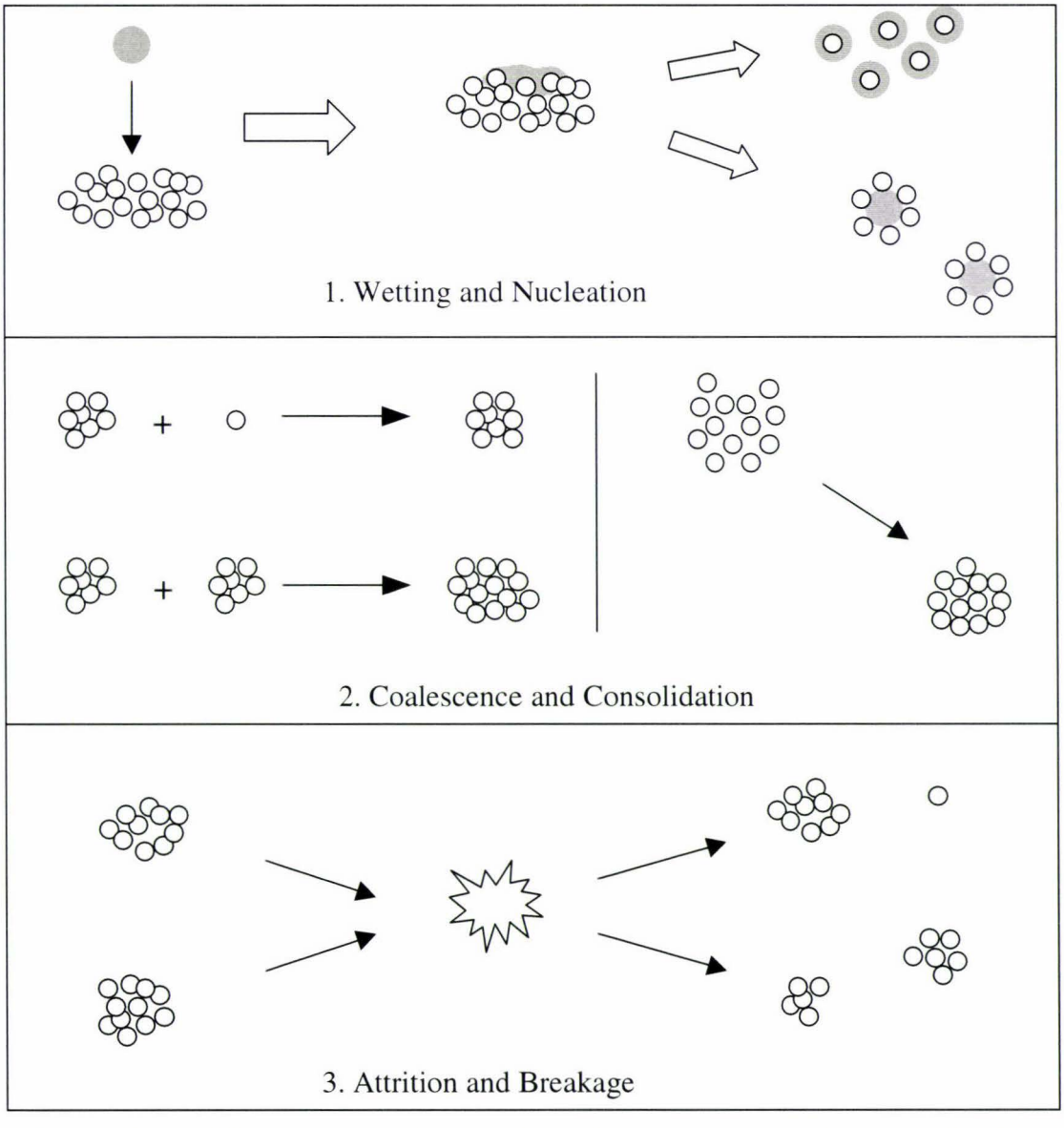


Figure 2A The present understanding of granulation mechanisms divided into three categories as described by Ennis and Litster (1997).

2.3.2 Nucleation and wetting

Nucleation occurs when the liquid binder is introduced and distributed throughout the dry powder bed. The formation of nuclei granules occurs and the distribution of the binder is important at this stage. Iveson *et al* (2001) state that current knowledge on the processes controlling nucleation is limited, although many researchers have identified the initial distribution and wetting ability of the binder as being important. The wetting of the binder into the powder is dependant on the contact angle between the solid and the binder and the spreading coefficient of the liquid phase over the solid phase.

Litster *et al.* (2001) used a dimensionless spray flux to define the spray-powder interaction for binder being sprayed onto a powder bed. The spray flux is a measure of the volumetric flow rate of liquid spray over the flux of the powder area covered by liquid spray droplets. A change in the spray flux was found to result in a change in the size and shape of the nuclei size distribution. The dimensionless spray flux is defined as the ration of the projected area of the droplet to the area of the bed and can be written as,

$$\psi_a = \frac{3V}{2Ad_d}, \quad \text{where} \tag{2.1}$$

V = volumetric spray rate flowrate of the spray nozzle [m^3/s]
A = flux of the powder surface through the spray zone [m^2/s]
d_d = average drop size. [m]

The variables of solution flowrate, powder flux and binder drop size are included in the analysis and using the dimensionless spray flux these values can be scaled-up for industrial processes. The suggested value of spray flux that should be used as a maximum is 0.1 to ensure a minimum of overlapping spray droplets. However, this is not practical in high-shear granulators because binder addition will take too long. Therefore, dimensionless spray fluxes of 0.8 or greater are typically used depending on the volume of the mixer (Hapgood, 2000).

2.3.3 Coalescence and consolidation

2.3.3.1 Coalescence

Coalescence is the mechanism under which already-nucleated particles grow by colliding and sticking together. A high proportion of research into granulation has focused on coalescence with current research almost at the point where modelling and simulation are feasible.

Ennis *et al.* (1991) described the collisions of rigid particles using dimensional analysis in an attempt to discover an *a priori* classification of coalescence and breakage. The dimensionless Stokes number was introduced with a critical Stokes number describing the point at which a collision between two particles would result in coalescence or separation.

The Stokes number, St, is given as,

$$St = \frac{4\rho u_o d}{9\mu} \quad \text{where,} \qquad (2.2)$$

ρ	= granule density	[kgm^{-3}]
u_o	= relative collisional velocity of particles	[ms^{-1}]
d	= harmonic average of granule diameters	[m]
μ	= binder viscosity	[Pa s]

Ennis *et al* (1991) further define a critical Stokes number, St*, to account for energy loss in collisions. This is given as,

$$St^* = (1 + \frac{1}{e_r}) \ln(\frac{h}{h_a}) \quad \text{where,} \qquad (2.3)$$

e_r	= coefficient of restitution = v_i/v_r	[dimensionless]
h	= binder layer thickness	[m]
h_a	= surface roughness or asperity height.	[m]
v_i	= velocity at impact	[ms^{-1}]
v_r	= rebound for normal collisions	[ms^{-1}]

The derivation of these quantities is complex and is not included in this discussion. The Stokes number describes the ratio between the kinetic energy of collision and the viscous dissipation energy. Ennis *et al.* (1991) identified three regimes of granulation depending on the ratio of Stokes number to critical Stokes number.

(i) $St < St^*$ Defined as the non-inertial growth regime. Coalescence always results providing binder is present.

(ii) $St \approx St^*$ Defined as the inerital growth regime. Coalescence may or may not occur, depending on the collisional forces and viscous dissipation forces.

(iii) $St > St^*$ Collisional forces are greater than viscous dissipation forces and no overall growth occurs.

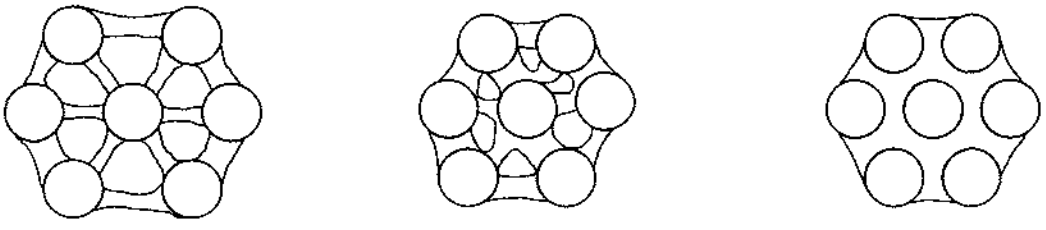
One of the major weaknesses of the Ennis *et al.* (1991) model was the assumption that the granules were rigid. Kristensen (1996) found that a granule becomes more deformable as the binder saturation increases, resulting in an improved probability of granules coalescing. The deformability of the granules may also depend on the density, porosity and particle size of the granules and the viscosity of the binder. A more deformable granule is more likely to absorb collisional energy and coalesce with another granule than a less deformable granule.

Liu *et al.* (2000) extended the model of Ennis *et al.* (1991) to include deformation during granule collisions. The Stokes deformation number, St_{def}, was introduced as the ratio of kinetic energy imparted during impact to energy dissipation due to plastic deformation of the granule. Two types of coalescence were identified depending on the Stokes number and Stokes deformation number. Type I coalescence is where the

viscous dissipation of collisional energy is high enough so that the solid particle surfaces will not contact and deform (equivalent to the non-inertial growth regime described by Ennis *et al.*, 1991). Type II coalescence occurs at higher Stokes numbers where the deformation of the particles becomes important and will determine whether the particles will rebound or not.

2.3.3.2 Consolidation

Consolidation during granulation describes the densification and subsequent change in porosity and liquid content of granules. The progression of consolidation has been described qualitatively by Newitt and Conway-Jones (1958) and is shown in figure 2B. York and Rowe (1994) made an addition, describing a droplet state and pseudo-droplet state as shown in figure 2C. The pseudo-droplet state accounts for included air bubbles.



(a) Pendular state (b) Funicular state (c) Capillary state

Figure 2B The progress of consolidation according to Newitt and Conway-Jones (1958) showing the pendular, funicular and capillary states.

(d) Pseudo-droplet state (e) Droplet state

Figure 2C. The York and Rowe (1994) addition to the states of consolidation showing the droplet and pseudo droplet states.

Ease of deformation relates directly to ease of consolidation. Therefore, any variation in process or formulation that affects deformation will also affect the ability of the granules to be consolidated. Binder viscosity has been found to affect the consolidation mechanism of granulation. An increase in viscosity will generally

cause a decrease in the rate of consolidation although this will only occur if the binder is over a certain minimum viscosity specific to each system. Below this minimum viscosity it is likely that inter-particle friction is the main force opposing consolidation (Iveson *et al*, 2001). Decreasing the particle size decreases the rate of granule consolidation, due to the consequent decrease in average pore size that the binder must be forced into.

Consolidation reduces the size of granules by removing entrapped air, resulting in a reduced porosity and stronger granules. Consolidation of granules may involve a period where little or no granule growth occurs but the porosity and density of the granules decreases and increases respectively (Iveson *et al*, 2001). The rate of consolidation will depend on the power input of the granulator, with an increase in power input resulting in more rapid consolidation.

2.3.4 Attrition and breakage

In a high-shear granulator attrition and breakage are occuring simultaneously to growth and consolidation (Ennis and Litster, 1997). Attrition is where small amounts of material are removed from the surface of the granule whereas breakage involves the shattering of the granule into two or more pieces through a plane or planes of fracture. The attrition and breakage of granules controls the maximum size of the granules in the granulator. The formulation of the granules will determine the strength and hardness and will determine the force required to break or shear a granule. An increase in the intensity of mixing will result in an increase in attrition and breakage, as more energy will need to be dissipated within the granules.

Kristensen *et al.* (1985a) found that the strength of a granule is determined both by the strength of the dry powder compact and the strength due to mobile liquid bonding, for fine powders compressed to porosities similar to granules manufactured through high-shear granulation. The strength of the dry powder compact depends on the interactive forces between particles as well as the ability of the particles to interlock. A fine powder generally has an increased strength compared with a coarse powder; a powder containing flat particles that can interlock is able to resist shear more effectively than a powder containing spherical particles.

Moisture content is also important to the strength of granules. Kristensen *et al.* (1985a) found that the strain required to fracture a moist agglomerate increased as the liquid saturation increased. The free liquid at the surface of the granule may also determine the type of size reduction process that occurs. Abrasion transfer (or attrition) may occur more readily for a surface dry granule whereas fragmentation of the granule or breakage may occur if a saturated granule has free surface liquid present. A surface dry granule will allow individual particles to be removed as a means of energy dissipation while a granule with binder fluid filling all of the interstitial spaces will have less attrition and will have an increased absorption of collisional energy. A wet granule will dissipate energy throughout the granule, resulting in more deformation of the granule; this may increase the probability of a breakage plane forming.

2.4 Process Variables

Most previous research on the effect of process variables on the granulation process has been empirical in nature. Campisi *et al.* (1999) used an experimental design technique to determine the effect of impeller speed and massing time in order to identify the optimal operating conditions for a high-shear mixer-granulator for a specific formulation in a ten litre granulator. Granulation is a notoriously difficult process to scale-up, and operating conditions may have to change markedly between different sizes of granulating processes (Johansen *et al.*, 1999, Schaefer *et al.*, 1993a).

2.4.1 Impeller speed

Theis and Kleinebudde (1999) found that binder concentration and impeller speed were the most important factors influencing granule growth. Schaefer *et al.* (1993b) found that an increase in impeller speed resulted in larger, smoother granules although the effect of this varied for different loads in the granulator. Knight *et al.* (2000) also found that impeller speed affected agglomeration behaviour but were able to further state that at low impeller speeds, the extent of size enlargement increased with increasing power input. Also, at high impeller speeds, the extent of size enlargement was low relative to the large power input with the difference being attributed to energy lost through breakage mechanisms.

Knight *et al.* (2000) suggested that breakage of granules is dependent on the impeller speed of a mixer-granulator. They found that at a low impeller speed of 450 rpm (revolutions per minute) the granules produced were highly spherical in nature while those produced using a high impeller speed of 1500 rpm were irregular in shape. The difference was attributed to granule breakage, as at higher impeller speeds the particle size distribution was wider with more fines being generated. Schaefer (1996a) suggests that the effect of viscosity on densification disappears when the impeller speed becomes sufficiently high.

2.4.2 Granulator component design

The design of the impeller was investigated by Schaefer *et al.* (1993a) to determine whether including a curved blade tip would aid in melt-granulation. They found that using a curved impeller blade resulted in a higher power input with more smooth and more spherical granules than a plane impeller blade in the same granulator.

Schaefer *et al* (1993a) tested the usefulness of including a stationary central cone suspended from the centre of the lid in an eight litre vertical axis mixer-granulator. They found that removal of the cone resulted in 25-30% of the material becoming stuck to the walls during the granulation. This was attributed to the granulator being too high and the cone acted to reduce the height of the bowl. They state that the inclusion of the cone reduces the ratio between the height and diameter of an eight-litre granulator from 0.95 to 0.56 and compare this to a fifty-litre granulator with a ratio of 0.5.

2.4.3 Power input

Ritala *et al.* (1988) found that the power consumption in high-shear granulation is influenced by the intra-granular porosity and by the surface tension of the binder solution. They suggest that power consumption is related to the strength of the liquid mobile bonding in the granules and showed that power consumption reflects the change in porosity during granulation.

Terashita *et al.* (1990) monitored the power consumption during granulation using two different mixer granulators with three different formulations of powder to be granulated and found that granulation could be separated into four stages, characterised by changes in the power consumption curve. The four stages of granulation were described as the formulation of agglomerates and nucleic particles, the growth into granular particles, granular refinement and the completion of granulation. The most spherical granules with the smallest size distribution were found at the beginning of the last stage.

2.4.4 Liquid saturation

Power consumption and granule growth rate are correlated with the liquid saturation of granules (Ritala *et al.*, 1988). As the powder becomes moist it becomes more difficult to strain, resulting in a higher torque requirement and increased power consumption. An increase in power input into the granulator has also been found to increase the granule growth rate. Kristensen *et al.* (1985b) state that at a certain range of liquid saturations, dependant on the properties of the powder, the granule growth rate increases significantly and the growth rate due to coalescence becomes the dominant growth mechanism. Johansen *et al.* (1999) found a correlation between the reproducibility of melt-granulation experiments and the binder concentration with the worst reproducibility coming from the highest binder concentration runs.

2.4.5 Temperature

The temperature conditions under which the granulation occurs will affect the particle growth rate due to a decrease in binder viscosity with increasing temperature (Schaefer and Mathiesen, 1996c). An increase in temperature will also cause thermal expansion of the binder, resulting in an increase in liquid saturation, and will also have an effect on the chemical properties of heat sensitive powders or binders, which may affect the final granule properties. Schaefer and Mathiesen (1996c) found that the intragranular porosity was affected by an increased temperature for the granulation of lactose monohydrate with PEG 6000 and they suggest that this was caused by evaporation of the water of crystallisation.

Temperature increases during wet granulation. Holm *et al.* (1985) state that the energy consumed in granulation is converted completely into heat, resulting in heating of the granular material. The mechanism for the energy conversion is through straining the moist agglomerates during agitation. Often, granulators are cooled in some way to reduce the effects of heat generation and to protect the granules from damage due to heat. Heat generation will be controlled naturally to some extent through evaporation of the binder during granulation. This will have the effect of

drying the granules, slowing granule growth, and increasing the amount of binder required to achieve granulation.

2.4.6 Humidity

Schaafsma *et al.* (1999) suggest that the adsorption of water on the particle surface increases at a higher relative humidity for a fluid bed granulation of lactose monohydrate with a binder of water with eight percent polyvinylpyrolidone (PVP). They further argue that a decrease in inter-particle forces with increasing relative humidity is possible since this will enhance conductivity, reducing electrostatic forces.

2.4.7 Batch load of the granulator

The amount of powder introduced into the granulator will affect granulation performance. Schaefer *et al.* (1993b) found that reducing the mixer load resulted in an increase in the amount of lumps for melt-granulation in a high-shear mixer. Theis and Kleinebudde (1999) also found that the granule growth was delayed with an increasing load in a batch high-shear mixer-granulator. This suggests that there is an ideal loading for each granulator under which granulation will occur rapidly, with a minimum amount of lumps. It is also likely that the load will depend on the type of powder to be granulated, as the bulk density of the powder may be important.

2.5 Composition

The composition of the material to be granulated plays an important role in the granulation process (Faure *et al.*, 1999). In many cases the formulation of the product needs to be altered to ensure a successful granulation. The choice of binder is also critically important, not only for the success of the granulation, but also because the final product will be composed of a mixture of the original powder and binder. Varying the formulation and binder is commonly done in the pharmaceutical industry but will not be discussed further here as a purely milk based product is the objective of this work. The following sub-sections discuss properties of the binder and powder that affect granulation and are aplicable to this project.

2.5.1 Effect of binder viscosity

The viscosity of the binder will affect the deformability of the granules and hence play an important role in the coalescence of particles; the magnitude of this effect will depend on the process conditions. Ennis *et al.* (1991) found that the viscous dissipation of collisional forces becomes significant and results in increased agglomeration if the viscosity is high enough. Conversely, Schaefer and Mathiesen (1996b) suggest that using a lower viscosity binder will result in increased agglomeration through the coalescence mechanism. They also found that using higher viscosity binders resulted in a greater likelihood of uncontrollable growth during granulation. Liu *et al.* (2000) found that granules deform more using a lower viscosity binder, resulting in an improved probability of coalescence due to an increased contact area between colliding granules. Johansen *et al.* (1999) found that a low viscosity binder granulates over a wider binder concentration range than a high viscosity before overwetting and uncontrollable granule growth occurred.

Mills *et al.* (2000) found that the viscosity of the binder affected both the rate of growth of granules and the mechanism by which the granules grew. They found that the granule growth rate increased with increasing binder viscosity up to a maximum viscosity of around 100 mPa s. The growth rate under these conditions was found to occur due to layering. Increasing the binder viscosity above 100 mPa s resulted in growth due to coalescence with no layering observed.

2.5.2 Particle size and shape

Schaefer (1996b) investigated the effect of melt-granulating varying amount of lactose with the plate-like compound Mannitol to discover if the physical properties of individual components of a mixture determines the agglomeration properties of the mixture, as was found in previous work on wet granulation (Oakunle and Spring, 1976a,b,c, Jaiyeoba and Spring, 1979, 1980a,b, Kleinebudde and Nymo, 1995). The plate-like structure of the Mannitol caused the granules to become less spherical with an increasing proportion of Mannitol. Schaefer (1996b) also suggested that the optimum amount of binder with Mannitol, Lactose and PEG was related to the packing properties of the powder.

Knight *et al.* (1998) found that the particle size was of critical importance in the size distribution and growth behaviour of high-shear mixer-granulation. During granulation a bimodal particle size distribution evolves that eventually progresses into a uni-modal size distribution. The rise of the bimodal distribution is described as being an intrinsic feature of all processes in which liquid is mixed into a fine solid. Scott *et al.* (2000) followed on from this by investigating the heterogeneity of particle size in high-shear granulation and postulated two hypotheses; the preferential nucleation hypothesis and the preferential growth hypothesis.

The preferential nucleation hypothesis states smaller particles are preferentially included into growing granules because the capillary force acting to engulf a particle will increase with decreasing particle size. Smaller particles are therefore more likely to be found in larger granules. Scott *et al.* (2000) found evidence that this is likely to occur in pour-on granulation for a PEG binder with 'Durcol 40' powder.

The preferential growth hypothesis further states that collisions involving larger particles are less likely to coalesce as the inertial forces seeking to disrupt the newly paired entities will be larger when the entities are large. Smaller particles are more likely to coalesce. This will result in larger granules being made up of coalesced smaller primary particles and smaller granules containing large primary particles.

2.5.3 Cohesiveness of the powder

Schaefer (1996a) found that it was difficult to control the granulation of a cohesive material. He granulated cohesive anhydrous lactose with PEG 3000 and PEG 6000 and found that the process was difficult to control because the liquid saturation had to exceed 100% to obtain a useful granule size. At such a high liquid saturation the granulation is likely to be difficult to control due to over-wetting, as the surface of the granules will be wet resulting in uncontrolled coalescence and growth. Addition of

coarse lactose made the mixture less cohesive, the growth rate more easily controlled, and increased the densification rate.

2.5.4 Solubility of the powder in the binder

Theis and Kleinebudde (1999) found that the amount of binder required to granulate is reduced with an increase in solubility of powder in the binder solution for the hygroscopic drug sodium valproate melt-granulated with glycerol monostearate. Schaefer *et al.* (1990) describe this effect as being due to the dissolution of the binder on the surface of the particles, reducing the particle interactions and improving the deformability of the moist agglomerates.

2.6 Instrumentation and Control of Granulation

2.6.1 Power consumption

Power consumption in high-shear granulation is related to the swept volume of the impeller blades and is a measure of the collisional energy that is imparted to the powder or granules in the granulator. Kopcha *et al.* (1992) investigated the measurement of power consumption, direct torque and reactive torque in an attempt to describe granulation in a high-shear mixer-granulator. They argue that a direct measure of power consumption may not provide a reasonable indicator of granulation as power consumption will be reliant on a power factor to represent the angular displacement between the current and voltage in an alternating current induction motor. A further deficiency identified is that the motor and gear assembly efficiencies will not remain constant over the lifetime of the granulating device, resulting in further inaccuracies over time. They prefer measurement of direct or reactive torque on the impeller shaft will avoid these problems.

2.6.2 Impeller shaft torque

Torque is a quantitative measure of the tendency of a force to cause or change rotational motion of a body (Young, 1992). Torque is related to power and total work by the equation:

$$W = \int P dt = \int 2\pi n \tau dt \qquad (2.4)$$

where W = Work done (J)
P = Power (W)
n = impeller revolutions per second (s^{-1})
τ = torque (Nm)

Kopcha *et al.* (1992) break down torque into direct torque and reactive torque, where direct torque is measured in the shaft strain in a rotating drive shaft and reaction torque is measured through the reaction of the motor to the shaft torque. The instrumentation used strain gauges to measure the torque in the impeller shaft (for the direct torque) and in the motor shaft (for reactive torque). It was found that the two methods of torque measurement as well as a direct power consumption measurement gave similar curves during granulation. The direct torque measurement was chosen as

the most reliable method of instrumentation as it does not include the motor and gearbox losses mentioned above.

Jones *et al (2002)* used a system of calcium carbonate granulated with an aqueous solution of 30 % PEG 6000. They found that the granulation end-point could be identified by a sharp rise in torque, which corresponded to wet massing when the granules had consolidated to their saturation point and moisture appeared on the granule surface. Wetted granules with a viscous surface layer are more difficult to mix, hence the increase in torque. They also investigated the effect of blade speeds on granulation and found that the time to end-point depended on the number of revolutions of the bottom mounted impeller, although this was dependent on the speed of the side-mounted chopper. Faster chopper speeds lowered the number of impeller revolutions required to reach the end-point.

2.6.3 Humidity control

Schaafsma *et al.* (1999) looked at the effect of humidity on fluidised-bed granulation and found that the minimum fluidisation velocity in the fluid bed is influenced by the relative humidity. This was explained by the fact that the equilibrium adsorption of water molecules on to particle surfaces increases at higher relative humidity. The adsorbed layer can enhance the inter-particle forces by the formation of liquid bridges. A possible decrease in inter-particle forces for increasing relative humidity is also possible as the enhanced conductivity can decrease electrostatic forces. They suggest that a good way to control the humidity of fluid-bed granulation is to use pulse spraying of the binder into the fluid-bed. No instances of using humidity control for high-shear granulation have been found.

2.7 Chemistry of Milk with Regards to Granulation

This section is a summary of the literature that was not covered prior to the experimental work but was found to be useful during the course of the project. This is only a brief synopsis due to the time constraints of the project. The composition of milk powders will affect the performance of granulation as has been explained in previous sections. Furthermore, the quality of the final product will be affected by the chemical changes that the powder composition undergoes during processing. This section gives a description of the composition of milk and the following section (§ 2.8) discusses potential changes that may occur during granulation and subsequent processing.

2.7.1 Introduction

Milk is a complex biological product, consisting primarily of protein, fat and lactose solids dissolved or suspended in water with variations of the type and amounts of these constituents depending on the time of the milking season and the species of cow (Graf and Bauer, 1976). The physical properties of each of these components may be important in the granulation of milk. Furthermore, interactions between these components could also have a significant effect on the granulation and on the product quality after granulation. The physical characteristics of natural mature cow's milk are shown in table 2.7.

Table 2.7 Composition of mature cow's milk (Macie et al, 1953).

Component	Mean	Range	Units
Lactose	4.8	2.1 – 6.1	g/100ml
Protein	3.3	2.1 – 6.4	g/100ml
- Whey Protein	0.58	0.2 – 0.94	g/100ml
- Casein	2.5	1.4 – 6.3	g/100ml
Fat	3.7	0.9 – 9.8	g/100ml
Other components (Ash)	0.72	0.35 – 1.21	g/100ml
Total solids	12.8	7.7 – 15.6	g/100ml
pH	6.62	6.22 – 6.77	

2.7.2 Lactose

Dry lactose is present in either an amorphous or crystalline form, with a number of variations to the crystalline form possible depending on the conditions under which it is crystallised (Bronlund, 1997). Amorphous lactose is formed when a solution of lactose is dried rapidly so that the increase in viscosity occurs fast enough to stop crystallisation from taking place (Nickerson, 1974). Amorphous lactose is known to become sticky if the glass transition temperature is exceeded, with an increase in stickiness being dependant on the amount the glass transition temperature is exceeded (Brooks, 2000). The presence of amorphous lactose during granulation will likely affect the quality of the granules produced, especially if the granulation occurs at temperatures much higher than the glass transition temperature. This is likely as the glass transition temperature is moisture dependent and granulation requires a relatively high powder moisture content. If the glass transition temperature is reached amorphous lactose will begin to crystallise.

The exact time required to initialise crystallisation under varying conditions has not yet been investigated making it difficult to predict whether lactose will crystallize during granulation and what effect crystallization will have.

Crystallized lactose monohydrate and anhydrous lactose are commonly used in granulation as an excipient. Schaefer and Mathiesen (1996c) found that melt granulation with anhydrous (amorphous) lactose resulted in stronger granules than when using crystalline lactose monohydrate due to the increased cohesiveness of anhydrous lactose. They also found that the agglomerate growth rate is decreased by the evaporation of the water of crystallisation when using lactose monohydrate; the exact decrease in growth depends on the temperature of the mixture.

2.7.3 Protein

The protein found in cow's milk can be divided into two main types, casein proteins and whey proteins. Casein makes up about 82% of the protein in cow's milk the remainder being whey proteins (Early, 1998). Casein proteins are found in milk as micelles and form due to the hydrophobic nature of the protein. The micelle aggregations are held together through hydrophobic and hydrogen bonds. Whey proteins will remain soluble in the milk serum after the destabilisation and sedimentation of casein by acidification. The protein in milk is responsible for much

of the chemical changes that can occur during heating and processing (Singh, 1995, McKenna, 2000). A brief description of some of these reactions is given in section 2.8.

2.7.4 Fat

Fat exists naturally in milk as globules of varying sizes surrounded by a membrane of protein and phospholipids. The fat globule membrane encapsulates and provides stabilisation for the hydrophobic fat globules (Early, 1998). Due to the increased surface area for smaller particles, emulsions of small fat globules are thermodynamically unstable systems (Fäldt and Bergenståhl, 1996). The size of the globules therefore may change depending on the amount of processing that the milk has undergone (van Boekel and Walstra, 1995). During processing the emulsions are known to coalesce, resulting in larger fat droplet sizes.

The size of fat globules is deliberately altered during homogenisation, or standardisation, by forcing the milk through a small orifice at very high pressures. This has the effect of producing a more stable colloidal sized fat globule that will not form a cream layer. The disadvantage of homogenisation is that the fat membrane is altered through changing the surface area of the fat, replacing the natural protein and phospholipid membrane with a membrane made up of predominantly casein (Kessler,1981). Further fat-protein reactions may occur a result, such as aggregation due to heat coagulation of the casein onto the fat globules (van Boekel and Walstra, 1995).

Sharma et al (1996) compared the surface composition of recombined milk with fresh homogenised milk. They found that the fat globule membrane of milk recombined from skim milk powder and milk fat, and subsequently homogenised, consisted of casein micelles, fragments of casein micelles and whey proteins. The coverage by these proteins was found to be less than for fresh homogenised milk. They explain this by saying that the protein coverage varied with the fat globule size, suggesting that the fat globule size was larger for recombined milk.

Early (1998) describes the chemistry of milk fat as being dominated by two types of reaction, hydrolysis and oxidation. Hydrolysis is the process whereby free fatty acids are liberated from fat globules by enzymatic means. This requires the presence of lipase, which is present in natural milk but is inactivated by pasteurisation. Oxidation of milk fat reduces the shelf life of the milk and is catalysed by light and by trace levels of some metals such as copper.

2.8 Processing of Milk by Granulation

The purpose of this project is to investigate granulation as an alternative to spray drying for the production of milk powders. Thus, it is important to broadly determine the effects that granulation will have on the functional properties of the milk. Further study of the effects of granulation of milk may be useful in the future, as the effect of processing dry milk powders is a largely unknown subject.

Granulation of milk powder has not been done before, therefore this literature survey infers expected behaviour by reporting processing effects on milk and how these may relate to granulation. Granulation is expected to affect the chemistry of milk through two mechanisms; (a), heating due to friction during mixing; and (b), shearing forces.

Heating of milk allows an increase in the rate of reaction for any natural reactions between milk components as well as supplying the activation energy for reactions that would not normally occur at ambient temperatures. Shearing may allow the rupture of both fat globules and protein micelles, resulting in an increase in undesirable reactions that will reduce the quality of the product (van Boekel and Walstra, 1995).

2.8.1 Heating of milk

A great deal of work has been undertaken to understand the effect of heating on liquid milk products, especially for milk heated to high temperatures such as in UHT treatment or heat sterilisation. The heat stability of liquid milk is determined by using the heat coagulation time (HCT), measured as the time it takes to coagulate milk at a given temperature (McCrae and Muir, 1995). This gives an indication of the state of the protein in the milk as coagulation is caused by association of casein micelles and whey proteins (Singh, 1995). The composition and the pH of the milk affect heat stability with the maximum heat stability at a pH of approximately 6.7 (Singh et al, 1995). The heat stability of milk will affect the amount of fouling that will occur during the processing of liquid milk and have an effect on the storage of UHT treated milk.

Fink and Kessler (1985) investigated the effect of heating raw (non-homogenised) cream with a fat content of 30% at various temperatures and holding times and found that the free fat content, or non-globular fat, and the electrophoretic mobility of the milk increased dramatically at temperatures between 110°C and 140°C. They concluded that the stability of the fat globule membrane was reduced at these temperatures. No measurable change in the free fat content of the milk was found below temperatures of 105°C.

Singh (1995) gives a summary of the literature on the effects of heating on casein in milk and states that above 0°C the casein micelles can associate reversibly, becoming larger. It is not until the temperature becomes greater than 70°C that irreversible reactions occur, with increasing rate of reaction as the temperature rises.

Crystallisation of amorphous lactose may occur if milk powder is heated and requires the glass transition temperature to be exceeded (Jouppila and Roos, 1994). The glass transition temperature is a function of the moisture content of the powder and has the result of making the powder sticky (Brooks, 2000).

Lactose may react with milk proteins via the Malliard, or non-enzymatic browning, reactions when heated. The Malliard reactions may reduce the nutritional quality of the milk and reduce the functional properties of the proteins (O'Brien, 1995). The Malliard reactions are complex and involve many reactants and stages, with most of the stages being either 1st order or 2nd order reactions (O'Brien, 1997).

Heating of milk powder during granulation may have similar effects on the milk chemistry as heating of liquid milk although previous research on the heating of liquid milk was done at much higher temperatures than is expected for granulation. The shearing forces that the milk will be exposed to during granulation may therefore have a more substantial effect on the milk chemistry. Crystallisation of lactose during granulation may occur because of the longer timeframe compared to spray drying

although the effect this will have on the granulation is unknown. Furthermore, the effect of granulation on the fat globule and lactose morphology is unknown and it is likely that some changes may occur through crystallisation of the lactose or rupture of the fat globules.

2.8.2 Shearing of milk

Shearing has been studied in detail for liquid milk. McKenna (2000) found that high shear processing of whole milk resulted in significant aggregation of casein micelles and fat globules when spray dried and subsequently reconstituted. In contrast, low shear processing resulted in less aggregation. The mechanism of aggregation was suggested to be the presence of whey protein hair-like structures that joined the aggregates together.

The effects of shearing are also dependent on the temperature of the milk. Speigel (1999) found that shearing solutions of whey protein concentrate and lactose resulted in protein unfolding and aggregation unless the temperature was below 85°C where the reaction was slowed significantly. The presence of lactose also reduced the rate of protein degradation.

2.9 Chapter Closure

This chapter has described the various granulation processes currently used in industry and has described the present understanding of the science of granulation. Granulation is expected to improve the physical qualities of dried milk although this must be achieved without altering the chemistry or reducing the functional quality of the milk. The constraints of this project include the use of a process that will use minimal additives so that a pure milk product can be produced. This will require an investigation into the effects of varying physical parameters of the granulation on the quality of the granules produced. Furthermore, an understanding of the chemical and physical changes that the granules undergo during granulation will be required to determine the suitability of the process conditions.

Chapter 3 – Equipment and Laboratory Techniques

3.1 Introduction

This chapter describes the major equipment used for the experimental work of this thesis. A mixer-granulator was available at the start of the project but required some modification to be suitable for the granulation of milk powders using milk concentrate as a binder. This developmental work is described in detail in chapter 4.

Granules were made using a batch high-shear mixer-granulator that had a capacity of 1.5 kg of powder per batch. The apparatus is shown in figure 3A. Milk concentrate binder was added via a spray nozzle onto the surface of the milk powder. The torque on the granulator motors was monitored during the granulation to obtain torque profiles for further analysis and to ensure that the granulation did not place too much strain on the motors. Granules were then dried for further analysis.

3.2 High-Shear Mixer Granulator

The granulator bowl is vertically mounted onto a supporting frame. A 4 kW impeller motor sits under the granulator bowl and drives the main impeller. A gearbox with a reduction ratio of 3.98:1 reduces the motor speed. Mounted on the side of the bowl is a 0.55 kW chopper motor.

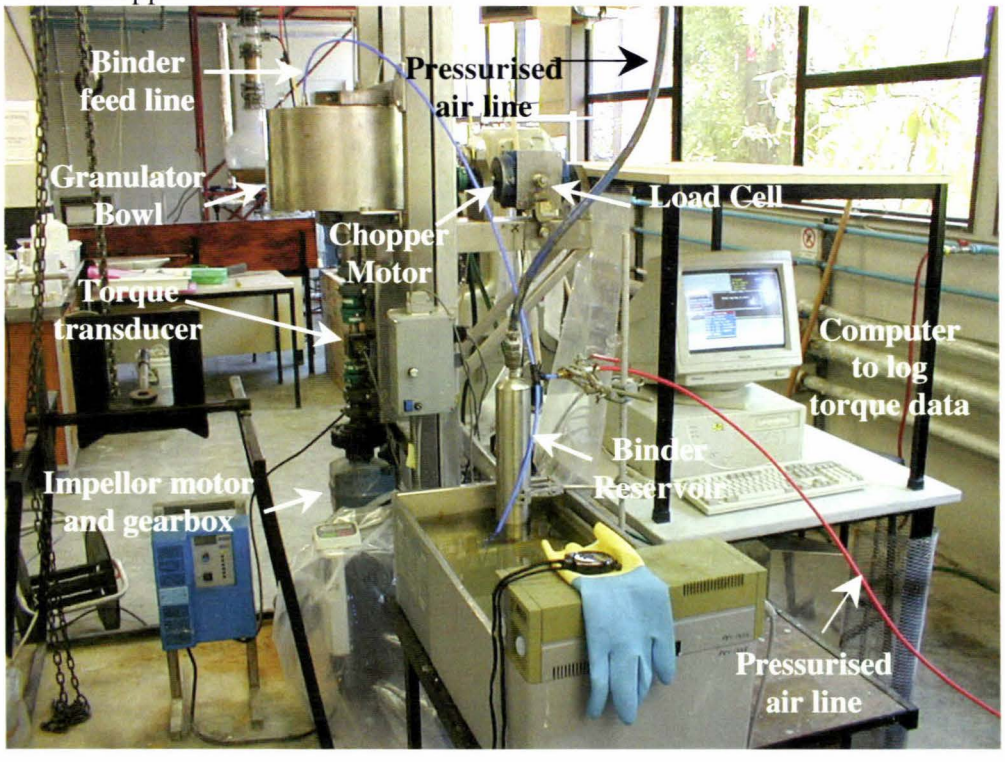


Figure 3A Experimental set up of the mixer-granulator

Binder is supplied from a pressurised reservoir and is added through a spray nozzle attached to the lid of the granulator bowl. The binder delivery vessel sits in a water bath to maintain the desired temperature of the binder and is pressurised using pressurised air.

3.2.1 Granulator bowl and mixing blades

The granulator has a large mixer blade through the central axis of the bowl and a small chopper blade located on the side wall of the bowl, perpendicular to the mixer blade as shown in figure 3B.

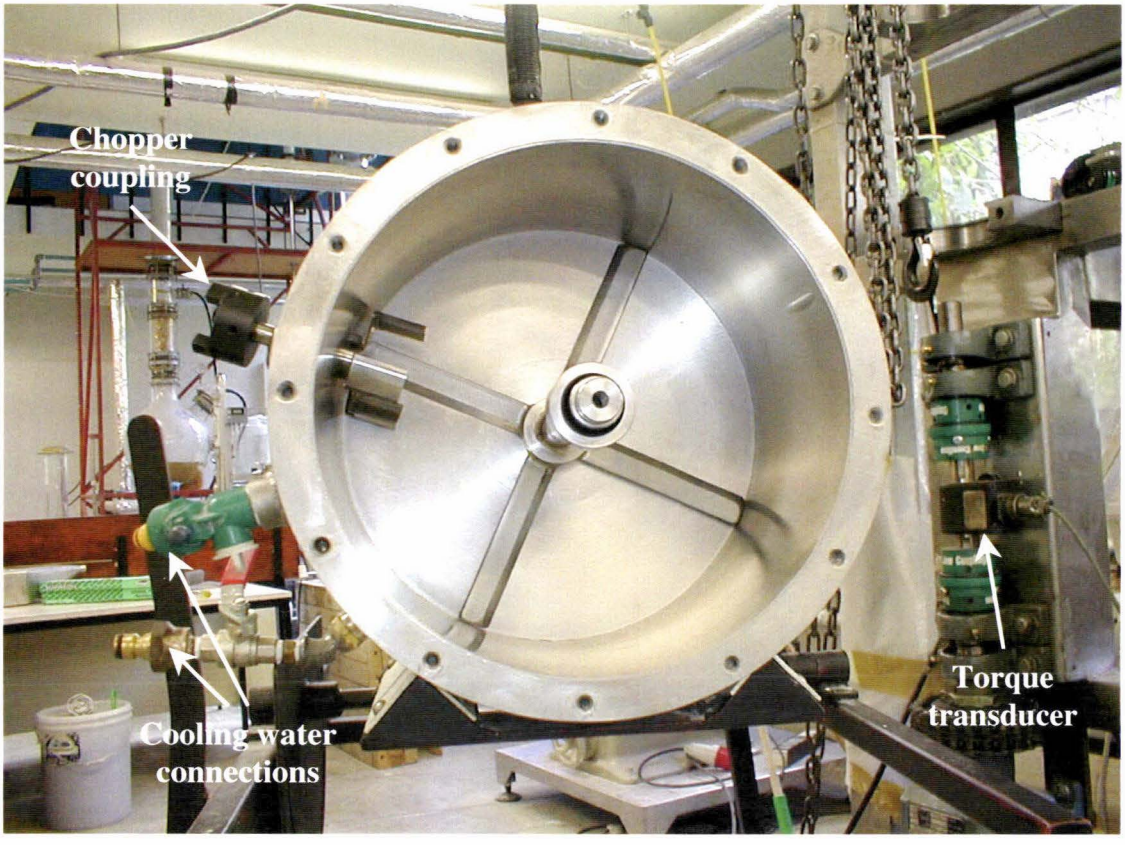


Figure 3B Granulator bowl and blades

Figure 3B shows the granulator bowl when looking down into the bowl. The internal dimensions of the bowl are 320 mm diameter and 270 mm depth. It can be removed from the motor assembly rig so that product can be removed and for ease of cleaning. The large mixer blade is removable to enable easy cleaning and to allow a change in blade design. The blade sits approximately 2 mm from the floor of the bowl the arms are tilted 9 degrees from the base of the granulator to reduce the risk of compaction of product under the blade during granulation. Each arm is 25 mm wide with a 40-degree bevel angle. At the left of the figure are the inlet and exit valves for cooling water. Above these is the chopper connection coupling. This is attached to the chopper by a shaft that protrudes through the granulator wall. The centre of the chopper sits 50 mm above the floor of the bowl and is 80 mm in diameter. Each of the three chopper blades is 40 mm long and 26 mm wide with a bevel angle of 31 degrees. The bowl is water-cooled to remove the heat generated by mixing.

3.2.2 Control and instrumentation

The motors that drive the mixer and chopper blades are operated through variable speed controllers used to change the speed of the blades. The mixer motor speed is controlled by a PDL controller, model X709, made by PDL Electronics Ltd. The chopper is controlled by a GEC MSC controller made by Flow-Line.

Two torque transducers measure the torque generated during granulation. The torque for the mixer blade is directly measured by a strain gauge situated in-line with the mixer shaft. This can be seen in figure 3B to the right of the figure. A load cell measures the reactive torque on the chopper motor by measuring the reaction of the chopper motor to the chopper shaft torque. A more in depth description of the reactive and direct torque is given in chapter two, section 2.6. The torque measurements are transmitted to a personal computer and are recorded as a torque versus time profile using a picolog data logging software package.

3.3 Binder Delivery

The development of the binder delivery system is described in chapter 4. The system described here was used for all of the experimental results described in chapters 5 and 6. The desired amount of binder to be added was weighed and then added to the reservior prior to each run. The binder was sprayed onto the powder surface in pulses to ensure the correct flowrate through the spray nozzle. Air was used to purge the binder delivery line between pulses, clearing the line of liquid and providing pressure between pulses to maintain a suitable spray pattern. Figure 3C shows the binder delivery set up.

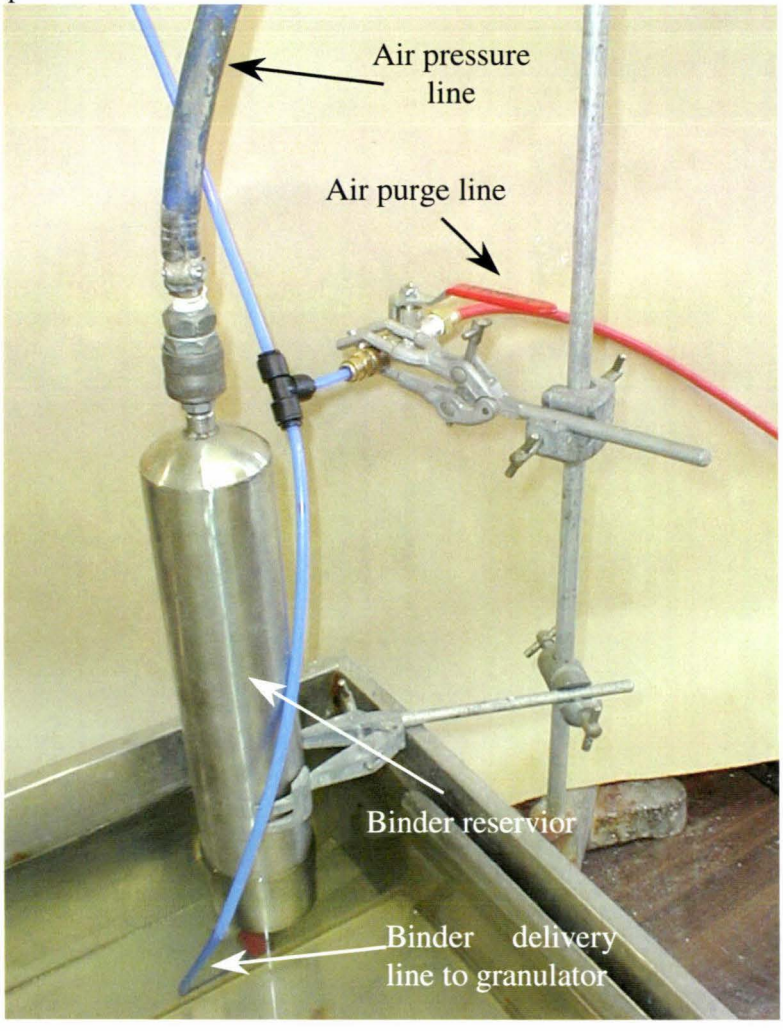


Figure 3C Binder delivery equipment

The binder reservior holds the binder for delivery. It sits in a water bath kept at 50 °C to ensure that the milk concentrate viscosity is suitable for spraying. The large hose

connected to the top of the vessel supplies air from a compressor at up to 6 bar. The binder is added to the granulator through the small line exiting the bottom of the vessel by opening the valve at the bottom of the vessel sitting in the water bath.

Binder is pulsed into the granulator for ten seconds at a time by closing the binder valve and opening the airline valve connected to the binder delivery line. Air then enters the line through the red air line shown in figure 3C and clears the line of binder, ensuring that the spray nozzle is kept clear and that the remaining binder in the binder delivery line is sprayed out at a suitable pressure. The next binder pulse is started by closing the air valve and opening the binder delivery valve.

The binder is sprayed onto the powder surface through a spray nozzle as shown in figure 3D.

Figure 3D Binder spray nozzle

The spray nozzle is a number 23 "TeeJet" model by Spray Systems Ltd., Auckland. The spray nozzle produces a flat spray and the direction and angle of the spray onto the top of the moving powder bed is altered to reduce wastage as explained in chapter four.

3.4 Drying Equipment

3.4.1 Open air drying

Granules were dried by leaving them in ambient conditions spread out over plastic trays. This enables the particle size distribution and physical properties of the granules to be observed without the possibly deleterious effect of the drying process. The depth of the granules on the trays was no more than 5 mm thick. This method was later found to be only useful in comparing the change in particle size distribution before and after drying as the total moisture content of the granules would only drop to around 8 % after 7 days. Storage of milk powders at high moisture contents results in chemical changes that lead to a reduction in reconstitutional performance (Baldwin, 2001). An investigation into these changes is shown in chapter 6.

3.4.2 Fluidised bed dryer

Granules that were to be used for further tests were usually dried in a custom made fluidised bed dryer. The dryer set-up is shown in figure 3E. Batches of up to 200 g or up to 2 kg could be dried by changing the main drying chamber from a 600 mm high by 80 mm diameter chamber to a 1.5 m high by 160 mm diameter chamber. The smaller drying chamber was used to dry samples for tests requiring only a small amount of granules while the large chamber was used to dry an entire granulation batch at a time.

The granules were dried for two hours as it was found that after this there was minimal reduction in moisture content. The air flow to fluidise a batch of around 1.7 kg of milk granules was found to be between 200 and 300 litres per minute. The air temperature used for drying granules that were to be further tested was 30 °C as granules dried at 40 °C or more were found to change colour noticeably after two hours of drying. A mesh hat made from 150 μm size mesh was used to filter fines from the small drying chamber while a fabric filter cloth 800 mm high was used to filter the large batch runs. Figure 3E shows the set up of the fluidised bed dryer running with the large drying chamber.

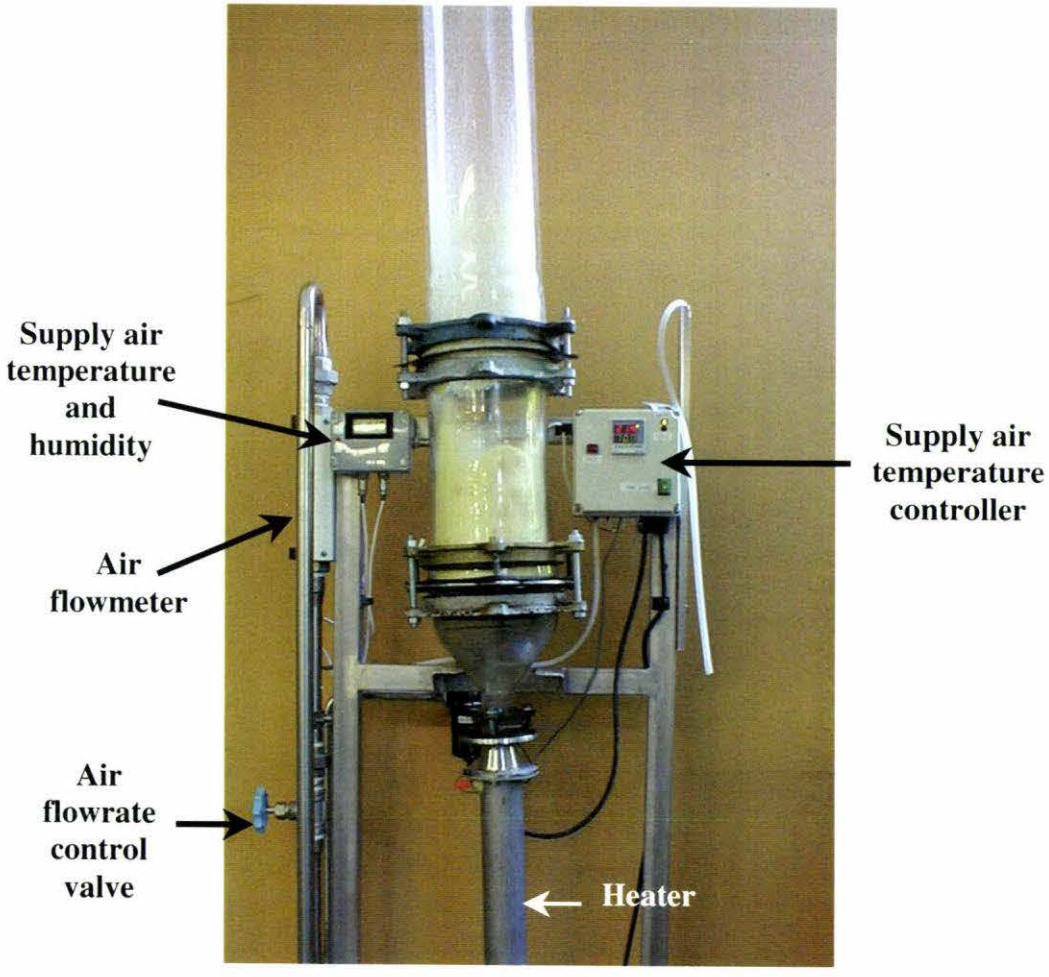


Figure 3E Fluidised bed drier

Air is introduced through a heating chamber situated under the main drying chamber. The inlet air temperature is controlled by a PI controller attached to the heater. The

airflow was manually controlled though a screw valve and monitored by a rotameter. The central glass chamber of the drier could be taken out to remove or replace product after drying.

The conditions for the fluidised-bed drying run were an air flowrate of 200 litres per minute at 30 °C for two hours. The conditions for open air drying were an air temperature between 8 °C and 20 °C with a relative humidity of around 50 %. The moisture content of the granules was 11 % total moisture before the drying, 4.2 % after the fluidised-bed drying and 8.2 % after the open-air drying.

The particle size distribution was found to change during drying when using the small drying chamber. Figure 3F shows the change in size distribution attributed to fluidised bed drying by comparing a sample dried by fluidised-bed with a sample from the same granulation batch left in the ambient air for 7 days. The size distribution was determined through hand sieving.

As can be seen from figure 3f, fluidised bed drying appears to increase the amount of medium sized particles (between 500 and 1000 μm). This is at least partially due to the agglomeration of the smaller particles to form larger particles or layering of smaller particles onto larger ones. This observation can also partially be explained by the loss of fines less than 150 μm in diameter through the mesh hat of the fluidised bed drier. However, the proportion of larger particles is roughly the same for both samples so this cannot completely explain the difference.

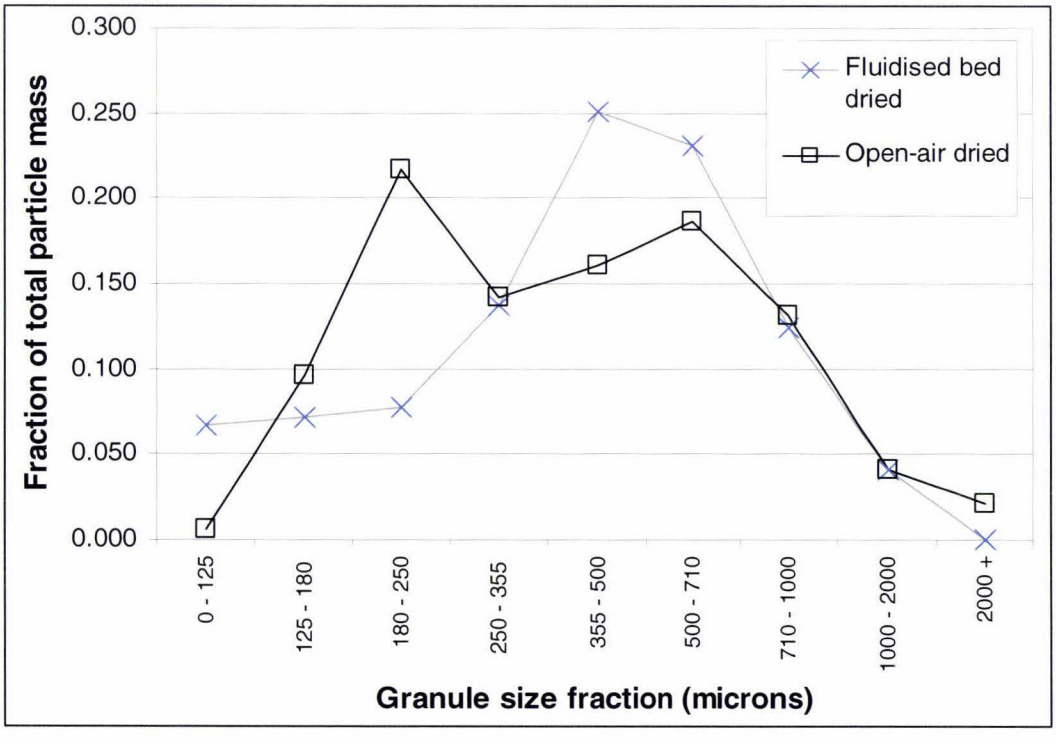


Figure 3F Fluidised bed drying effect on particle size distribution

The granules that came out of the fluidised bed dryer appeared to be more rounded than before the drying. This apparent reduction in asperity would add to the reduction

in surface area due to size enlargement of the particles. Ths may affect the functional quality of the granules by reducing the rate of dispersion during reconstitution.

3.4.3 Glatt fluidised bed dryer

A Glatt Uniglatt fluidised bed dryer was used to produce a drying curve for the granules. The dryer was situated at the DRI pilot plant, Palmerston North. Figure 3F shows the set up of the Glatt dryer.

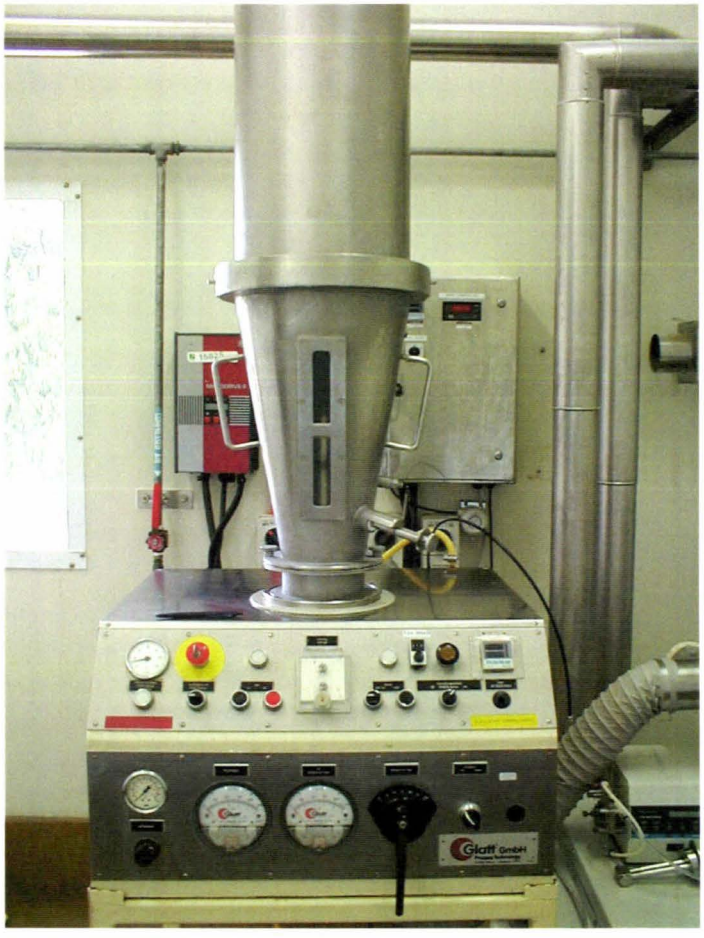


Figure 3G Glatt fluidised bed dryer

A vacuum draws air through the drying chamber and the filter sock. Samples of granules were removed at certain time intervals for moisture analysis and this data was used to determine the batch drying curve (BDC) for the granules. Details of this work are found in chapter 6.

3.4.4 Moisture content analysis

The total moisture content of the granules was determined by drying the granules at 105 °C for at least six hours. Bronlund (1997) states that under these conditions the water of crystallisation of lactose of α-lactose monohydrate is not completely removed; a temperature of 120 °C is required to do this. The amount of moisture removed from α-lactose monohydrate when dried at 100 °C for 2 hours was found to be around 0.25 % (g/g dry solid) compared with around 5.5 % (g/g dry solid) when dried at 120 °C. This suggests that the moisture content determination of milk powder or granules that contain crystallised lactose may have some error in the

measurement. The amount of crystallised lactose present in the milk granules that were dried in this project is unknown.

An alternative to the oven method was the Karl-Fisher moisture content test to determine the total moisture content. This was carried out by NZDRI staff using a Mettler DL18 Karl Fischer titrator in accordance with the NZDRI milk powder technology methods II test number MP-MT-KF01. The Karl-Fisher method of moisture analysis removes all of the chemically bound moisture including the water of crystallisation (NZFRC Milkpowder Methods II). The variation in measurements of the same sample between the 105 °C oven method used here and the Karl Fischer method was found to be less than 0.05 % total moisture over a range of powder and granule moisture contents. This suggests that the difference between the 105 °C oven method and the Karl Fischer method of moisture analysis was not significant and could be used interchangeably.

3.5 Other equipment used

3.5.1 Viscometer

In an attempt to ensure reproducibility of binder between runs, the viscosity of the milk concentrate was measured. A Paar Physica Rheolab MC1 rheometer was used with a number 2 spindle under a constant shear rate of 350 s^{-1}. The measurement of viscosity was of little use however as milk concentrate is non-Newtonian, having age thickening and shear thinning characteristics as well as the problem of protein gelation during storage (Caric, 1994, Pisecky, 1986). This meant that the viscosity would vary depending on the time it took to take the measurement, a factor that was difficult to control.

3.5.2 Milk Reconstitution

Milk concentrate was made by slowly adding spray dried milk powder to water pre-heated to 50 °C. Figure 3G shows spray dried milk powder being reconstituted.

Figure 3H Milk Powder being reconstituted

A mixer blade was used to agitate the liquid at the maximum speed possible before bubbling (and hence foaming) occurred. The addition of the powder took approximately thirty minutes and was followed by a gentle mixing period of a further thirty minutes. After this, the liquid was sieved though a 210 micron sieve to remove any lumps and to reduce the foam. The concentrate was then used straight away to avoid age thickening.

3.5.3 Mercury Porosimeter

The pore size distribution of granules was measured by using a mercury pore sizer, made by Micrometrics, model number 9300. The pore sizer measures the intrusion of mercury into the granules over a range of pressure settings. The pore size at each pressure is then calculated using the Washburn equation (Marshal, 2001).

3.5.4 Water Activity Measurement

The water activity of powders was recorded using a humidity meter attached to an airtight container containing the powder to be measured. Figure 4 shows the experimental set up.

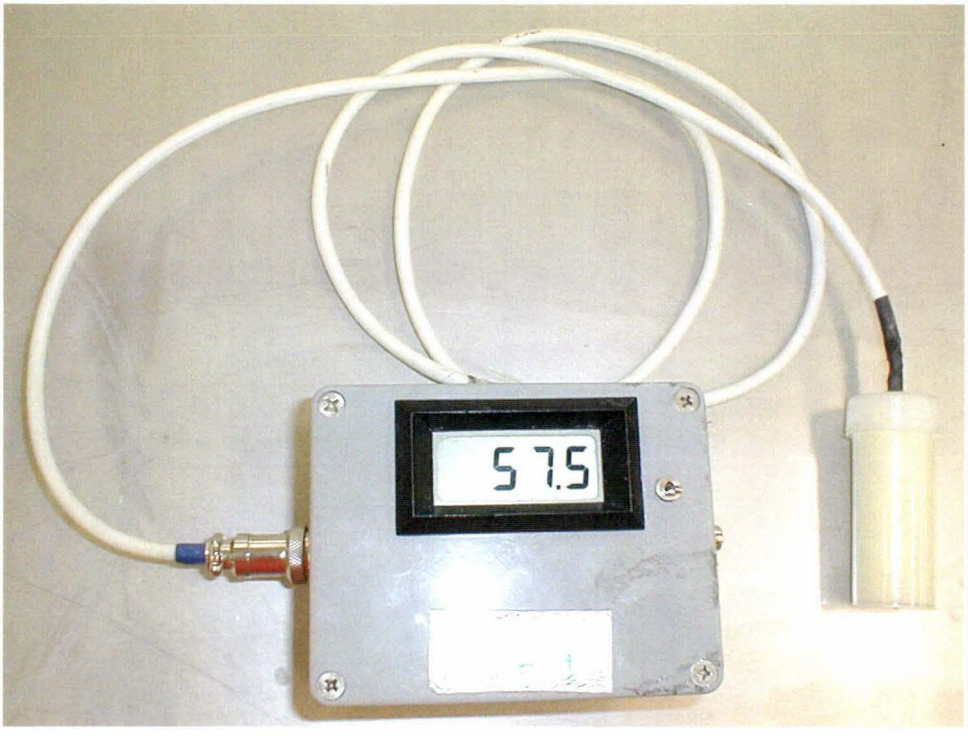


Figure 3I Water activity measurement

The water activity is measured by reading the humidity of the air filling the pores in the container. Each sample was read after the measurement was stable for at least five minutes after being left for at least one hour to equilibrate. Bronlund (1997) states that the system should be left for three hours to reach steady state using this method of measuring water activity although there was no significant difference found when the two methods were compared.

3.6 Laboratory Testing Methods

3.6.1 Particle size analysis

3.6.1.1 Sieving

Sieving was used to analyse the particle size distribution or for separating out specific size fractions of granules for further analysis. Originally, an Endecott test sieve shaker model EFL 1 Mk 11 was used for 10 minutes at fifty cycles per second for each sieve run. Either 100 mm diameter or 200 mm diameter stainless steel sieves were used depending on the amount of granules to be sieved.

Sieving for size analysis was only possible under instances where the granules were dry and hard enough to resist attrition. When granules were made in less than 5 minutes or with moisture contents less than the optimal (see chapter 5), the granules were porous and brittle, resulting in attrition during sieving. This caused the sieves to block, reducing the sieving efficiency, and resulted in an inaccurate measurements. Sieving of undried granules resulted in granules sticking to the sieve, blocking the sieve holes and caused the fines to become agglomerated into the larger granules. Sieving for particle size analysis was therefore undertaken by gently shaking by hand each sieve individually for five minutes. This technique took considerably longer but

gave much more accurate results. All reported particle size analysis results that used sieving were obtained using the hand-sieving technique.

3.6.1.2 Particle Size Analysis using the Malvern Mastersizer

Particle size testing was also undertaken using a Malvern Mastersizer made by Malvern Instruments. This work was carried out by NZFRC staff at the NZFRC powder testing laboratory, Palmerston North. The Malvern was used to determine the size distribution of both dried particles and suspended particles for reconstituted milk. The disadvantage of using the Malvern is that it could not be used for particles over 2 mm in diameter. This meant that the granules larger than 2 mm had to be hand sieved and removed before measurement could begin. This introduced an inconsistency into the results as the Mastersizer measures the volume percentage of each particle size fraction while sieving attempts to measure the diameters of particles.

3.6.2 Functional testing using NZDRI standard tests

A series of functional tests were performed on granules to test the effect of granulation on milk powder. The results of this work are shown in chapter 6. Table 3A shows a summary of these tests and the relevant NZFRC reference.

Table 3A Summary of NZFRC powder and granule functional tests

Test	Description	Reference	Methods Manual Number
Solubility Index	Determines how much insoluble material is present in a 50 ml sample to give an indication of the chemical change of a powder	MP-MT-SI01	I
Bulk Density	The density of the powder or granules including inter-particle spaces when packed into a standard size container	MP-MT-BD01	I
Particle Density	The density of the particles including particle pores but excluding inter-particle spaces	MP-MT-PD01	II
Karl-Fisher Moisture Determination	Determines the total moisture content of a sample including the chemically bound water	MP-MT-KF01	II

The methods and equipment used are described in NZFRC Milk powder technology methods manual I and II.

Chapter 4 - Development of Experimental Methods

4.1 Introduction

An existing pilot scale high-shear mixer-granulator was modified for this project. This chapter describes these modifications which involved improvement of the spraying system and altering the granulator bowl so that condensation did not occur during the binder addition phase. A section on the methodology used to determine the best binder reconstitution procedure is also included. This work has been kept separate from the other descriptive and experimental chapters because the developmental work described is not only useful in determining an experimental set-up that would allow for successful granulation using the present granulator but also for discovering the conditions under which granulation could be carried out in other systems at a larger scale.

4.2 Development of Spraying Device

4.2.1 Nozzle selection based on spray pattern and flowrate

The flowrate of the spray through a spray nozzle depends on the pressure drop that is observed across the tip of the nozzle as well as the viscosity and surface tension of the liquid (Masters, 1991). The viscosity and surface tension will depend on the temperature, age and the history of processing of the milk concentrate (Early, 1998 and Kessler, 1981). In the experimental work for this thesis, the temperature, solids concentration and pressure drop across the spray nozzle were controlled. The freshest milk concentrate with the minimum processing was used to minimise any other variables. The temperature of the concentrate was chosen to be 50°C.

Three spray nozzles were tested for flowrate and ability to handle milk concentrate. The nozzles were made by Spray systems Ltd. Auckland, and had approximately 170, 230 and 500 micron sized orifices that produced a flat spray. The flat spray. by observation, proved to be suitable as the droplets were distributed evenly across the powder bed. The orientation of the spray could be altered to maximise the width of the spray, resulting in a good distribution of spray droplets into the powder bed.

The flowrate of water through the spray nozzles was tested at a pressure drop of 6 bar. This was the maximum pressure consistently available from the compressor that was used for most of the experimental work. A binder volume of 300 ml was used for the test as this was estimated to be the amount of binder that would be used during granulation. For simplicity, water at ambient temperature (approx 15 °C) was used for the test: water at this temperature was found to have a similar spray flowrate to 30 % solids milk concentrate heated to 50 °C. It was found that the 230-micron size spray nozzle had the most suitable spray rate, spraying 300 ml of water in approximately 30 seconds. The 500-micron nozzle took around 5 seconds to spray 300 ml and the 170-micron nozzle took over 10 minutes. The smaller nozzle was also tested using 20 % solids milk concentrate and was found to block very easily, giving further reason not to use it.

A binder spraying time of around 4 or 5 minutes is needed for adequate granulation (Stringer, 2001) meaning that the 230 micron nozzle, at 30 seconds, was faster than

required. The flowrate can be lowered by reducing the pressure drop across the spray nozzle. However, this will also reduce the quality of the spray by altering the droplet size distribution and leading to larger droplets (Kufferath *et al*, 1999), and possibly increasing the chance of the pressure nozzle blocking during operation. It was therefore decided to use the maximum available pressure drop for granulation.

4.2.2 Determining Spray Distance and Pattern

The distance from the spray nozzle to the powder bed will affect the separation of the spray droplets when hitting the bed as a spray naturally spreads out as is moves away from the nozzle. Therefore, it is logical to have the spray as far away as is practical from the powder surface to reduce the chance of droplets overlapping (Hapgood, 2000). The maximum distance between the nozzle and the bed depends on the dimensions of the powder bed and the characteristics of powder flow at the bed surface. The spray must not hit the walls as sticking and caking can cause significant product loss. Milk powder is hygroscopic and will become sticky when wetted so the powder will tend to stick to the walls and blades of the granulator if the spray wets the walls.

The spray was found to absorb into the powder bed quickly when the granulator blades were moving and it was proposed that under the correct blade speeds, spray pattern and binder flowrate, sticking of powder to the walls could be minimised. Therefore, it was important to characterise the spray to determine the spray pattern and to determine how far the spray nozzle should be from the moving powder bed to maximise the efficiency of the spray and to reduce any sticking that might occur.

A 'snapshot' of the spray was taken under various conditions to determine the width and the dispersion of the spray at various distances from the spray nozzle. The equipment consisted of a sliding plate with a vertical slit in the centre of the plate. The plate is pulled horizontally through its frame by dropping a weight attached to the side of the plate over the edge of a table, as shown in figure 4A. The spray is allowed to run for 10 seconds before the weight is dropped to allow a steady stream to develop. The slit through the plate exposes a piece of transparent sheet taped to the frame to a 'snapshot' of the spray pattern, allowing individual drops to be seen as dots on the sheet.

Figure 4A shows a diagram of the experimental set up. A 9.45 kg weight was used to pull the plate and this was dropped onto a marking on the ground from the same height each time in an effort to reproduce the speed of the slit as it passed through the spray. The string attaching the weight to the slide was loose before the weight was dropped so that the slide was pulled with a short and sharp impact rather than a gradual increase in speed. The weight fell approximately 100 mm freely before the string attached to the slide became taught. This method gave a faster slide velocity meaning that a better image of the spray pattern was possible. Tests in which the weight was dropped with the string tight meant that the spray pattern was difficult to visualise due to droplets overlapping as the slit moved more slowly across the spray.

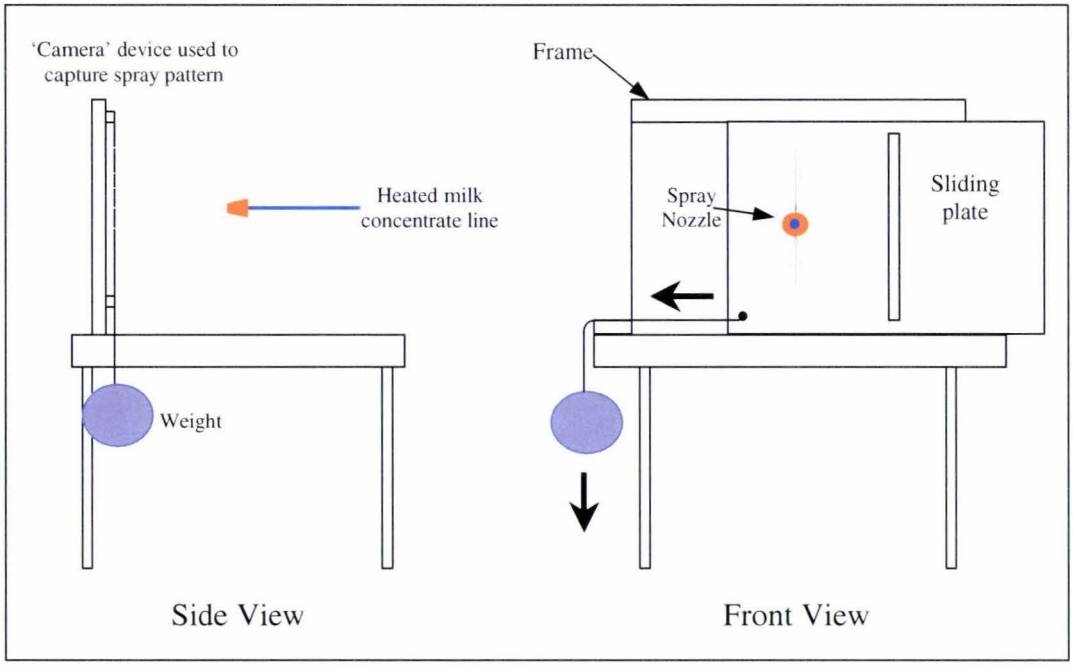


Figure 4A Experimental set-up for recording spray pattern

A transparent cellulose acetate sheet was found to be the most suitable means of recording the spray pattern as the milk did not soak into the sheet. However, the viewing of the spray pattern was difficult due to the semi-transparent nature of milk and the light colour.

An attempt was made to improve the contrast of the spray droplets through both dyeing the milk with blue food colouring and through staining the cellulose acetate sheet containing the spray profile with Safranin and Crystal violet stains after the spray pattern had been recorded. Both methods improved the contrast of the spray droplets although staining after the spray had dried proved to be difficult due to extra stain droplets affecting the spray pattern. It was difficult to stain the sheet without washing off some of the milk droplets when rinsing the stain. A better quality image was produced through dyeing the binder although the viscosity and surface tension of the milk concentrate may have been affected. It was assumed that the effect of adding the dye had minimal effect on the physical properties of the spray.

Table 4.1 Spray pattern footprint at varying distances

Spray Distance	Length of spray footprint	Width of spray footprint	Overlapping of droplets
5cm	100mm	15mm	Over most of spray
7cm	120mm	25mm	Sides and centre only
9cm	150mm	40mm	2 small areas at the sides

Table 4.1 shows the spreading of the spray and the extent of the overlapping of droplets at each distance. As can be seen, the spray has less overlapping of droplets

with increasing distance. These results also show that the spray pattern is not even and a higher concentration of spray is found at the center of the spray.

The distance from the spray nozzle to the moving powder bed in the granulator was found to be approximately 7 cm although it was later found that the powder 'bulks up' during granulation and the distance is reduced to around 5 cm. It was also observed that the spray penetrates the powder bed better from close range meaning that the spray is mixed into the powder better as the inertia of the powder aids the mixing. However, overlapping of droplets caused by short spray distances is likely to affect the particle size distribution of the granules and result in poor moisture distribution.

4.2.3 Spray angle

The spray system shown in figure 3D was adjusted to allow the maximum distance between the spray nozzle and the powder surface. The S-shaped design of the feed line was required as the impeller shaft bearing on top of the granulator restricted the placement of the hole through the top of granulator for the binder feed line. The S-shaped design allowed the spray nozzle to be closer to the centre of the granulator giving a better angle onto the powder surface and meant that the spray nozzle angle could be easily adjusted. The angle of the spray nozzle was approximately 30° from the vertical and was sprayed directly towards the toroidal powder flow surface.

Optimisation of the spray angle was carried out through trial and error by observing the flow pattern of the powder whilst dry. The best spray angle was a combination of two factors; firstly, to direct the spray at a right angle to the powder to reduce splashing of the powder, and secondly, to have the spray directed diagonally across the face of the toroid as shown in figure 4B. It was found that having the spray hitting the powder at any angle less than perpendicular to the powder bed resulted in powder splashing around the inside of the granulator. This resulted in powder adhering to the walls of the granulator and being wasted. By directing the spray diagonally it is possible to minimise the overlapping of droplets and also increase the area over which the spray can land. However, this meant that the distance between the spray and the powder surface varied over the length of the spray so that at one side (the high side of the spray) the spray was closer to the powder surface compared with the other (lower) side. It is unclear whether this had an effect on the granule size distribution although using this method produced the minimal amount of powder sticking to the sides of the granulator.

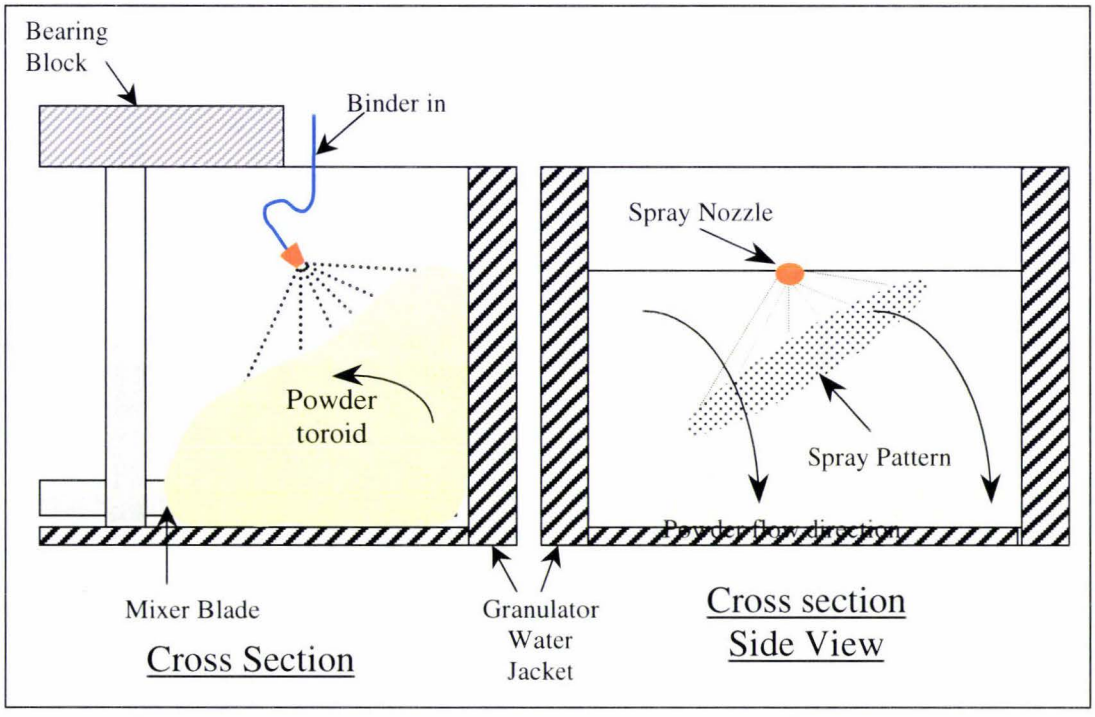


Figure 4B Spray angle and spray footprint (not to scale)

4.2.4 Spray timing

As described in section 4.2.1 the flowrate of the binder through the spray nozzle was found to be faster than desired. The overall flowrate could be reduced by pulsing the binder in a similar manner to Shaafsma *et al* (1999), where fluidised bed granulation was studied by controlling the binder pulse length and flowrate. However, pulsing binder into the granulator may pose difficulties as the drop in pressure across the spray nozzle will reduce the quality of the spray and cause large drips to form. This was observed when milk concentrate was pulsed into the granulator. In addition, powder in the granulator often adhered to the drips between the pulses, affecting the spray pattern and causing the nozzle to block.

Air was blown through the spray system between the pulses to clear the binder delivery line of milk concentrate and to prevent nozzle blockage. This required having two valves on the binder delivery line as shown in figure 3C. The timing of the pulses was chosen through trial and error. A ten second pulse of binder was selected. Pulses longer than 15 seconds reduced the distribution of the binder by over-wetting the surface of the powder bed and resulted in more product becoming stuck to the walls.

The torque on the mixer blade was found to increase during each binder pulse and pulses 10 seconds or less resulted in a manageable increase in torque that dropped after the pulse. This is shown in figure 4C, after an initial dry mixing phase. The increase in torque occurs for more than 10 seconds because the timing of the pulse is based on how long the binder delivery valve is open. After the binder delivery valve is closed, the air purge takes a further 5 – 10 seconds to clear the delivery line of binder depending on the binder temperature and pressure which will vary depending

on air temperature, the temperature of the delivery line and fluctuations in the compressor pressure.

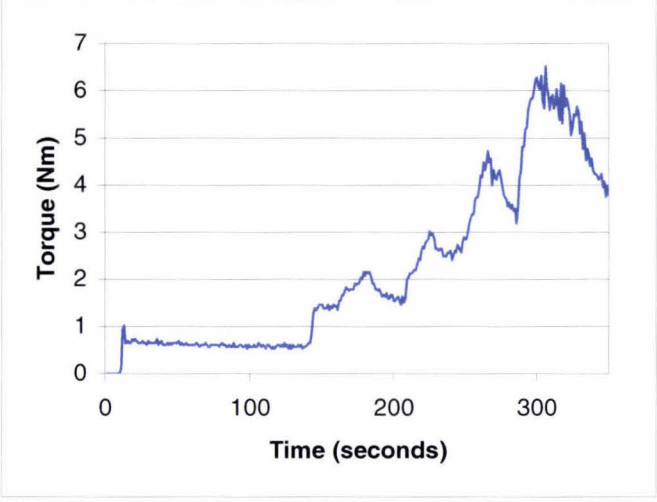


Figure 4C Torque profile for mixer drive shaft during binder addition

One of the problems with this technique is that it requires the operator to follow a timer and to synchronise the opening and closing of valves. As the potential for error increases with the speed of switching, the pulse duration was chosen to be 10 seconds to minimise this operator error.

To remove the possibility of operator error from the binder addition an automated binder delivery system is suggested for future work. This would improve the control and reproducibility of the granulation as well as allowing shorter binder pulses to be used. A three-way automated valve or two automated valves linked to open and close simultaneously could be used. A three-way automated air valve was tested at the end of the experimental work and it was found to be unsuitable as the binder liquid leaked through the valve when the valve switched to an air pulse. A robust valve design would therefore be required that could handle both liquid and air and remain sealed at the pressure drop required.

To test the optimal pulsing length, a milk concentrate dyed blue was sprayed into the granulator while running at the binder delivery speed. The dyed binder could easily be seen in contrast to the powder. The time taken for the blue binder to colour the full surface of the powder bed was between one and two seconds. This means that pulsing the binder in using one second bursts could prove beneficial to the granulation. Care would have to be taken to ensure that there was not a periodic relationship between the pulsing rate and the circulation time of a particle about the toroid and around the mixer so that one area of the bed received a disproportionate amount of binder.

4.2.5 Using a higher pressure to spray

The pressure for the spray system provided a pressure drop of around 6 to 7 bar. This was suitable for granulation however it may be possible to improve the spray, and possibly narrow the granule size distribution, by increasing the pressure of the spray. A compressor with the ability to provide up to 10-bar pressure was used for one granulation run at the end of the project. As expected, the spray was finer than using

a 6-bar pressure drop and it was even found that the milk concentrate could be sprayed through a spray nozzle of 170-micron diameter without blocking the spray nozzle.

In the test of the higher spray pressure the air to purge the binder delivery line was taken from a 6-bar compressor. This was needed to prevent the pressure in the reservoir of the 10-bar compressor from dropping too quickly, as the compressor did not have a high capacity. This meant that any operator error in the timing of the valve switching could cause air or binder to flow up the lower pressure air line, dirtying the air supply and valves. An instantaneously actuated valve system is necessary to ensure pressure drop between the binder delivery and the purge air is minimised. Therefore, a fast-acting non-mixing valve system would be desirable for further research using this design.

4.3 Binder Reconstitution

4.3.1 Introduction

Most of the binder solutions were made by reconstituting milk powder and the development of a procedure that was repeatable was required. During the course of the experimental work the concentration of the milk concentrate was increased from 30 % to 40 % total solids due to improvements in the technique used to reconstitute the binder. The concentration used for the binder used in experiments producing granules for functional testing was 40 % solids. It is possible to reconstitute milk up to 50 % solids. However, it was found that above 40 % solids the physical properties of the concentrate changed so rapidly that handling of the concentrate was difficult. Table 4.2 shows the increase in viscosity of increasing solids concentration for milk concentrate found using a Brookfield viscometer at 50 °C and a shear rate of 330 s^{-1}.

Table 4.2 Increasing viscosity of milk concentrates at a shear rate of 330 s^{-1}

Solids concentration of milk concentrate	Viscosity (mPa.s)
25 %	2.43
31 %	4.48
36 %	6.91
47 %	22.5

An increase from 36 % total solids to 47 % shows a large increase in viscosity resulting in increased likelihood of problems during granulation. The main problem that could occur is 'setting' of a skin on the concentrate between pouring the binder into the binder delivery vessel and starting the spray. With an increase in viscosity there is also an increasing chance that some of the binder will stick to the binder delivery equipment as a film and not be sprayed. This may result in blocking of the spray nozzle when parts of the film are sloughed off.

4.3.2 Reconstitution Methodology

The reconstitution method was developed through trial and error and was not specifically studied apart from observations during other experiments. Initially, the

method involved slowly adding the correct amount of powder to pre-heated water that was mixing gently. The water was mixed gently to reduce the generation of foam, which will occur when fat leaks out of fat globules and coats an air bubble (Kessler, 1981). Theoretically, more foam will occur with an increase in shearing because of the increase in 'free fat' and air bubbles formed. Under these conditions it took 40 minutes to an hour to add all of the powder. Even so, sieving still caught a number of lumps of milk solids. These lumps were attributed to the powder aggregating while being added to the water and not being broken up by the impeller blade.

A number of impeller blade designs were trialed; a summary of these and the observed mixing patterns are shown in table 4.3. Reconstitution was carried out in a 2 litre plastic container approximately 150 mm in diameter.

Table 4.3 Impeller design and success of reconstitution

Size	Design	Blade diameter	Blade width (if flat)	Bubbles at low speed? (180 rpm)	Bubbles at high speed? (360 rpm)
Small	round 3-blade propeller	25 mm	-	no	no
Standard	flat 2-blade propeller	80 mm	15 mm	no	minimal
Large	paddle	115 mm	40 mm	minimal	yes

The large impeller was initially used at low speeds but when the impeller speed was increased in an attempt to break up the lumps the smaller blades had to be used to stop excess foam forming. The smaller blade did not provide enough turbulence to mix the concentrate well, even at the maximum speed of the mixer. The standard size impeller was then used for subsequent milk concentrate reconstitution.

Reconstitution was improved by increasing the mixer blade speed whilst tilting the impeller at an angle and offsetting it from the centre of the container. This reduced the vortex formed and produced far fewer bubbles. It also resulted in fewer lumps. The best position for the impeller was found by running the mixer at a fast speed and moving the impeller around to determine the location that provided the least amount of bubble generation before the powder was added. The impeller was then slowed while the first third of the powder was added. The mixer speed was then increased as the rest of the powder was added. This was necessary as the volume of the liquid was less at the start of the reconstitution and increased as the powder dissolved. The final improvement was to leave the concentrate mixing with the mixer running at a slow speed for 30 minutes, which allowed for further dissolution of solids. After this procedure, no solid material was found on the sieve.

4.4 Humidity of the Granulation

Powder build up on the walls of the granulator bowl was a significant problem early on in the experimental program. In these early experiments the bowl was sealed so that there was no mixing of the air in the granulator with the external air. This meant that the air in the bowl quickly became saturated during binder addition. Because the

walls were water cooled, this resulted in water condensing onto the interior wall surfaces of the granulator. This meant that powder hitting the walls would stick rather than rebound as would happen with perfectly dry walls.

This problem was solved by exposing the granulator to the outside atmosphere by taking the lid off a viewing port. In addition, blowing air into the granulator between binder pulses aided the reduction in product wastage by allowing a flow of humidified air out of the granulator and reducing the powder sticking to the walls. A further reduction in product loss was found by polishing the granulator walls and mixing blades before a run was carried out with a dry piece of tissue. Under ideal cases where the granulation conditions were optimal (see chapter 5) and these precautions were observed a negligible amount of product loss occurred.

The investigation into the humidity of the granulator during granulation was undertaken in an attempt to estimate the surface temperature of the granules by assuming the granules were in equilibrium with the air inside the granulator. The temperature could then be estimated by the use of a psychrometric chart. This assumption of equilibrium is unlikely to hold due to the hygroscopic nature of milk powder. The saturation of the air followed by condensation of water onto the granulator walls (and the humidity probe) also meant that measurements of humidity could not be used to estimate the temperature. Using humidity to investigate granulation mechanisms may be possible if the humidity of the system does not reach saturation such as in the work by Schaafsma et al (1999).

Chapter 5 – Granulation of Whole Milk Powder

5.1 Introduction

This project is the first known attempt to produce milk powder by a granulation process using milk as the sole ingredient. The principal relies on spraying milk concentrate onto milk powder that has already been granulated and dried. Screening then separates a product fraction and a recycle.

This chapter focuses on the batch high-shear granulation process, the operating parameters that successfully yield granules, and the sensitivity of the product to changes in these parameters. The results described here are useful when considering scaling to larger batches or to continuous granulation processes. Exact scaling laws have not been established for high-shear granulation, even when scaling between batch sizes, and certainly not for scaling from batch to continuous processing. Therefore scale up will necessarily require pilot scale experimentation.

The optimal process conditions to granulate whole milk powder will be judged on the quality of the granules produced. In a granulation circuit, where out-of-specification product will be returned to the process, the most important quality of the granules will be the size distribution. An ideal granulation produces granules that have a desirable mean size with a narrow size distribution. The effect of process conditions on the reconstitution and storage quality of the granules will also be important and will be covered in chapter 6.

5.1.1 Process Selection

High-shear granulation is the best available process. Chapter 2 contains a lengthy account of high-shear granulator performance. It's potential advantages over spray drying are:

- intensified process
- small footprint
- lower capital cost

These reasons justify the present investigation.

5.1.2 High Shear Granulator

The high-shear granulator described in chapters 3 and 4 has a range of operating parameters shown in table 5.1.

Table 5.1 Granulation parameters to be tested

Parameter to be tested	Possible range of testing	Actual values tested
Mixer speed	0 to 1450 rpm	524, 700 and 900 rpm
Chopper speed	0 to 7000 rpm	1250, 1324, 1900 and 2500 rpm
Amount of binder addition	0 to amount where over-massing occurs	121 g, 175 g, 204 g and 233 g into 1.5 kg of powder to achieve 8, 10, 11 and 12 % total moisture of granules
Length of granulation after binder addition	0 minutes +	2, 5, 10, 20 minutes
Binder solids concentration	0 to ~60 % total solids	40 % and 50 % using evaporated milk or 30 % and 40 % using reconstituted milk

Other parameters have an impact on the granulation, but were not tested in this work. These include:

- dimensions of the chopper blade
- dimensions of the mixer blade
- bowl dimensions
- inclusion of a cone insert on the lid of the granulator
- spraying system – multi spray or finer spray designs
- cooling system – using a cooler water jacket temperature

This project used a single design of mixer and chopper blades, a bowl of cylindrical dimensions with a single spray and cooling system as described in chapter 3.

5.2 Achieving Successful Granulation

5.2.1 Experimental Set-Up

The binder to powder ratio is the most significant parameter in any granulation (Ennis and Litster, 1997). For this reason, preliminary experiments examined this ratio at moisture contents of 8%, 10% and 12%, with all other variables held constant as given in table 5.2. The moisture content was calculated as the total moisture weight to total solids weight. The total moisture weight includes the moisture associated with the powder and the milk concentrate.

The amount of powder added to the granulator was initially tested by using the same mass used for previous work on granulation with calcium carbonate (Hamilton and Jones, 2001). This was found to be too high for milk powder as the powder had a

lower bulk density than calcium carbonate and 'bulked up' during binder addition and resulted in the surface of the powder contacting the spray nozzle. A powder load of 1.5 kg was the most suitable amount of powder as the powder flowed well during dry mixing and granulation and there were no problems encountered due to 'bulking up' of the powder during binder addition.

The blade speed of the mixer and chopper were chosen by running the granulator with dry powder and choosing speeds at which the powder seemed to flow well, with limited splattering of powder around the inside and onto the lid of the granulator. Other experimental conditions are described in detail in chapter 4.

Table 5.2 *Operating conditions for preliminary experiment to determine correct binder volume.*

Mixer speed:	524 rpm
Chopper speed:	1324 rpm
Amount of milk powder added:	1.5 kg
Binder solids concentration:	30% (total solids content) reconstituted whole milk powder
Amount of binder added:	121g (8 %), 175g (10 %), and 233g (12 %)
Binder added after:	two minutes of dry mixing
Length of granulation:	10 minutes granulation after binder addition

Table 5.3 gives qualitative observations for the experiments at 8, 10 and 12 % total moisture. Granulation occurs between 10 and 12% total moisture. At 8% total moisture the product was still powdery with no visible granules present. At 10% most of the product was still powdery with a few granules produced that were more round and larger than the feed powder particles. At 12% most of the product was either granulated or stuck to the sides and lid of the granulator. Significant build up of caked milk solids occurred under the mixer and chopper blades resulting in browning and a strong cooked smell.

Table 5.3 *Observations for determining optimal binder amount.*

	8% total moisture	**10% total moisture**	**12% total moisture**
Granules produced?	No	Few small granules	Yes
Product loss	No	Minimal	Significant
Smell	Slight	Slight	Strong
Colour change?	Minimal	Minimal	Noticeable

In conclusion, granulation of whole milk powder occurs at a moisture content between 10 % and 12 %. However, it must be remembered that this is specific to the conditions shown in table 5.2. The following sections of this chapter detail the effect of other variables on the granulation and examine the size distribution of the granules produced as a measure of the granulator performance.

5.3 Optimising mixer speed, chopper blade speed, and the amount of binder

A three-factor experiment was performed to evaluate the importance of the mixer speed, chopper speed, and the amount of binder added to the granulation. Three factors were chosen to keep the number of experiments to a manageable number. The levels of each parameter are shown in table 5.4. The constant parameters in this table are investigated in later sections of this chapter.

The amount of binder added to the granulation is the most important parameter in any granulation system. Therefore, the amount of binder was given three treatment levels. The chopper and motor speeds are likely to affect the granule attributes such as granule size distribution and density (Jones *et al*, 2002), but not likely to affect the success of the granulation so were limited to two levels of treatment. The experimental design is given in table 5.5. The order of the experiments was randomised to minimise time dependent trends in data due to extraneous sources such as ambient temperature or humidity.

Table 5.4 Experimental conditions for three-factor investigation into mixer speed, chopper speed and binder amount.

Parameter	Is parameter being investigated?	Level(s) Low → High
Mixer Speed	Yes	524 rpm 900 rpm
Chopper speed	Yes	1250 rpm 2500 rpm
Amount of binder*	Yes	175 g 204 g 233 g
Binder temperature	No	50 °C
Granulation time after binder addition	No	20 minutes or if over-massing occurs
Binder solids concentration	No	30 % Total solids
Water cooling	No	Ambient temperature (approx. 17°C)
Dry mixer prior to binder addition	No	2 minutes
Binder addition technique	No	10 second pulses with 30 second air purging

* The binder amount corresponds to final granule moisture content of 10, 11 and 12 % respectively

The experimental procedure involves a dry blending stage, a binder addition stage and a granulation stage. The speed of the chopper and mixer blades were set at values of 800 rpm and 450 rpm respectively during the initial 2 minutes of dry mixing and

during binder addition. Just prior to the granulation stage the speeds were then increased to their set values shown in table 5.4.

The duration of the granulation stage was 20 minutes for these experiments. This is much longer than would be expected in a normal granulation because the effect of time on the granulation was unknown at this stage. The granulation was to be stopped if over-massing occurred. In previous work on granulation of calcium carbonate powder with polyethylene glycol binder, over-massing was observed when a sharp increase in the torque occured (Hamilton and Jones, 2001).

Table 5.5 *Experimental design to investigate the effects of mixer and chopper speeds and the amount of binder used for granulation*

Mixer Speed	Chopper Speed	Binder Amount	Experiment number
Low	Low	Low	3
Low	High	Low	6
High	Low	Low	11
High	High	Low	1
Low	Low	Medium	4
Low	High	Medium	9
High	Low	Medium	12
High	High	Medium	7
Low	Low	High	2
Low	High	High	8
High	Low	High	10
High	High	High	5

The variables observed after each run were chosen to be,

- the mass of loose granules that could be removed from the granulator by lightly brushing the granulator walls with a brush
- the temperature of the granule mass immediately after granulation was stopped
- the particle size distribution
- the moisture content of the granules produced
- colour

5.3.3 Results and discussion

Table 5.6 summarises the results of the amount of granules removed after granulation, the temperature of the granules immediately after granulation and the total moisture content of the granules measured using the 105 °C oven method described in chapter 3.

Table 5.6 *Summary table of mixer speed, chopper speed and binder addition experimental results.*

Run	Chopper Speed	Mixer Speed	Binder	Mass of Granules (g)	Granule Temp (°C)	Measured granule total Moisture Content (%)
1	H	H	L	1353	26.5	10.62
2	L	L	H	1072	29.5	10.91
3	L	L	L	1494	21.0	11.57
4	L	L	M	1178	25.0	12.35
5	H	H	H	276	40.0	7.5
6	L	H	L	1458	22.5	11.17
7	H	H	M	806	45.0	9.21
8	L	H	H	755	43.0	8.65
9	L	H	M	1180	24.0	11.68
10	H	L	H	903	39.0	8.42
11	H	L	L	1462	24.5	11.16
12	H	L	M	1070	32.0	10.60
H = high M = medium L = low levels (see table 5.4)						

The most surprising result from this experiment is the resultant moisture content of the granules after the granulation is completed, as shown in figure 5A. These results appear to be independent of the moisture content that was planned for each experimental run. The granulation runs that used a total moisture basis of 10 % (the low treatment) all resulted in minimal granules being formed but all had a final moisture content between 10.62 % and 11.57 %. These results are difficult to explain.

Two sources of experimental error have been identified that would lead to an under-estimation of the final granule content. Firstly, the moisture content of the original milk powder that was used as feed to the granulator was assumed to be 3.5 % due to estimates given by the NZFRC. The moisture content of the powder was measured after the experiment was finished to investigate the discrepancy in results and it was found to be 4.5 % total moisture. This difference accounts for extra moisture that would make the final estimate of total moisture 11.3, 12.3 and 13.3 % for the expected moisture levels of 10, 11 and 12 % respectively. However, the moisture in the original powder is likely to be chemically bound moisture and may not affect the stickiness and subsequent agglomeration and granulation properties of the powder particles.

The other source of experimental error is in the binder reconstitution. Some difficulty was found in reconstituting the binder to the desired solids concentration during these experiments due to some of the powder not dissolving and necessarily being removed by sieving prior to binder addition to avoid blockages in the spray nozzle. The error

in the solids concentration was investigated towards the end of the experiment; it was found that the actual solids concentration of the binder was between 27.5% and 29% total solids through the use of the 105 °C oven method described in chapter 3. Underestimating the solids concentration of the binder resulted in more moisture being added to the granulation, as the moisture concentration was higher than expected in the binder. This error accounts for an extra 1.5 to 4.4 g of moisture for the low binder quantity treatment. The estimated corrected expected moisture levels are shown in figure 5A that account for higher than planned powder moisture contents and lower than planned binder solids concentrations.

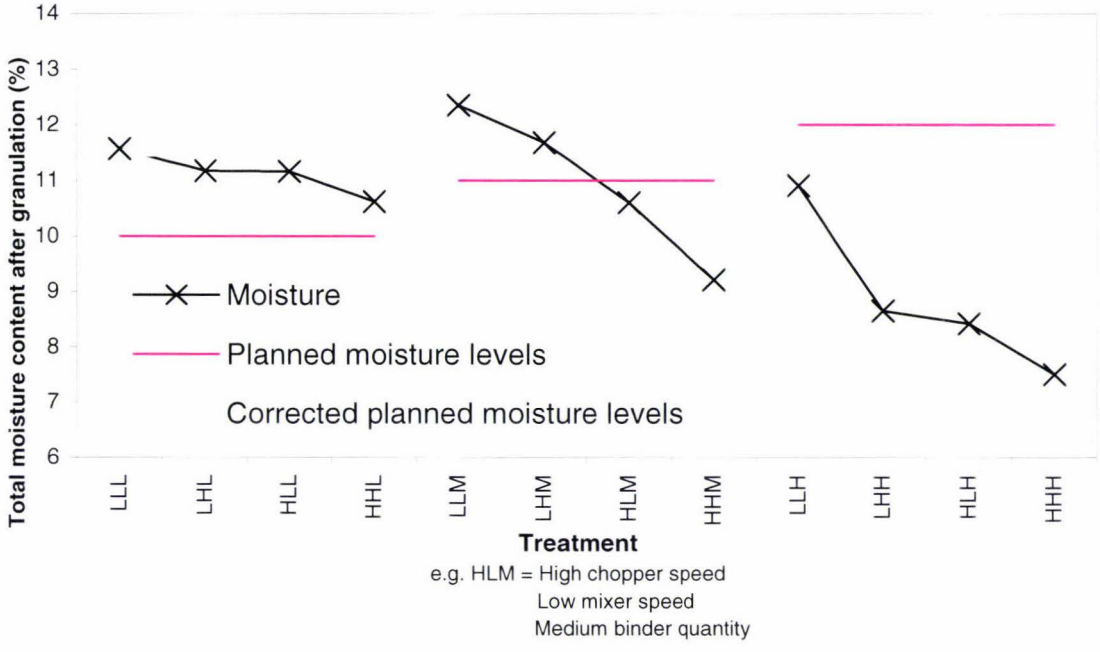


Figure 5A Moisture content for experiments with varied chopper speed (L=1250 rpm, H= 2500 rpm), mixer speed (L=524 rpm, H=900 rpm) and binder amount (L= 10%, M= 11%, H=12% total moisture).

Significant differences are still evident between the corrected planned moisture content and the actual moisture content of the granules. The runs with an expected moisture content of 11 % or 12 % vary significantly in the actual moisture content. This can be explained through evaporation of moisture during the granulation, as the granulator was open to the air with some air flowing out of the granulator with the air purging the binder delivery nozzle. This evaporation was found to be greatest for experiments with both high chopper and mixer blade speeds, because they input the greatest amount of energy into the powder. Evaporation is also higher for runs containing more moisture as the powder is initially stickier which results in more friction and heat generation. Additionally, it is likely that with more moisture added, more surface moisture is present.

Uneven moisture distribution may also contribute to the differences. The runs with a high amount of binder addition had fewer granules removed from the granulator as more powder was stuck to the walls. This powder was not measured but may have higher moisture content than the granules removed from the granulator after granulation. Particles with a high moisture content are stickier and more likely to adhere to the walls. Moisture distribution problems may also be compounded by

moisture condensing on the walls of the granulator due to the effect of the cooling jacket.

Figure 5B shows the graph of the mass of granules removed and the granule temperature at the end of granulation for all runs. The data has been separated into three sub-groups of low, medium and high binder addition to aid understanding.

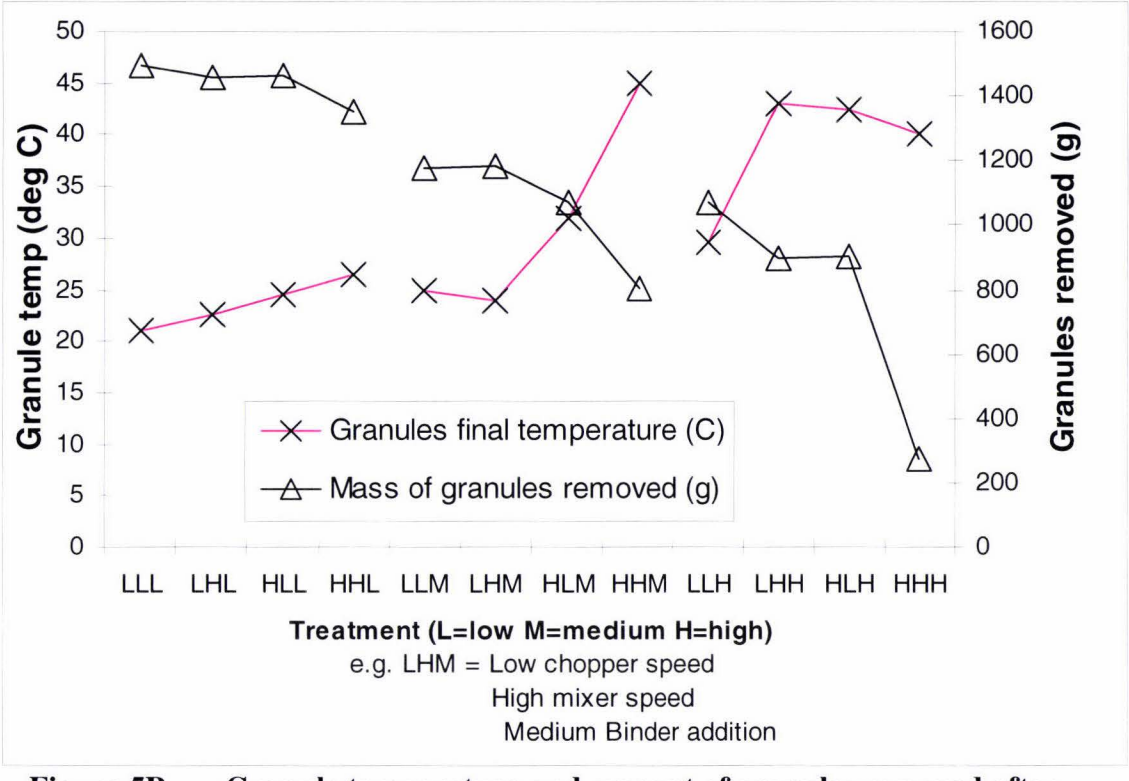


Figure 5B Granule temperature and amount of granules removed after granulation for experiment with varied chopper speed (L=1250 rpm, H= 2500 rpm), mixer speed (L=524 rpm, H=900 rpm) and binder amount (L= 10%, M= 11%, H=12% total moisture).

Figure 5B shows that a decreasing mass of granules removed is correlated with higher temperatures. The remaining powder left in the granulator at the end of granulation was caked on the walls of the granulator. This insulated the granules from the cooling jacket, causing increased temperatures.

Increasing the chopper and mixer blade speeds increases the energy input into the powder which results in a higher final granule temperature. Increasing the chopper speed results in a greater temperature increase, and subsequent decrease in granule yield, than increasing the mixer speed. This can be seen by comparing the HLL and HLM runs with the LHL and LHM runs. This is unexpected as the chopper provides less energy into the granulation than the mixer due to the much smaller blade size (Hamilton et al, 2001). However, this can be explained by the ability of the chopper to throw powder against the walls and lid of the granulator, resulting in caking, reduction in yield and increasing temperature due to insulation of the granulator from water jacket cooling by wall build up. The chopper disturbed the powder flow greater than the mixer at higher speeds during observations of these runs.

Increasing binder content causes an increase in powder stickiness, with a resulting increase in the effects of increasing chopper and mixer blade speeds. This is because greater stickiness increases the chance that powder will stick to the walls of the granulator.

Figure 5C shows a comparison between the particle size distributions of selected granulation runs and the original powder. All show significant size enlargement compared to the original powder. The HLL run (high chopper speed, low mixer speed and low binder addition) is the next finest. It differs from the other runs shown because it has low binder addition whereas the others have medium binder addition. Binder addition is the most significant factor affecting granule size distribution which is apparent when comparing HLL to HLM.

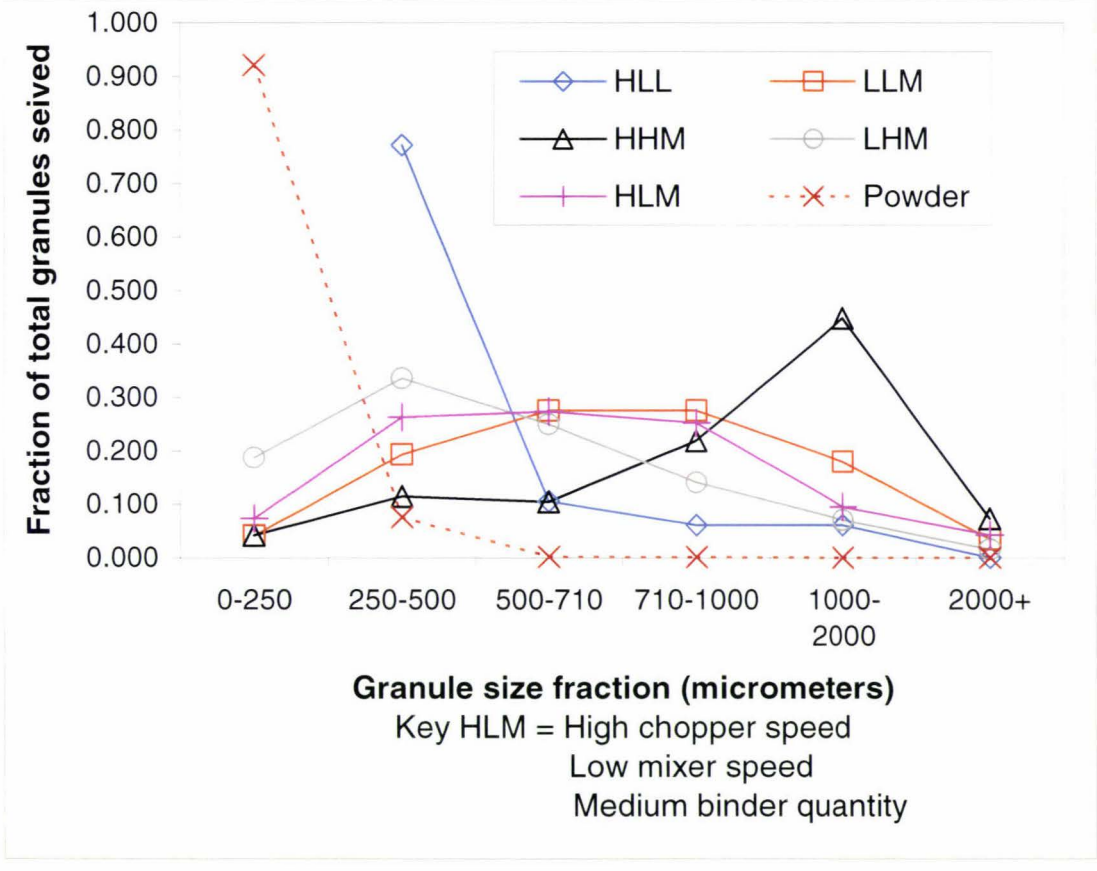


Figure 5C Particle size distributions for varied chopper speed (L = 1250, H = 2500 rpm), mixer speed (L = 524, H = 900 rpm) and amount of binder (L = 175 g, M = 204 g, H = 233g) compared with the original powder.

Comparing HLM to LLM shows that chopper speed has little effect on the granule size distribution. The effect of mixer speed is observed by comparing HLM to HHM (at high chopper speed) or by comparing LLM to LHM (at low chopper speed). In the first comparison the size distribution is coarser with increasing mixer speed; while in the latter, it is finer. This difference is difficult to explain directly although the following general observations of the runs reduce the significance of these results. Generally, the distributions are broad with significant proportions of particles between 250-500, 500-710, and 710-1000 microns. This was not observed in later runs reported in chapter 7, where distributions were much sharper, similar to HHM here. It is possible that attrition occurred during drying, which took 2 hours with 30 °C air in

a fluidised bed prior to sieving. The granules produced during the HHM run, which has the most vigorous blade action, may have been stronger and did not break down as much as the other runs.

Blinding of sieves was also a problem with some runs, particularly those with low binder addition, represented here by the HLL run. This size distribution is the finest of the trials shown but is likely to be even finer than shown because the 0-250 micron and 250-500 micron results are combined into the 250-500 fraction due to blinding of the 250 micron sieve. Figure 5C does not show any results for the high binder addition runs as the granules produced after drying were sticky and blinded even the larger sieves. However, qualitatively their size distributions appeared coarser than for the medium binder addition runs.

Due to the 20 minute granulation time the granules produced were generally more yellow then the original powder, often with some browning of powder that had stuck to the walls of the granulator behind the chopper or mixer blades. The runs containing the low amount of binder (10% total moisture) did not produce granules of a significant quantity while the runs using both medium and high amounts of binder produced granules and had a strong oxidised fat smell. The runs using a high amount of binder (12 % total moisture) had significant sticking of the powder to the walls of the granulator and had brown areas of burnt milk. The runs with the medium amount of binder addition gave the best granulation results with the exception of the HHM treatment where the mass of granules removed was less and the granule final temperature was higher than the other medium binder addition runs due to high blades speeds as already described.

5.3.4 Conclusions

This experiment provided valuable experience for the development of further granulation trials using milk powder. A few qualitative conclusions can be made regarding the chopper speed, mixer speed and the amount of binder added to the granulation.

The amount of binder added to the granulator is of vital importance. These results show that a total moisture content of the wetted powder mass should be around 11 % using 30 % solids milk concentrate as binder for a successful granulation to occur. A total moisture content of 11 % was therefore chosen as the basis of further granulation work.

The effect of mixer blade speed alone on granule size distribution is difficult to determine due to the limited range of blade speeds tested. The granule size distribution seemed only to be affected when both the chopper and mixer blades were set at their high speeds and this resulted in an increase in mean granule size and a narrowing of the granule size distribution. This may be because the measurement of the granule sizes was easier with the granules formed under these conditions as they may have been harder and less prone to attrition during sieving.

A mixer blade speed of 700 rpm was chosen for further work as this was between the two mixer speeds tested and appeared to flow well with minimal turbulence and splashing of powder when dry powder was mixed at this speed.

The effect of changing the chopper speed is also difficult to determine, except for the HHM run as discussed above. The runs using a higher chopper speed all had more powder stuck to the roof of the granulator after each granulation when compared to runs with a lower granulation speed. This suggests that the chopper speed of 2500 rpm was too great for optimal granulation conditions.

A chopper speed of 1900 rpm was therefore chosen for further granulation work as this was between the two chopper speeds tested and, at this speed, powder and wetted granules did not appear to hit the roof of the granulator when observed.

The granulation time used in this experiment of twenty minutes was too long. It compounded problems of wall build up and heat generation. In general, more energy input into the granulation resulted in a greater amount of product wastage through sticking of the powder to the walls and a greater final temperature of the granules after granulation. Shorter granulation times are investigated in subsequent sections of this thesis.

5.4 Effect of changing the solids concentration of the binder

The previous experiments highlighted the importance of moisture content on granulation. In order to optimise the granulation it is important to deliver the moisture in as high a solids content milk concentrate. This necessitates a study into the effects of changing the binder solids concentration. The experiments discussed above used reconstituted milk as a binder because of the ease of preparation. It was necessary, however, to use evaporated milk concentrate fresh from the plant, to simulate real process conditions and to test the validity of using reconstituted milk. Evaporated milk concentrate was available from the NZFRC but at a solids concentration of 50 % solids rather than the nominal 30 % solids used in the above reconstituted binder trials. This provided a means to test both the use of a higher solids binder and the effect of using an evaporated milk concentrate binder. An industrial process will use evaporated milk concentrate rather than reconstituted milk as the milk from evaporators is made continuously and has undergone much less processing.

The purpose of this section is not to optimise the solids concentration of the evaporated binder as there was not enough evaporated milk concentrate to do a thorough study. Instead, this section aims:

- to investigate whether successful granulation could occur using higher solids concentrate binder
- to further investigate the affect of varying the moisture content of the granules
- to compare evaporated milk concentrate to reconstituted milk concentrate for use as a binder.

5.4.1 Experimental set-up

Four experimental runs were carried out using evaporated milk concentrate at different solids concentrations. The concentrate was only available from NZFRC on

two occasions so a limited number of experimental runs could be completed. Table 5.7 shows the set-up of the experimental runs. Runs 13 to 15 used concentrate provided from the same evaporation run. Run 13 used fresh concentrate approximately 1 hour after being evaporated while run 14 used concentrate that was cooled to 5 °C then heated approximately 7 hours after evaporation. Run 15 used concentrate that was cooled to 5 °C then stored overnight and reheated the next morning after approximately 12 hours. Run 16 used freshly made evaporated concentrate produced on a different day to the other runs that was used approximately one hour after production.

Relatively high solids concentrations are used compared to previous experimental work, as these concentrations were available from the NZFRC and will allow investigation into the effect of using concentrates that are similar to those presently produced in industrial milk evaporators (Caric, 1994, Pearce, 2001). A new bag of milk powder was used for this experiment to ensure the freshness of the powder. The moisture content of the powder was found to be 4% total solids and this value was used to estimate the desired powder wet mass moisture.

Some solids separation was observed during the storage of the concentrate, which introduced error into the experiment through inaccurate estimation of the solids content. The solids content for run 15 was estimated to be 36% as some separation was observed during storage. The measurement of the solids prior to the separation was 38% solids so a reduction of 2% was added as a qualitative estimate to account for the removal of a solid skin layer after storage.

Tables 5.7 *Experimental set-up to investigate the effect of using evaporated milk concentrate binder at different solids concentrations.*

Run	Estimated binder total solids	Desired granule total moisture	Amount of binder added
13	50 %	11 %	270 g
14	50 %	12 %	292 g
15	36 %	12 %	234 g
16	40 %	11 %	215 g

Moisture contents of 11% and 12% of the wetted powder mass were tested to investigate whether the higher solids concentration binder, with a subsequent higher viscosity, could be granulated at higher moisture contents. This may have an advantage in an industrial process, as a higher moisture content will result in faster granule growth.

The granulation conditions set for the experimental runs are shown in table 5.8. Each experiment has a dry mixing period of 2 minutes, a binder addition period of 8 minutes and a variable granulation period. The shorter granulation time for run 16 is because this run was undertaken after the time of granulation experiment (section 5.5) where it was found that extended granulation times were detrimental to granule quality.

Table 5.8 Experimental conditions for the investigation into the effect of evaporated binder concentration on granulation.

Mixer speed:	290 rpm for mixing and binder addition 700 rpm for granulation
Chopper speed:	800 rpm for mixing and binder addition 1900 rpm for granulation
Amount of milk powder added:	1.5 kg
Binder added after:	two minutes of dry mixing
Binder addition added over:	8 minutes
Length of granulation:	Runs 13, 14 & 15, 10 minutes; Run 16, 2 minutes

5.4.2 Results and discussion

Table 5.9 shows the granule moisture and yield results for this experiment. A discrepancy exists between the desired granules total moisture and the measured granule total moisture, which is partially due to the discrepancy between the nominal binder solids, measured when supplied from the NZFRC, and the actual binder used during the experiment. Because granulation is highly dependant on moisture content, it is necessary in an industrial process to tightly control moisture content so that variations are no worse than the variations shown in this experiment.

Table 5.9 Results of using 40 % and 50 % solids evaporated milk concentrate binder at total granule moisture contents of 11 % and 12 %.

Run	Nominal binder total solids	Measured binder total solids	Desired moisture content	Measured final moisture content	Granule yield
13	50 %	48.2 %	11 %	10.5 %	1544 g
14	50 %	48.2 %	12 %	11.2 %	1514 g
15	40 %	32.2 %	12 %	11.6 %	1366 g
16	40 %	43.3 %	11 %	11.2 %	1664 g

Granule yield is related to moisture content. The lower granule yield for run 15 can be attributed to the higher moisture content of the granulation resulting in more sticking of the wetted powder to the granulator walls. The relatively high yield for run 16 can be attributed to the shorter granulation time of 2 minutes compared with 10 minutes for the other runs. A shorter granulation time results in less product being lost due to sticking. Granulation time is further discussed in section 5.5.

Figure 5D shows that the granule size distributions are very similar for runs 13 to 15. The granules produced during these runs were fluidised-bed dried for 2 hours at 30 °C prior to sieving (see chapter 3). They appeared to be less sticky than those produced in previous runs using reconcentrated binder at lower solids concentration. Therefore, sieve blinding was not a significant problem for these granules.

The data from run 16 is not included in figure 5D as this run used a different granulation time and was not sieved to the same size fractions. The granule size distribution of run 16 is shown in table 5.10. The distribution is considerably coarser which suggests that the time of the granulation is an important factor in the size distribution of milk granules. This initial broad size distribution is common in granulating systems (Knight et al, 1998), which narrows with time as loose, large agglomerates are broken down and fines are absorbed.

Table 5.10 Granule size distribution for run 16. Evaporated concentrate binder at 40 % solids to achieve an 11 % granule moisture content.

Granule size fraction	Fraction of total
0 to 250 microns	0.131
250 microns to 1 mm	0.224
1 mm to 2 mm	0.529
2mm +	0.116

The size fractions of granules that are likely to be useful for reconstitution purposes are between 250 μm and 1 mm (Pearce, 2001). As can be seen from table 5.10, 35 % of the granules from run 16 are above 1 mm in size. This is undesirable, therefore, longer granulation times are needed to narrow and reduce the size distributions.

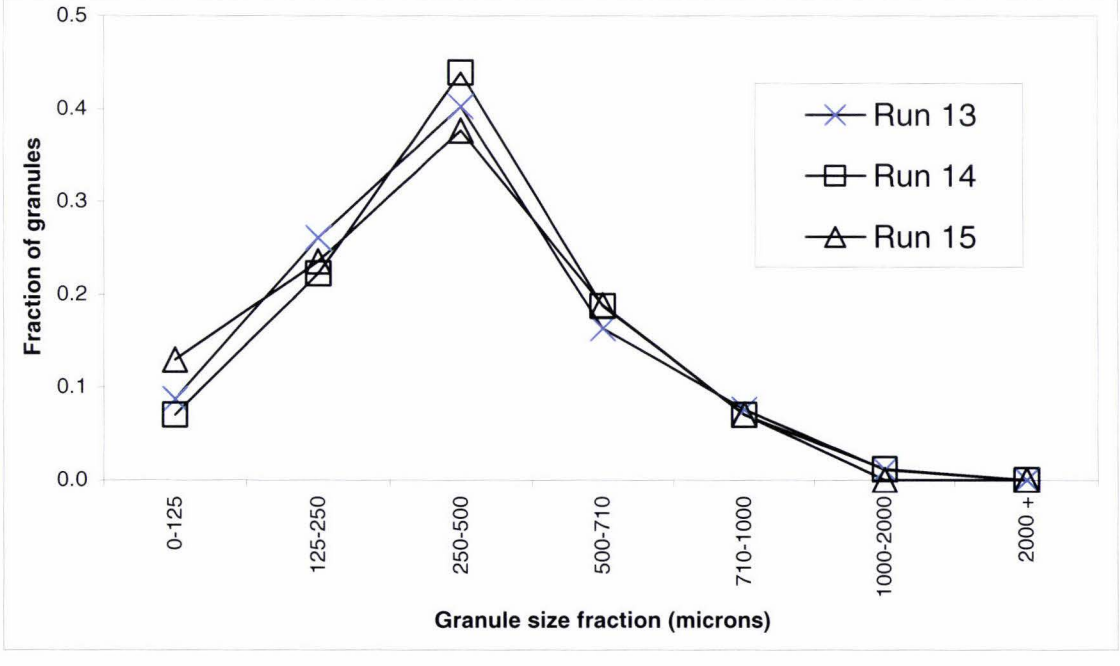


Figure 5D Granule size distribution for runs using evaporated concentrate as binder at 40 % and 50 % solids to produce granules of 11 % and 12 % total moisture. ***Run 13 = 50 % solids binder and 11 % moisture granules; Run 14 = 50 % solids binder and 12 % moisture granules; Run 15 = 40 % solids binder and 12 % moisture granules.***

5.4.3 Conclusions

Milk concentrate was found to granulate easily even though the solid content of the binder was higher than previous experiments using reconstituted milk powder. Binder solids concentrations were lower than expected which impacted on the final granule

moisture contents. The resultant granule size distributions were similar for the runs 13-15 that used 10 minutes granulation time.

The granule yield decreased with an increase in granule moisture content for the range tested, with a significant drop in granule yield at a granule moisture content of 11.6 % moisture. A granulation moisture content of 11 % total moisture is therefore suitable for granulating milk with an acceptable range of ± 0.5 % total moisture. These conditions appear to be suitable using evaporated or reconstituted binder between 20 % and 50 % total solids.

The shortened 2 minutes granulation of run 16 gave a higher granule yield but also gave a broad granule size distribution with too many over-sized granules.

5.5 Effects of changing granulation time

In many granulation processes a granulation end point is defined where the optimal size distribution of the granules is obtained. Sometimes this is measured by a rise in torque (Hamilton *et al*, 2000). However, from a processing perspective, especially for large capacity granulators, the granulation time should be as short as possible to avoid heat generation. The main objective of this experiment was to investigate the effect that granulation time has on the granule size distribution. The effect on the granule yield and the final granule temperature was also investigated as well as a brief look into repeatability of the granulation experiments.

5.5.1 Experimental set –up

Four experimental runs were conducted at 2, 5, 5, and 10 minutes granulation time. The granulator was run for two minutes of dry mixing prior to binder addition. Binder was added over an eight minute period that consisted of about three minutes of binder pulse spraying and five minutes of further mixing to allow the binder to wet and mix into the powder before the granulator blade speeds were increased. The experimental conditions are shown in table 5.11.

Table 5.11 Experimental conditions for the investigation into the effect of granulation time on milk granules.

Mixer speed:	290 rpm for mixing and binder addition 700 rpm for granulation
Chopper speed:	800 rpm for mixing and binder addition 1900 rpm for granulation
Amount of milk powder added:	1.5 kg
Binder solids concentration:	30 % (total solids content)
Amount of binder added:	204 g (11 % granule moisture content)
Dry Mixing:	two minutes
Binder added over:	five minutes
Length of granulation:	2, 5 and 10 minutes

Reconstituted milk concentrate at approximately 30 % total solids was used as the binder. A freshly opened bag of spray dried milk powder was used for granulation and reconstitution of the binder. The moisture content of the milk powder used was measured at 3 % total moisture using the 105 °C oven method described in chapter 3. The granulation moisture content was set to be 11 % total moisture assuming the milk powder used had a moisture content of 3 %.

A replicate of the five-minute granulation run was also included in the experiment to investigate the reproducibility of the granulation. The granules were fluidised-bed dried at 30 °C for two hours before sieving.

5.5.2 Results and discussion

Table 5.12 shows that the yield decreases with increasing granulation time, granule temperature increases with increasing granulation time and that granule moisture content varied between runs although the same intended amount of moisture was used. Some difference in evaporative losses is expected with differing granulation times. Also, this was partially due to an underestimate of the moisture content of the milk powder used. Post-experimental measurement of the powder found a moisture content of 4.2 %. It is likely that the powder absorbs moisture when exposed to the open air due to the presence of amorphous lactose. The water activity of the bag of powder used was found to be 0.25 when measured after the experiment, with an ambient air relative humidity of approximately 0.5. This suggests that the powder will absorb moisture readily, as spray dried milk powder is a hygroscopic material (Kessler, 1981).

Table 5.12 Results of varying granulation time runs.

Run	Time of granulation (minutes)	Granule moisture content (%)	Granule temp (°C)	Granule Yield (g)
17	2	12.3	28.5	1434
18	5	11.5	33	1202
19	5	12.9	30	1278
20	10	10.1	41	1012

The granule yield decreases with increasing granulation time. This was also observed in the previous experiment described in section 5.4 and is due to more powder being stuck to the walls of the granulator as time progresses. It is therefore desirable to minimise the granulation time to reduce product wastage. The temperature of the granules also increases with increasing granulation time. This is likely due to the build-up of powder on the walls of the granulator, preventing heat loss to the water jacket.

The torque profiles are shown in figure 5E and 5F for the motor shaft and chopper respectively (for measurement methods see chapter 3). Both figures show that all experiments follow the same profile suggesting that there is little variation of the physical conditions inside the granulator between each of these runs. The first two minutes of the torque profiles shows the dry mixing stage before binder is added. Following this is a period of increasing torque as the binder is added in pulses. The binder addition finishes at around five minutes and here the torque drops to a

relatively steady rate. Torque increases at ten minutes when the blade speeds are increased for granulation. The granulation is ended when the motors are switched off and the torque drops to zero.

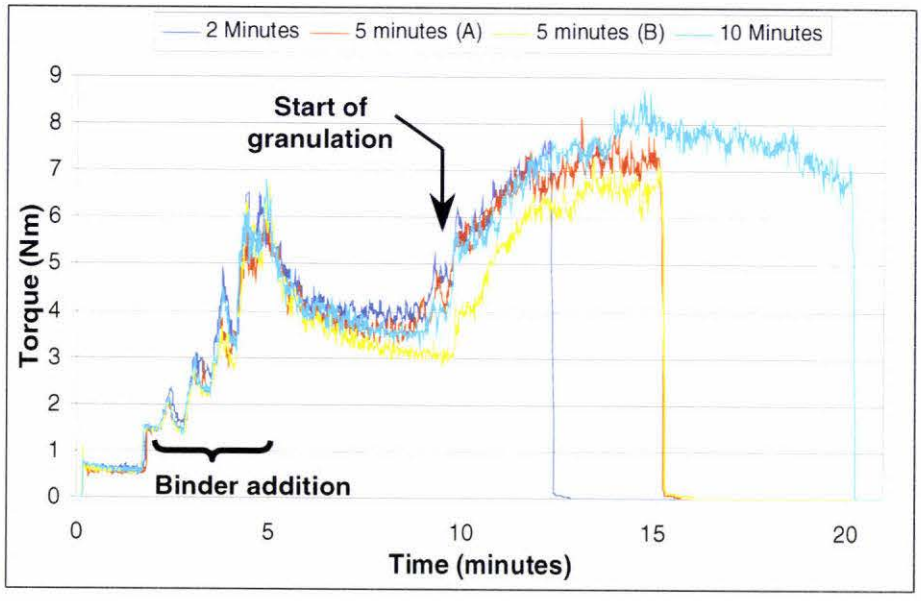


Figure 5E **Mixer torque profiles for granulation runs with varied granulation time.**

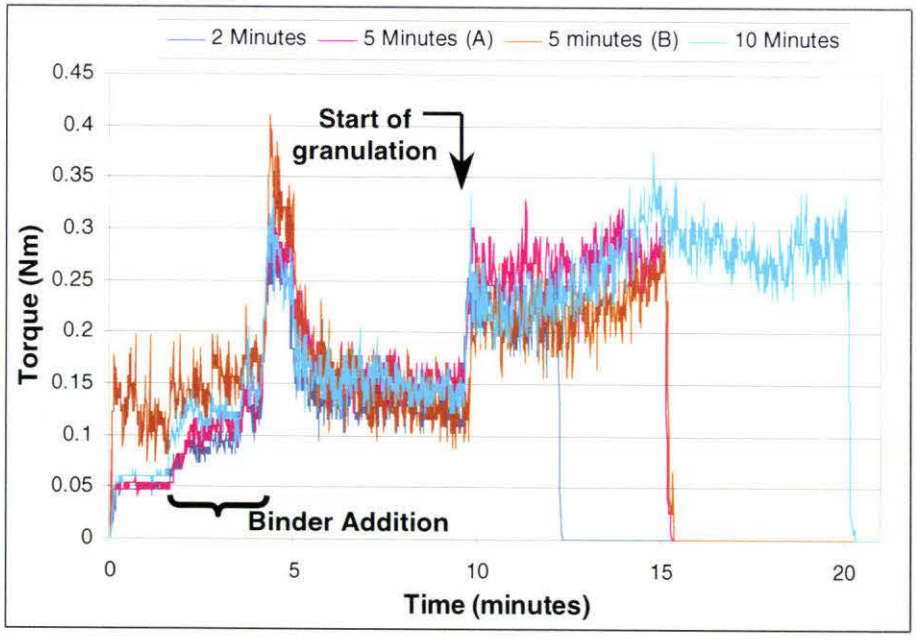


Figure 5F **Chopper torque profiles for granulation runs with varied granulation time.**

At around 15 minutes, or after five minutes of granulating, the torque profile of the mixer blade appears level off and begins to drop. This is likely to be because of the removal of powder from the granulation as wall build-up occurs resulting in less energy being required to move the remaining granules around the granulator.

Figure 5G shows that increasing the granulation time increases the average granule size with a narrowing of the granule size distribution. The 2 minute granulation had

the broadest size distribution. This broad and somewhat bimodal behaviour is expected in the early stages of granulating systems (Knight et al, 1998). Granule growth can be seen after 5 minutes and by ten minutes all fines have disappeared and the distribution is both narrow and consists mostly of large particles. This indicates that the granulation mechanism in later stages is one of layering of the remaining fines and small granules onto the larger granules.

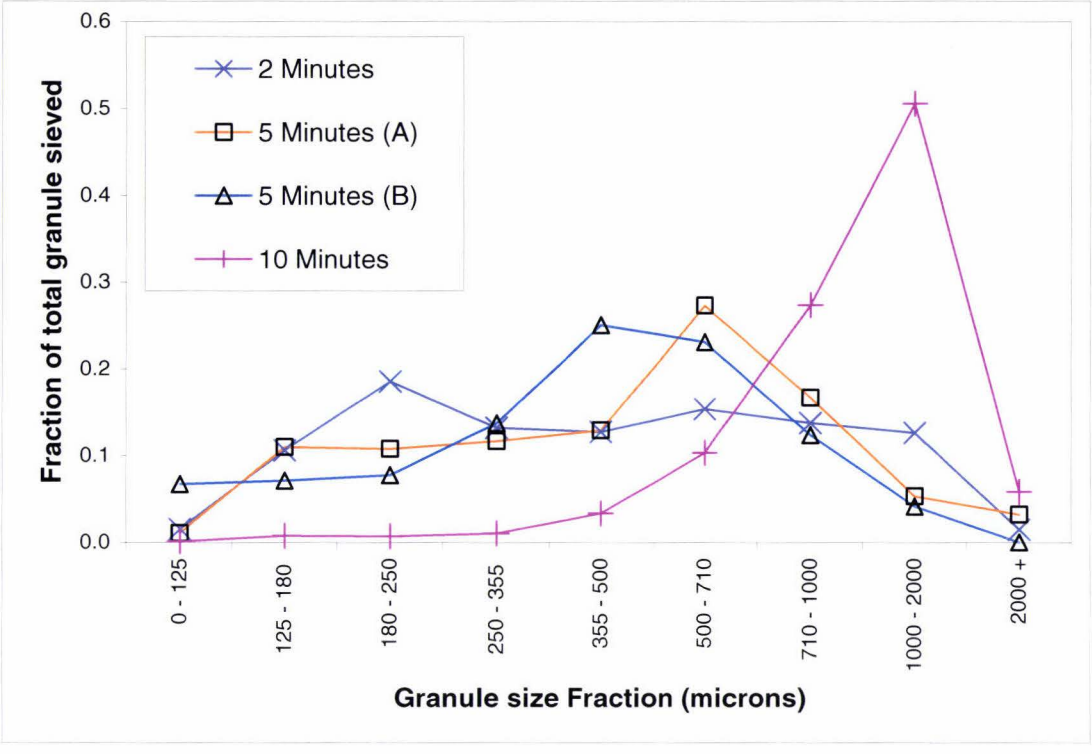


Figure 5G Granule size distributions for granulation runs with varied granulation time.

Repeatability was tested by the two runs using a five-minute granulation time. They produced slightly different granule size distributions even though the granulating conditions were identical which cannot be explained. This is a surprising result considering very good repeatability of earlier reported runs 13 and 14 using evaporated milk concentrate. The most noticeable difference between runs 18 and 19 and the earlier runs 13 and 14 is the solids concentration of the binder which was 30 % here, but 50 % previously. This will affect the binder viscosity, which has been shown to be an important parameter in granulating systems (Keningley et al, 1997); in some systems a critical binder viscosity exists above which granule formation is highly dependant on viscosity. This will affect the way moisture distributes within the powder. Exactly how viscosity may affect repeatability is not known and is an area requiring further work.

5.5.3 Conclusions

Increasing granulation time has the effect of reducing the granule yield, increasing the average granule size and narrowing the granule size distribution.

After two minutes, the mechanism of granulation appears to occur through layering of fines onto larger granules resulting in a growth in the average granule size and a narrowing of the granule size distribution.

Some variability was found between the granule size distribution of runs with similar granulation conditions. This cannot be explained, except to note that a discrepancy in the moisture content of the granules occurred which can only be linked to a variation in moisture content of the initial powder.

5.6 Chapter closure

This chapter investigated many of the variables of granulation in an attempt to find suitable conditions for producing granules. Mixer and chopper blade speeds were found to have an effect on the temperature of the granules and the amount of product lost through sticking to the walls of the granulator. A mixer speed of 700 rpm and a chopper speed of 1900 rpm were chosen for further granulation work as the flow of powder within the granulator was vigorous but did not throw powder against the non-mixed areas of the granulator walls, when wall build up was shown to be a problem.

An 11 % total moisture content is optimal for granulation of whole milk powder; something important to monitor in an industrial process. The optimal amount of binder required to achieve successful granulation was found to be related to the amount of total moisture in the wetted powder mass.

Granulation time was found to affect the average granule size and the granule size distribution. The average granule size increased with an increasing granulation time, which is attributed to the layering of milk powder fines onto larger granules that had been formed during the binder addition phase. This resulted in the granule size distribution becoming narrower with time, which will lead to more favourable handling properties.

Chapter 6 – Functional testing of granules

6.1 Introduction

This chapter investigates the functional and physical properties of milk granules. The work described here was undertaken to investigate the feasibility of producing milk granules commercially. The scope of the testing was limited to NZFRC functional tests and tests to determine the physical characteristics of the granules, such as porosity and the drying rate of granules. The methods used for the testing are described in detail in chapter 3.

The granules used in testing were produced under identical conditions as described in table 6.1 except where noted.

Table 6.1 Granulation conditions of granules produced for functional tested

Mixer speed:	290 rpm for mixing and binder addition 700 rpm for granulation
Chopper speed:	1324 rpm
Amount of milk powder added:	1.5 kg
Binder added:	204 g of reconstituted 30 % total solids milk concentrate
Binder added after:	30 seconds
Length of granulation:	2, 5 or 10 minutes granulation after binder addition
Drying conditions:	Fluidised bed for 2 hours at 30 °C (see chapter 3) or open air dried

6.2 Reconstitution functional testing

The most important functional test for a dried milk product is the solubility index test, in which the amount of insoluble material present in the sample is measured using the standard NZFRC solubility index (SI) testing procedure described in chapter 3. A change in solubility of the powder is an indication that chemical changes have happened to the components of the milk resulting in an undesirable reduction in quality. Therefore, the solubility of a granulated product should not be affected during granulation or subsequent drying. The maximum acceptable level of insoluble material in reconstituted milk is 0.1 ml of insoluble material per 50ml of sample (NZFRC Milk Powder Methods I).

6.2.1 Isolating the storage problem

Granules dried in ambient open-air conditions were found to give unacceptable levels of insoluble material when reconstituted. The results of the solubility index (SI) tests are shown in table 6.2. The granules that were tested were taken from the four granulation runs described in section 5.5.

Table 6.2 Solubility Index (SI) results for ambient open-air dried granules produced with various granulation times.

Time of granulation (minutes)	Time between manufacture and SI testing (days)	SI at 24 °C (ml insoluble material/ 50 ml sample)
2	4	1.6
5 (A)	6	2.4
5 (B)	10	3.8
10	5	2.4

The granules were stored under ambient open-air conditions in an attempt to dry them without altering the granule size distribution, as described in chapter 3. The difference in the time between manufacture and testing between runs is due to the testing for all samples occurring on the same day while the granulation runs occurred on different days. As can be seen from table 6.2, the solubility index is significantly higher than the acceptable level of 0.1 for all of the runs. Baldwin (2001) found that the generation of insoluble material during storage of skim milk powders was found to be dependant on the temperature and the moisture content of the powders with a significantly higher rate of insoluble material generation for a moisture content of around 10 % compared with around 3.5 %. Moisture content measurements were not made, but it is likely that the presence of insoluble material in the granules is due to the storage at higher moisture contents of the ambient open-air dried granules.

The effect of storage time was eliminated by repeating the experiment and testing for the solubility index on the same day. The solubility index was measured for wet granules and for granules dried in the fluidised bed to isolate the effects of granulation and drying on the SI. Furthermore, the SI was performed at 24 °C and 50 °C; the latter temperature being typical of reconstitution conditions in industry. The results are shown in table 6.3.

Table 6.3 Solubility Index (SI) for milk granules produced under favourable conditions.

Sample	SI at 24 °C (ml/50ml sample)	SI at 50 °C (ml/50ml sample)
Powder	0.10 ± 0.02	0.10 ± 0.02
Unsieved Wet granules	0.10 ± 0.02	0.10 ± 0.02
Wet granules 250 μm to 1mm diameter	0.10 ± 0.02	0.12 ± 0.02
Dried granules	0.08 ± 0.02	0.10 ± 0.02

These results show acceptable solubility indices. The range of readings from the same samples was found to be ± 0.02 ml per 50 ml sample so there was no significant difference in solubility between any of the samples tested. The results show that neither granulation nor the test temperature has a deleterious effect on the SI. The problem appears to be a combination of storage time and storage conditions.

6.2.2 The effect of time and moisture on the solubility index

The effect of storage time and storage conditions were examined in detail by storing milk powder (control), granulated wet granules and granulated dried granules for up to ten days. Solubility index tests were performed on days 1, 3, 6 and 10. Samples were stored in 25 ml screw top plastic containers filled up to the top with the sample and sealed with Sellotape. The samples were stored in a dark incubator at 20 °C. The water activity and moisture content were also recorded to compare any changes between the samples.

Table 6.4 Granule storage conditions for functional property changes over time experiment

Sample	Water activity	Moisture content
Powder	0.21 ± 0.02	4.2 ± 0.1 %
Wet granules	0.71 ± 0.02	10.5 ± 0.4 %
Dried granules	0.19 ± 0.02	5.5 ± 0.4 %

Table 6.4 shows that the water activity and moisture content of the samples did not vary greatly over the ten day storage time. There were no noticeable trends in the small variations that were observed. The water activity of the dried granules was lower than the powder even though the moisture content was higher. This result is unexpected and suggests that the mobility of the water in the dried granules is lower. This could be due to water becoming chemically bound as water of crystallisation. Crystallisation of lactose may be possible during granulation because of the longer time frame over which drying occurs compared to spray drying.

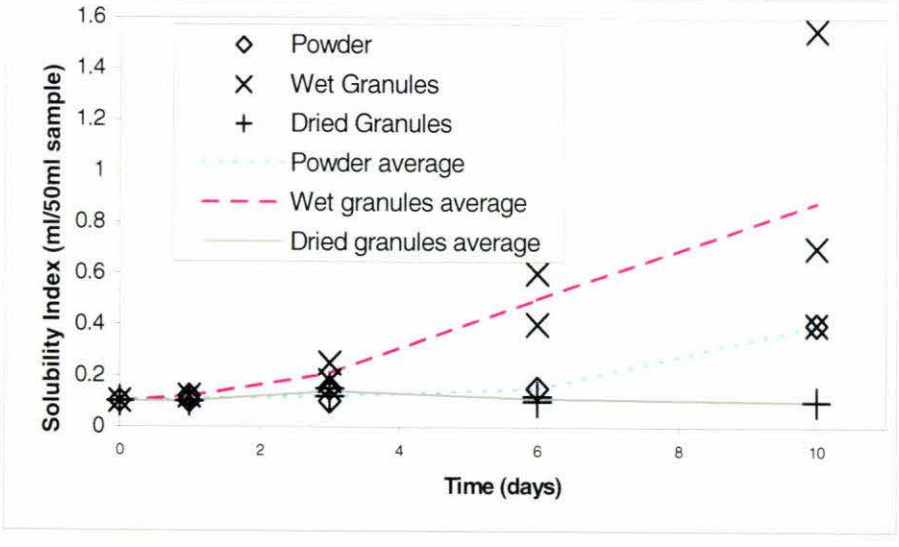


Figure 6A Insoluble material generation over time as a function of granule storage

The dried granules and the powder have no significant difference between each other until the tenth day, where the powder has an increase in insoluble material while the dried granules do not. The increase in insoluble material for powder at day ten is unexpected and can not be easily explained, as powder taken from the bag did not have a significant increase in insoluble material even after a month of the original opening of the bag.

The wet granules had an increase in the variability between samples and an increase in the average amount of insoluble material over time. This result agrees with the findings of Baldwin (2001) for skim milk powder that insoluble material generation is due to storage at elevated moisture contents. These results also give further evidence that the granulation process, and subsequent drying does not affect the solubility of whole milk powder.

6.2.3 Wettability of milk granules

The wettability of dried granules was tested using a standard wettability test described in NZFRC Milk Powder Methods II. The granules were dropped into a water bath at 24°C and the time taken for the granules to sink to the bottom of the container was measured. The granules tested did not have a surfactant such as lecithin added to improve the wettability. Granules were produced under the conditions described in table 6.1 with granulation times of 2, 5 and 10 minutes.

None of the granules tested were found to have a suitable wettability as they did not break the surface of the water for at least six minutes. Because granules are consolidated it is likely that the pores of the granules were too small to allow rapid moisture infiltration. In addition, although the surface properties of the granules were not measured, it is known that the surface of whole milk powder has a high fat content (Kim et al, 2002). If granules have a similar surface fat content, combined with significantly smaller pores, then moisture infiltration may be obstructed.

The granules sank when mixed gently or if the container was tapped suggesting that the surface tension of the water contributed significantly to the failure of the granules to become wet. Therefore, granulation may not 'instantise' the milk as well as the currently used method of agglomeration of recycling powder fines to the top of a spray drier, where all of a powder sample is expected to sink within 6 minutes of the wettability test. Further work may be required to investigate the reasons why granules are not easily wettable and to find ways to improve their wettability. The addition of lecithin to the granules will improve this but was not tested in this work.

6.3 Bulk density, particle density and porosity of granules

A key industry concern in transporting powders is consolidation of the powder over time which results in unutilised head space and an appearance of partially filled containers. The amount of variation in bulk density that occurs over time is found by the difference between the loose or 'poured' bulk density and the 'tapped' bulk density. This gives an indication of the amount of packing of the powder that will occur when transported. The particle density is a measure of the true density of the solid without the air spacings between particles.

6.3.1 Bulk Density and Particle Density Changes

The bulk density and particle density of granules and powder were measured according to NZFRC standard tests (Milk Powder Methods II). The granules were produced using the conditions described in table 6.1 with a granulation time of 5 minutes. Figure 6.5 compares the results of the bulk density and particle density measurements made for granules and powder.

Table 6.5 Bulk density and particle density changes due to granulation

Sample	Loose bulk density (g/cm^3)	Tapped bulk density (g/cm^3)	Change in bulk density	Particle density (g/cm^3)
Spray dried whole milk powder	0.44 ± 0.02	0.58 ± 0.02	32 %	0.85 ± 0.02
Granulated whole milk powder	0.67 ± 0.02	0.70 ± 0.03	5 %	0.92 ± 0.02

The granules have a significantly higher bulk density and a slightly higher particle density. The spray dried milk powder has a porous structure (Masters, 1991) while granules produced using high-shear mixer granulation are generally dense (Ennis and Litster, 1997). In addition, the granules are spherical whereas the spray dried milk powder particles appear more like a bunch of grapes (Caric, 1994). Spherical particles pack better and result in a higher bulk density. The comparison between the granule size distribution and powder size distribution is shown in figure 6B.

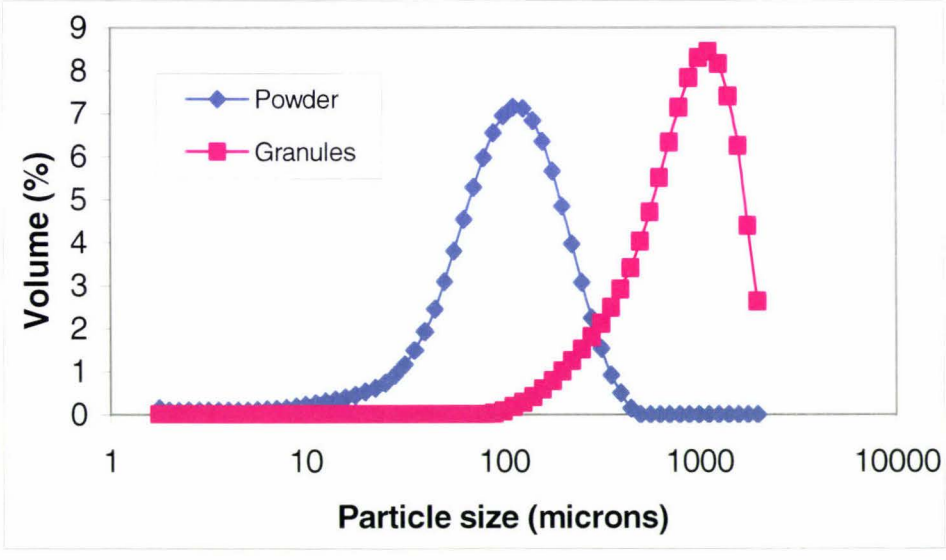


Figure 6B Comparison of particle size distributions for powder and granules using a Malvern Mastersizer 2000.

6.3.2 Porosity of granules

A mercury porosimeter was used to estimate the pore size distribution of the granules as described in chapter 3. Figure 6C shows the results of the pore size distribution for granules made under the conditions described in table 6.1 with a granulation time of five minutes. The apparent step in the trend is due to the boundary between the intergranular pore spaces and the actual granule pores (Marshall, 2001) although it is possible that some pores are larger than this boundary.

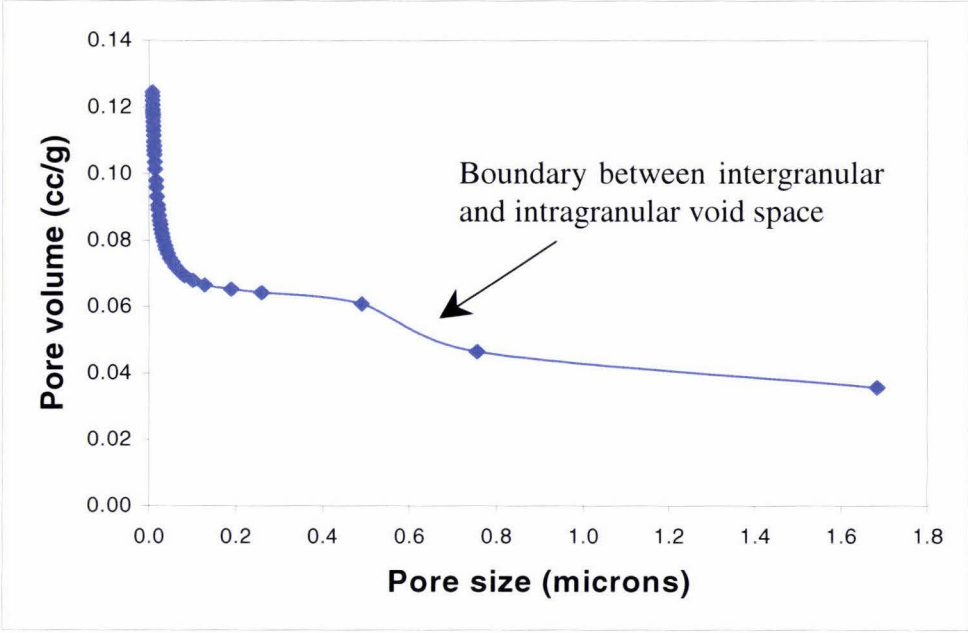


Figure 6C Pore size distribution for granules

As can be seen from figure 6C the pore sizes observed are very small with the largest pore volume occurring below 0.1 microns. This provides evidence as to why the granules were not acceptably wettable as evidenced in section 6.2.3. Small pores reduce the infiltration of water into the granules, reducing the instantising ability of the granules. This suggests that milk granules may not be suitable as an instantised product and may be limited to a role as an improved product for reconstitution where the wettability is not vitally important due to the high shear of the reconstitution process.

6.4 UHT treatment

Ultra high temperature (UHT) treatment involves sterilising the milk by heating to a high temperature for a short time. The changes in the milk that may occur during UHT treatment are reflected in colour changes, gelation and coagulation, the separation of fat during storage, sedimentation during storage and a change in viscosity due to protein agglomeration (Newstead and Mason, 1997). The extent of change depends on the heat stability of the milk (McCrae and Muir, 1995). This will be dependent on the state of the protein in the milk (Singh, 1995) and the composition and pH of the milk (Singh et al, 1995).

UHT treatment trials were performed with reconstituted milk made both from milk powder and milk granules. The purpose was to investigate differences in fouling rate of the UHT heat exchanger. A high fouling rate indicates that granulation affects the heat stability of milk. Also investigated was the fat rise and sedimentation of the UHT treated reconstituted milk during storage. The amount of fat rise gives an indication of the chemical changes that occur during granulation and reconstitution in the fat globules. Sedimentation indicates an irreversible reaction in the components of the milk resulting in insoluble material forming and settling out. It is preferable that both fat rise and sedimentation are kept to a minimum.

6.4.1 Fouling during UHT treatment

Granules were made under the conditions described in table 6.1 with a granulation time of 5 minutes. They were made over a ten-day period, as 10 kg of granules were required for the testing. The granules were flushed with nitrogen after each granulation run prior to storage and were stored in an airtight bag sealed by goosenecking with two rubber bands. Storage was in a dark incubator at 10 °C.

The granules were reconstituted up to 12.5 % total solids prior to the UHT treatment and screened to remove any visible non dissolved solids. A control batch of spray dried milk powder was also reconstituted under the same conditions. The control milk powder was taken from the same bag of powder used to make the granules. The reconstituted milk was passed through the NZFRC small scale UHT plant where it was heated to 140 °C and held for 4 seconds.

Across the final heat exchanger used to heat the milk, the inlet and outlet temperatures of the reconstituted milk and the heating fluid were measured, allowing a log mean temperature difference (θ_{lm}) to be calculated. The θ_{lm} is a measure of the temperature difference between the heating medium and the milk being heated. An increase in this value indicates fouling because the heat transfer across the heat exchanger is reduced. The fouling rate was determined by plotting the θ_{lm} across the final heating heat exchanger. Figure 6D shows a comparison between the fouling rates of the control powder and the granules.

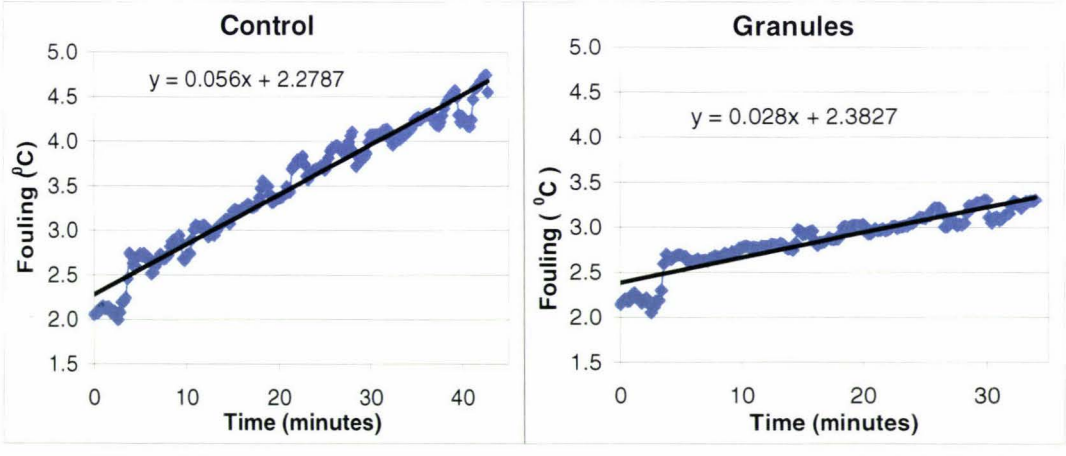


Figure 6D **Comparison between fouling rate of reconstituted milk during UHT treatment for spray dried milk powder (control) and granulated spray dried milk powder**

The trend in figure 6D shows that the increase in fouling (θ_{lm}) over time for the control run is faster than for the run using granules. This is further illustrated by plotting a comparison of the change in heat transfer coefficients for the final heat exchanger as shown in figure 6E, in which the heat transfer coefficient at any time (U) is divided by the initial heat transfer coefficient (U_o) and plotted over time. U/U_o was used instead of U as other factors in the equation for determining U were not accurately known but can be assumed constant and can be cancelled from the calculation by dividing U by U_o.

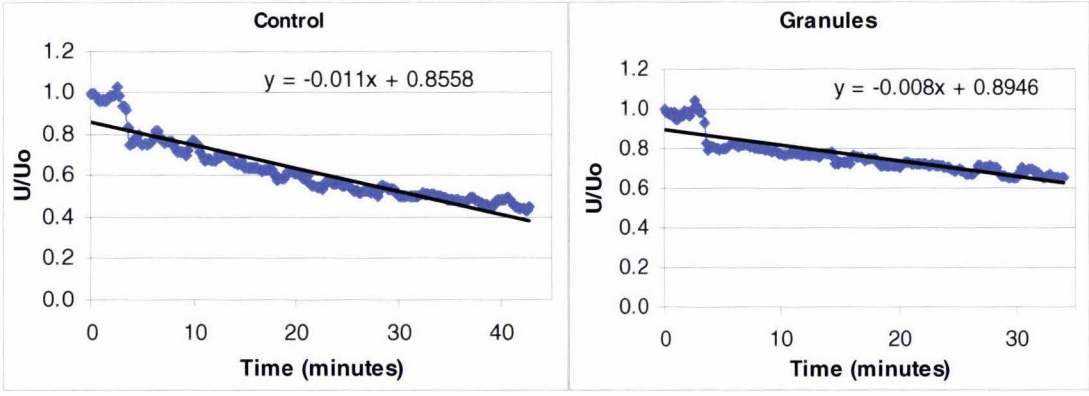


Figure 6E Comparison of change in heat transfer coefficient over time during UHT treatment.

The sudden drop in U/U_0 at around 3 minutes is the start of the milk flow through the heat exchanger. The rate at which U/U_0 drops is more rapid for the control run. This shows that fouling occurred at a slower rate in the granule run suggesting that the reconstituted milk made using granulated milk powder was more heat stable. The reason why this occurs is not clear and is an area of further work.

6.4.2 Fat rise and sedimentation during storage

The UHT treated milk from both trials above were stored to test for fat rise and sedimentation. The fat rise test was taken from the visual, subjective method described in Newstead and Mason (1997) with the samples stored in 50 ml cylindrical plastic screw top containers for two months at 30 °C and left for two hours at ambient conditions before measurement were taken. The milk used was from the UHT run described in section 6.4.1 and was not homogenised prior to storage. Reconstituted milk made for UHT treatment is usually homogenised before packaging to reduce the fat separation but during this experiment homogenisation was not done to help clarify any differences in fat separation between granulated and non-granulated reconstituted milk.

The thickness of any fat layer or sedimentation in the milk was recorded by measurement with a ruler. The samples were separated into three batches depending on the stage of the UHT run. The results of the measurements are shown in table 6.6.

Table 6.6 Fat rise and sedimentation during storage of UHT treated reconstituted milk made from granules

		Control			Granules		
		Start	**Middle**	**End**	**Start**	**Middle**	**End**
Fat rise (mm)	mean	10.7	10.7	10.5	8.5	9.4	8.4
	s.d.	0.68	0.89	1.41	2.1	0.90	0.98
Sedimentation (mm)	mean	0	0	0	0	0	0

As can be seen from table 6.6, no sedimentation was observed for any of the runs. The average fat rise for the run using reconstituted granulated milk powder is slightly

lower than the control run although this lies well within two standard deviations suggesting that there is no statistical difference between the two runs. The amount of fat rise was relatively high for both runs, as no noticeable fat layer should form before two months for good quality milk (Newstead and Mason, 1997) although leaving out the homogenisation step prior to storage should have greatly enhanced the fat rise. This experiment has provided further evidence that granulation of milk powder has no noticeable adverse effect on the chemistry of milk.

6.5 Batch drying curve for granules

The drying characteristics of granules will be important in determining the feasibility of milk granulation. The physical properties of granules that differentiate them from spray dried powders such as a higher density, larger size and lower porosity may negatively affect the rate of drying. In order to determine this an experiment was undertaken to measure the rate at which granules dried. This could then be compared with previous work done using spray dried and instantised spray dried milk powders. This work was limited in scope due to time constraints. Further study of this area is therefore suggested to determine better understanding of granule drying behaviour.

Granules were made under the conditions described in table 6.1 with a five-minute granulation time and no drying. The wet granules were then transferred to the Glatt fluidised bed drier described in chapter 3. The time between the end of granulation and the start of drying was approximately ninety minutes due to transport and other logistical barriers. The granules were then dried using 95°C air over a two-hour period in which samples were taken for moisture analysis. Figure 6E shows the results of the run and includes for comparison instant and non-instant spray dried milk powder drying curves adapted from Pearce and Buchanan (1996). The moisture content of the granules was measured using the 105°C oven method described in chapter 3 while the work from Pearce and Buchanan (1996) used the Karl Fischer method of moisture analysis. The two methods of moisture analysis were compared using granules and were found to vary by 0.02 %, which was deemed acceptable to be within experimental error.

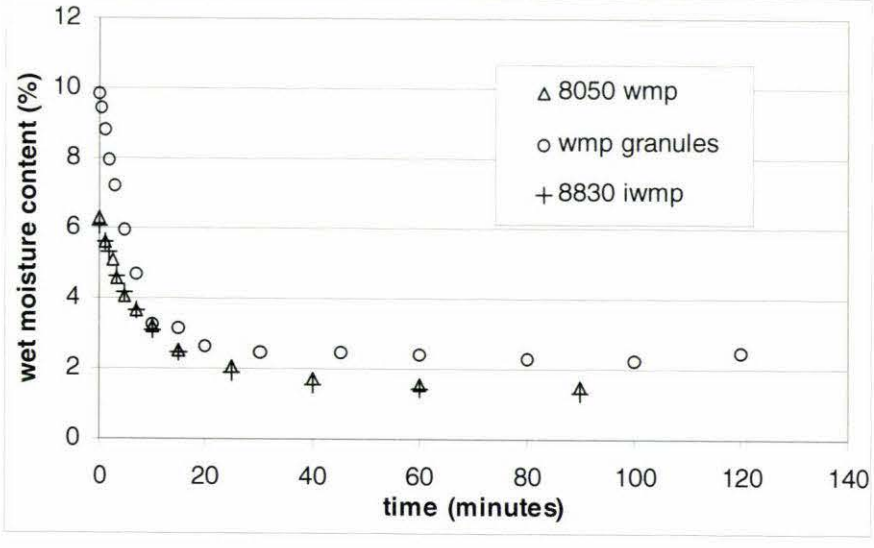


Figure 6F Batch drying curves for granules compared with instant (8830) and non-instant (8050) spray dried milk powders.

Figure 6E shows that the granules have a higher initial and final moisture content. The difference in initial moisture contents is because the powders of Pearce and Buchanan were removed from the start of a continuous industrial sized fluidised bed drier while the granules were taken straight out of the granulator. The final moisture contents of the powders are lower than the final moisture content of the granules. The reason why is not immediately clear but may be due to water becoming chemically bound as water of crystallisation in the lactose during granulation. When dried at 105°C during the moisture measurement procedure, some of the water of crystallisation will be removed (Bronlund 1997) resulting in a higher moisture content reading for the granules. Different operators and equipment between the drying curves for the powders and the granules drying curve may also explain some of the differences.

The final moisture content of the granules, around 2.2 %, is an acceptable level for dried milk powders as milk powders are normally dried to around 2.4 % to 3 % total moisture for optimal storage conditions (van Mil and Jans, 1991). It is also noticeable that below 3 % total moisture the drying rate is significantly slower. This is most likely due to an increase in resistance to moisture transfer due to smaller pore sizes of the granules compounded with a larger average particle size compared to spray dried powder. As has been shown in section 6.3.2, the pore sizes in the granules are very small so moisture transfer from the smallest of these pores will be slow and drying will consequently take longer. However, pore impeded drying only occurs below 3 % moisture, which is the target moisture content for optimal storage of dried powder. Therefore, in the useful range of drying both granules and whole milk powder have similar drying characteristics.

This result is limited to only one drying run due to time limitations. Pearce and Buchanan (1996) used four drying runs for each powder to determine a standard batch drying curve that could be used for the design of fluidised bed dryers. The comparison between the granule drying curve and the previous work is subsequently not entirely thorough and a more comprehensive study will be required to conclude that granules dry as well as powders.

6.6 Chapter closure

This chapter presents evidence for the feasibility of milk granulation as a performance-enhanced milk powder. Compared to spray dried powder, granulated milk has many favourable characteristics; better bulk density, a lower difference between loose and tapped bulk density, equivalent solubility, a lower fouling rate during UHT treatment of reconstituted milk and equivalent fat rise and sedimentation behaviour. Drying behaviour is slightly different, but not unfavourably so. Only the wetting behaviour of granules is poorer than for spray dried powder.

The bulk density measurements of the granules gave the most favourable credence to the use of milk granules as an alternative to spray dried powders. The change in bulk density measured from loosely poured to tapped, as would be experienced in extended transport of a freshly packaged product, was found to be 5 % for granules compared with 32 % for spray dried milk powder of the same origin. The tapped bulk density of granules was also higher than spray dried powder by 0.12 g/cm^3. This gives granules

an economic advantage over spray dried powders because of the reduction in transport costs due to the reduced volume of product.

The solubility of milk was found to be unaffected by granulation suggesting that granulation does not alter the chemical properties of the milk powder. This was further confirmed through testing of reconstituted milk heated during UHT treatment and subsequently stored. The fouling of the UHT heat exchanger was found to be slightly less for reconstituted granules than for reconstituted milk powder signifying greater or no significant difference in heat stability for the granulated reconstituted milk. No significant difference was found in fat rise between reconstituted and UHT treated milk over two months storage. Also, no sedimentation was found in the storage showing that no irreversible denaturation reactions occurred to the milk components.

A study of the drying characteristics of granules was conducted but was limited to only one experimental run due to time constraints. This experiment found that there was no noticeable reduction in the drying rate of granules when compared with spray dried powders until the moisture content dropped below three percent moisture where the drying was slower for the granules. This may be attributed to a resistance to moisture movement caused by small pore sizes and the larger size of the granules when compared with spray dried powders. The final moisture content of the granules was 2.2 % compared to 1.8 % for the spray dried powder, for fluidised bed drying with 90 °C air. This does not have any practical relevance as dried powders are not normally dried below 2.4 % due to problems of fat oxidation at extremely low moisture contents.

Granules do not wet as well as instant milk powders. Wettability may be improved by altering granulation conditions to increase pore size. Excipient addition, such as the use of a surfactant, will also improve the wetting by lowering the contact angle. The granule pore size distribution was measured and it was found that the size of the pores in granules were very small, with the largest pores occurring at less that 0.1 microns. This may help explain the poor wettability performance of the granules as the infiltration of moisture is restricted at these small pore sizes.

Chapter 7 – Continuous granulation and further work

7.1 Introduction

The previous chapters show that granulation of milk powders is technically feasible and that the quality of the product is not affected by granulation and in some areas is improved. This chapter aims to further test the feasibility of commercially producing granules by investigating and simulating a continuous granulation process. Also included is a discussion of potentially useful further work that can follow on from this masterate project.

7.2 Continuous granulation

7.2.1 Introduction

Industry currently uses continuous processes to produce low-value products in high quantities such as fertiliser granules while batch processes are used for products with a high value such as pharmaceutical products. A continuous process may be more suitable for milk granulation as the production volumes are high and the cost of manufacture must be low. However, batch processes offer more control over product quality as each batch can be monitored independently. Before moving to a continuous process the effect of process parameters on product quality must be understood and minimised. In the following sections a brief investigation into continuous granulation is given with an attempt to determine whether continuous granulation will have a detrimental effect on the quality of the milk.

7.2.2 Continuous granulation process design

Continuous granulation involves producing and drying granules continuously through the use of a granulation circuit. Product not falling into the desired size range is recycled around the circuit. The recycle ratio is the flowrate of material recycled divided by the product flowrate and is generally controlled by a size classification device, such as a double screen, that feeds oversized material into a size reduction process before being returned to the granulator. Fines or undersized product from the screens are recycled directly to the granulator. The recycle ratio is controlled by the quality and size distribution of the granules desired as product but can also be dependant on the amount of material that is required to be recycled through the granulation circuit.

Two process designs have been devised for the production of granulated milk depending on whether a spray drier is to be included in the process. Granulation of spray dried powder will improve the quality characteristics described in chapter six, but is also applicable to the granulation of powder blends, affecting the same quality characteristics, but having the additional benefits of eliminating segregation and improving flowability. A diagram of a process including a conventional spray drier is shown in figure 7A.

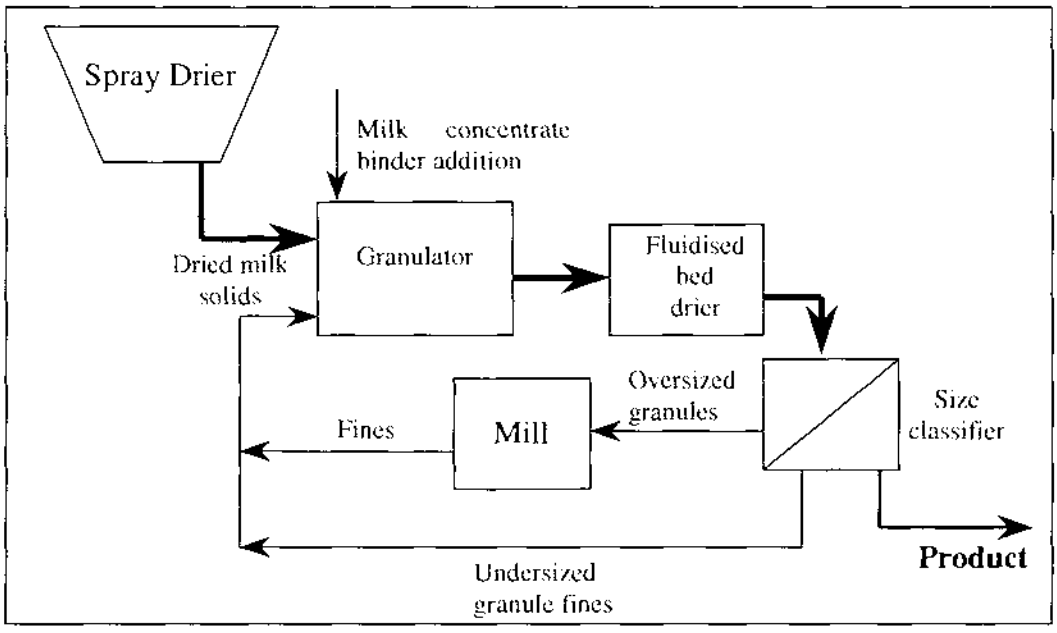


Figure 7A Continuous granulation circuit including spray dried milk powder in solids feed.

Spray dried milk powder constitutes most of the feed solids to the granulator, but a small amount is added as milk concentrate for use as a binder fluid. Ideally, the amount of recycle should be reduced to nothing to avoid re-processing the product and increasing the throughput. Product quality may be further improved through the use of a roller drum placed between the granulator and drier to reduce the granule size distribution; this will further spheronize the granules and will consume fines that survive granulation. It must be noted that vigorous particle movement in the fluid bed drier can have an adverse effect on the particle size distribution as has been shown in chapter three, therefore the operation of the fluidised bed drier must be carefully controlled.

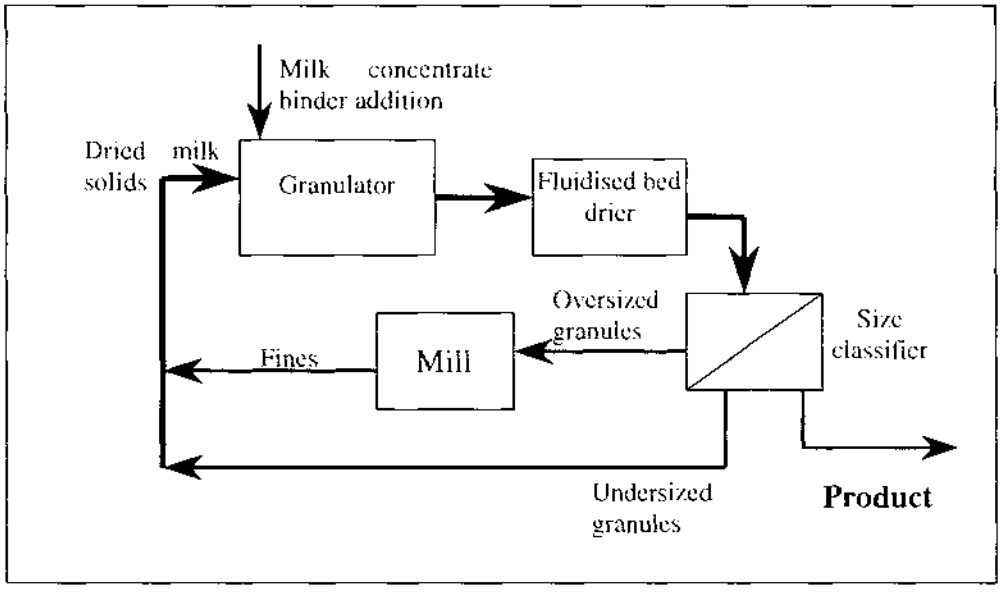


Figure 7B Continuous granulation circuit design using milk concentrate as the sole milk solids input.

Figure 7B shows a hypothetical granulation circuit that does not require a spray drier. Instead the milk concentrate is sprayed directly onto the fine granules and powder, which then granulate. The wet granules pass into a fluid bed for drying. This circuit is considerably smaller and hence less costly than existing spray driers. This is because granules do not require large volumes to keep 'tacky' droplets apart. However, the same amount of moisture will need to be removed as the process shown in figure 7A. This moisture will need to be removed in the fluid bed dryer. The economic significance of replacing a spray drier with a larger fluidised bed drier would need to be investigated to compare the capital costs of each process.

The process shown in figure 7B requires a fixed amount of milk solids to be resident in the process during operation. Start-up of the operation would require loading of the circuit with dried milk powder, preferably granulated product from a previous run. This is important because it will take some time before the initial powder charge has completely left the circuit. The product remaining in the circuit on shut-down will also be substantial and strategies will need to be put in place to reserve this powder, possibly as the start up charge. If this can be done effectively, minimal product will be lost.

The granulator will necessarily be a different design to that described in chapter three for the batch trials in this thesis. It will most likely be a Lödige style ploughshare granulator. After granulation, the granules need to be dried continuously in a fluidised bed. Such systems already exist in the dairy industry. After drying, the granules are classified to remove oversize and undersized particles which are recycled to the granulator. Ideally, both these processes require the inclusion of a continuous granulator, although using a batch granulation system is also possible by having a batch drying set-up or a series of batch granulators feeding a continuous drier. The design of a continuous granulator for carrying out further work to test these processes could be in the form of a Lödige granulation device as described in Parikh (1997), Hoornaert *et al* (1998) or Wellm (1997).

7.2.3 Mass balance and process design

An analysis of the process shown in figure 7B was undertaken by performing a mass balance over the proposed granulation circuit in an attempt to understand the desired recycle rate. This would give an indication of the processing time required for each unit operation so that further testing could be carried out on the effect on milk quality. A mass balance is not required for the process shown in figure 7A as the recycle rate is not a limiting factor. Figure 7C shows a diagram of the mass balance assuming typical moisture content values and where both oversize and undersize recycle streams are combined for simplicity.

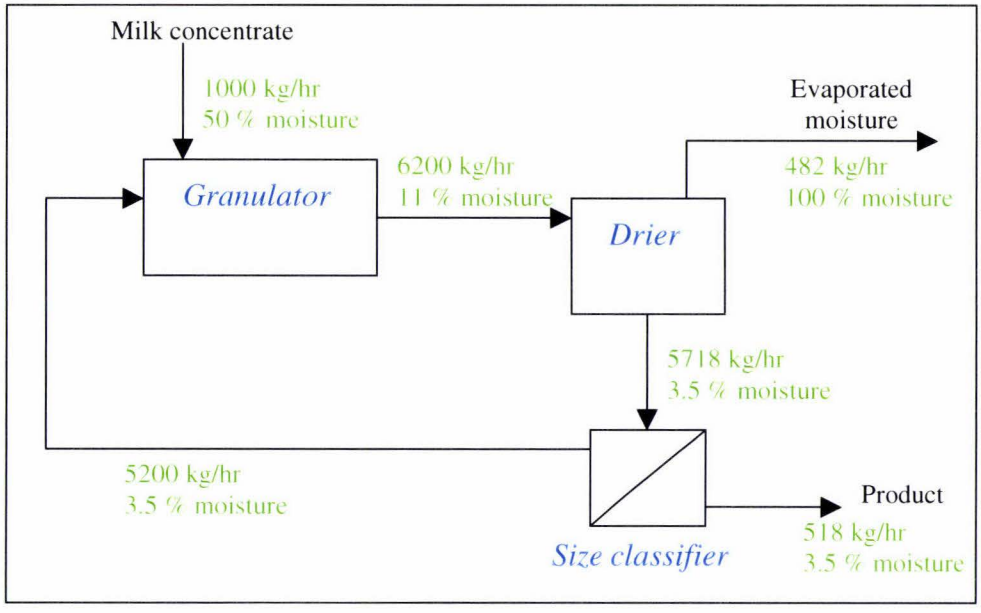


Figure 7C Mass balance diagram for continuous granulation circuit

The amount of product removed from the circuit is dependent on the amount of milk concentrate added. The recycle rate is therefore around ten times the product flowrate to ensure the bulk moisture content of the material flowing into the granulator is at the optimal 11 % level. This may be a disadvantage, as milk solids will likely be processed ten times that may result in a reduction of quality before exiting as granules. One possibility to reduce the effect of processing on the milk solids would be to reduce the amount of time the solids spend in the granulator. The feasibility of this would be best investigated using further experimental testing using a continuous granulation circuit.

Increasing the solids of the feed concentrate could further reduce the recycle rate. The effect of this on the quality of granules produced and the efficiency of the granulation is unknown as binder concentration was only able to be tested up to 47 % solids during this work.

7.3 Simulation of continuous conditions

An experiment was designed to test whether extended processing of milk granules resulted in chemical changes. The solubility index (SI), change in bulk density, and particle size distribution were measured for granules that had been through a batch granulation cycle and subsequently dried three times. This was done in an attempt to simulate the conditions that dried milk would be exposed to during a continuous granulation process as described by figure 7B.

As shown by figure 7C the milk powder is likely to be recycled through the granulation circuit around 10 times although due to the extended time taken for each batch run only three subsequent batch and drying steps were used. A granulation time of 5 minutes was used for each granulation stage was chosen to give a total granulation time of 15 minutes. This may be enough time to simulate the conditions for the process shown figure 7B if the residence time of the powder in the granulator was reduced to less than 90 seconds.

7.3.1 Experimental set-up

Spray dried whole milk powder was granulated using reconstituted whole-milk concentrate (WMC) as a binder. The granules produced were then dried in a fluidised bed drier as described in chapter 3, section 3.4.2. The dried granules were then re-granulated using WMC and subsequently dried two more times. The experiment took place over a two-day period with two granulation stages occurring during the first day and the final granulation and laboratory testing occurring on the second. The granules were stored dry overnight at 10 °C in a nitrogen-flushed plastic bad sealed by goosenecking. The experimental conditions are summarised in table 7.1.

Table 7.1 Experimental conditions for continuous granulation simulation

Mixer speed:	290 rpm for mixing and binder addition 700 rpm for granulation
Chopper speed:	1324 rpm
Amount of milk powder or granules added prior to granulation:	1.5 kg initial powder or maximum granules available from previous run
Binder added:	40 % total solids WMC to make granules up to 11 % total moisture*
Binder added after:	30 seconds
Length of granulation:	5 minutes granulation after binder addition
Drying conditions:	Fluidised bed for 2 hours at 30 °C

* The amount of binder added to each granulation run was estimated by assuming that the granules dried to 4% total moisture after 2 hours of drying.

After each granulation and drying run, a blending stage was added in an attempt to break up the larger granules, effectively simulating the milling process. The blending was done in a Sunbeam blender set on maximum for five minutes. However, this stage was removed after the first simulated loop of the circuit due to excessive heat build up and caking that occurred as the milk fat melted. The small proportion of the 1.5 kg batch that was blended in the sunbeam was omitted from the next granulation batch. This illustrates that milling must be done in a cooled environment. Further work is needed in this area.

7.3.2 Results and discussion

No difficulties were found in re-granulating the milk granules. After the first granulation stage the product was run through a 2mm sieve to remove the largest granules, which made up about 40 % of the weight. No large granules were removed from the 2nd and 3rd stages due to the reasons stated above.

It was difficult to estimate the true bulk moisture content of the material in the granulator. Previous work in chapter 5 shows the critical importance of moisture content; 11 % is optimal but 12 % leads to substantial product losses through caking

of powder to the sides and blades of the granulator. In these experiments, the moisture content of the granules entering the next simulated loop was estimated to be the same as previous granules dried in the fluidised bed drier. This was not correct and consequently too much binder was added leading to a higher final granule moisture content. Table 7.2 summarises the moisture content of the granules measured after each run and shows the resulting successive reduction in milk solids recovered from the granulator due to product loss caused by caking.

Table 7.2 Granule moisture content effect on amount of granules recovered from granulator

Run	Total milk solids entering granulator (g)	Moisture content of granules during granulation (%)	Wet granules recovered (g)	Milk solids recovered (g)
1st	1526	11.02	1700	1513
2nd	1560	11.88	1666	1468
3rd	1500	11.9	1576	1388

This slight difference in the amount of material in the granulator is likely to have an insignificant effect on the granulation performance (Theis and Kleineudde, 1999) compared with the difference in bulk moisture contents between runs as shown in chapter 5.

Table 7.3 shows the functional test results of the granules from the continuous simulation compared to the powder and batch produced granules. The only significant variation between the granules produced during this experiment and granules produced previously is the unacceptably high insolubility index measurement. This result is significant, as it has shown that the generation of insoluble material may not be completely explained by the storage and moisture hypothesis described in chapter 6, section 6,2,1. Extended processing of milk granules may therefore be a cause of product quality loss. This may mean that the granulation circuit shown in figure 7B could have difficulties with product quality although without further actual testing of a continuous granulation circuit it is difficult to make definite conclusions.

Table 7.3 Functional testing comparison of granules made using continuous simulation with batch granules and powder

Sample	Solubility Index (ml/50ml)	Loose bulk density (g/cm^3)	Tapped bulk density (g/cm^3)	Particle Density (g/cm^3)
Powder	0.1 ± 0.1	0.44 ± 0.02	0.58 ± 0.02	0.85 ± 0.01
Normal granules	0.1 ± 0.1	0.67 ± 0.02	0.70 ± 0.03	0.92 ± 0.01
Continuous simulation granules	0.5 ± 0.1	0.70 ± 0.02	0.72 ± 0.02	0.93 ± 0.01

7.3.3 Conclusions and recommendations

It is likely that the production of milk granules will require a feasible method of continuous granulation to achieve commercial success. The limited experiment carried out here using a batch process to simulate a continuous granulation circuit has shown that when milk powder is exposed to extended processing of granulation conditions a reduction in milk quality can occur. These conditions are more likely to occur in a process where powder is recycled through the granulation circuit many times, such as in the process described by figure 7B. To avoid over processing of milk solids, recycle must be kept to a minimum, which suits applications such as product blends, as indicated by figure 7A. Further work is required to determine if this reduction in quality is significant in a true continuous granulation process.

7.4 Further work

7.4.1 Skim milk powders

This thesis has focused almost singularly on whole milk powder. A very brief investigation into skim powder granulation found that the conditions required for skim powder granulation, if it is possible, differ greatly from those discovered as ideal for granulation of whole milk powder. In particular, rapid moisture absorption during skim powder granulation caused a significant and uncontrollable rise in impeller torque as binder was added. In these experiments 40 % solids reconstituted binder was added to skim powder in an attempt to make granules up to 11, 10 and 8 % total moisture with binder being added using the pulsing technique described in chapter 4, section 4.2.4 and with a larger period between pulses. None of the runs were successful and all had a hard and rubbery substance caking onto the blades and sides of the granulator when the experiment was stopped. During each run the binder addition could not be completed. Only a small fraction of the material was affected by caking with the rest being visibly identical to the original powder with no observable granules formed.

The most likely explanation of the failure of these experiments is that the components of the skim powder went through chemical changes resulting in the formation of undesirable products. Both lactose and spray dried milk proteins are hygroscopic and will undergo reactions if wetted. Lactose will undergo crystallisation while proteins will undergo gelation. Both of these reactions will result in an increase in the viscosity and shear sensitivity of the powder bed. Further analysis of these reactions during granulation conditions is required if skim powder granulation is to be attempted further. The crystallisation of lactose during granulation has not been investigated in this work but would be useful for further granulation work and may be critical for the development of skim milk granulation.

As has been described in the previous section, crystallisation of lactose is likely to cause problems during skim milk powder granulation while it does not appear to limit the granulation of whole milk powder. Further investigation into this phenomenon may be useful for further improvements into continuous granulation process design.

Chapter 8 – Conclusions and Recommendations

High-shear granulation of whole milk powder using a reconstituted milk concentrate binder was achieved using a 22-litre pilot-scale batch mixer-granulator. The granules had a higher bulk density than milk powder with less change in bulk density during storage. Granules were easily reconstituted using conventional reconstitution methods making granules an ideal replacement for spray dried milk powder as a reconstituted product. However, the granules made did not meet the wettability requirements for an instantised product suggesting that their use should be limited as a reconstituted product unless an improvement in wettability can be found.

The moisture content of the wet granulating mass was found to be the most important processing parameter with the best granulation occurring at 11 % total moisture. This value gives the greatest portion of granules within 250 μm to 1 mm and results in minimal product loss due to caking during granulation. An 11 % total moisture was found to be suitable using binder of varying solids concentration between 20 % and 50 % total solids with no significant differences between the granules produced. A reduction in the average granule size occurred at moisture contents lower than 11 %, while excessive product loss due to smearing and caking of product onto the granulator walls was observed at moisture contents higher than this.

Increasing chopper and mixer blade speeds results in an increase in the granule average size and a narrower granule size distribution. At high chopper speed the amount of material lost due to smearing and caking onto the granulator walls and lid increased. This was also observed with an increasing mixer blade speed, although to a lesser extent. Increasing the time of the granulation results in a narrowing of the granule size distribution with an increase in the average granule size. The mechanism of granulation after two minutes was found to be layering of fines onto pre-formed granules. A reduction in granule yield was also observed with an increasing granulation time due to an increase in the amount of material sticking to the walls of the granulator.

Apart from the wettability tests no evidence was found to suggest granulation affected the quality of the milk. Granules were stored without any observed degradation in quality for at least ten days provided drying occurred directly after granulation. Granules dried as rapidly as spray dried milk whole milk powder in a fluidised bed drier down to 2.2 % moisture. Reconstituted milk made from granulated powder was found to have a lower fouling rate during UHT treatment than spray dried reconstituted milk suggesting the granulated milk was more heat stable. Fat rise during storage of reconstituted UHT treated milk was found to be slightly lower for granulated powder compared with spray dried powder.

Multiple granulation stages caused an increase in the insoluble material of the granulation. This needs to be further investigated.

The effects of mixer speed, chopper speed and the amount of binder addition were found by investigating the batch granulation of spray dried milk powder granulated with reconstituted milk concentrate binder.

Although granulation is a seemingly easy process to understand and carry out, the fundamental physical and chemical processes are complex, especially for a complicated food product such as milk. Often, in the pharmaceutical industry, a large proportion of the ingredients added to the granulation recipe are excipients that are there solely to improve the physical properties of the powder mixture and have to functional use to the consumer. This project has shown that whole milk powder can be successfully granulated without any excipient addition with insignificant product loss during processing.

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