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Optimisation of granola breakfast cereal manufacturing process by wet granulation and pneumatic conveying

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Publications and Presentations

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Abstract

This study has considered the optimisation of granola breakfast cereal manufacturing processes by wet granulation and pneumatic conveying. Granola is an aggregated food product used as a breakfast cereal and in cereal bars. Processing of granola involves mixing the dry ingredients (typically oats, nuts, etc.) followed by the addition of a binder which can contain honey, water and/or oil.

In this work, the design and operation of two parallel wet granulation processes to produce aggregate granola products were incorporated:

a) a high shear mixing granulation process followed by drying/toasting in an oven.

b) a continuous fluidised bed followed by drying/toasting in an oven.

In high shear granulation the influence of process parameters on key granule aggregate quality attributes such as granule size distribution and textural properties of granola were investigated. The experimental results show that the impeller rotational speed is the single most important process parameter which influences granola physical and textural properties. After that binder addition rate and wet massing time also show significant impacts on granule properties. Increasing the impeller speed and wet massing time increases the median granule size while also presenting a positive correlation with density. The combination of high impeller speed and low binder addition rate resulted in granules with the highest levels of hardness and crispness.

In the fluidised bed granulation process the effect of nozzle air pressure and binder spray rate on key aggregate quality attributes were studied. The experimental results

show that a decrease in nozzle air pressure leads to larger in mean granule size. The combination of lowest nozzle air pressure and lowest binder spray rate results in granules with the highest levels of hardness and crispness. Overall, the high shear granulation process led to larger, denser, less porous and stronger (less likely to break) aggregates than the fluidised bed process.

The study also examined the particle breakage of granola during pneumatic conveying produced by both the high shear granulation and the fluidised bed granulation process. Products were pneumatically conveyed in a purpose built conveying rig designed to mimic product conveying and packaging. Three different conveying rig configurations were employed; a straight pipe, a rig consisting two 45° bends and one with 90° bend. Particle breakage increases with applied pressure drop, and a 90° bend pipe results in more attrition for all conveying velocities relative to other pipe geometry. Additionally for the granules produced in the high shear granulator; those produced at the highest impeller speed, while being the largest also have the lowest levels of proportional breakage while smaller granules produced at the lowest impeller speed have the highest levels of breakage. This effect clearly shows the importance of shear history (during granule production) on breakage during subsequent processing. In terms of the fluidised bed granulation, there was no single operating parameter that was deemed to have a significant effect on breakage during subsequent conveying.

Finally, a simple power law breakage model based on process input parameters was developed for both manufacturing processes. It was found suitable for predicting the breakage of granola breakfast cereal at various applied air velocities using a number of pipe configurations, taking into account shear histories.

Nomenclature

ε_i	local turbulent eddy dissipation rate, (m^2s^{-3})
ε_{th}	threshold turbulent eddy dissipation rate, (m^2s^{-3})
φ	aggregate restructuring coefficient, (-)
η	attrition propensity parameter, (-)
ξ	selection function, (-)
ξ_i	initial breakage ratio, (-)
ξ_f	final breakage ratio, (-)
ΔD_{50}	particle breakage along pipeline, (μm)
Δt	exposure time in Eq. 2.6, (s)
Br	breakage ratio in Eq. 5.1 and 5.2, (-)
c	correlation parameter in Eq. 2.2, (-)
d	particle equivalent diameter, (m)
d_0	initial median granule size, (mm)
d_T	median granule size after conveying passes, (mm)
E	particle attrition in Eq. 2.7, (-)
F	force in Eq. 3.1, (N)
F	fine particle increment rate, (-)
G	average shear rate, (s^{-1})
H	hardness, (Pa)
i	number of transport repetitions, (-)
K	constant, (-)
k	constant of the breakage model in Eq. 2.6, dimensions can vary
K_c	fracture toughness, ($Nm^{-3/2}$)
k_e	constant in Eq. (2.9), (-)
l	distance, (mm)
l_i	characteristic particle size, (m)
LL	linear distance, (-)
m	correlation parameter, (-)
n	Power-law coefficient for proposed breakage model, (-)
n	number of impacts in Eq. 2.3, (-)
p	breakage ratio distribution, (-)

S	relative granule diameter based on mass median diameters, (-)
t	exposure time, (s)
v	impact velocity, (ms^{-1})
v_{50}	median velocity, (ms^{-1})
v_a	air velocity, (ms^{-1})
v_p	particle velocity, (ms^{-1})
x	fraction of the initial feed, (-)
X_a	fine particle ratio after transport, (-)
X_b	fine particle ratio before transport, (-)

Chapter 1

Introduction

1.1 Introduction and background

Breakfast cereals have been defined as ‘‘processed grains for human consumption” (Fast, 1987). The importance of breakfast is well-recognized; accordingly, many people start their day with a bowl of their favorite ready-to-eat cereal. The major grains used in the manufacture of breakfast cereal are corn, rice, wheat, oats, and barley. Breakfast cereal products can be divided into: (1) ready-to-eat breakfast cereals; (2) hot breakfast cereal; and (3) alternative breakfast products based primarily on cereal grains, such as cereal bars, toasted pastries, waffles and other frozen products, muffins, and bagels (Caldwell et al., 2004). Ready-to-eat cereals are frequently made from mixtures of one or several grain components with other ingredients; they require extensive processing, are usually fortified with vitamins and minerals, and are specially packaged to protect their flavour, texture and nutrition during storage as well as to display their contents in such a way as to visually appeal to and entice the consumer. The global breakfast cereals market grew by 2.9% in 2008 to reach a value of $22,209 billion and ready-to-eat cereals dominate the market with 88.1% of the market share (Anonymous, 2009).

Granola is a breakfast and snack food consisting of rolled oats, nuts and honey which is baked until crispy, and is quite a friable material. Processing granola involves mixing the dry ingredients followed by the addition of a binder which can contain honey, molasses and/or oil. The product is then baked at temperature in the range 150 – 220°C until the granular product is toasted to the desired extent. Natural ready-

to-eat cereals are alternatives to consumers as they match both the health and flavour related requirements of consumers by being free of preservatives, additives and added sugar (LaGrange et al., 1991). To maintain sweetness, many cereal manufacturers use pure and natural honey, which imparts a sweet flavor and golden colour that many consumers prefer (LaGrange and Sanders, 1988). Granola sales have risen steadily because consumers are looking for products that are lower in fat and richer in healthy and more natural ingredients (Celis et al., 1996; Liesse, 1993). Such products contain healthy, natural ingredients (cereals, nuts and fruit) and can therefore contribute to a balanced diet.

In the present study granola is manufactured by two processes; one involves high shear granulation and the other involves fluidised bed granulation. Granulation is a size enlargement process in which small particles are formed into larger, physically strong agglomerates, in which primary particles are still distinguishable (Benali et al., 2009) and where the role of particle breakage is significant in terms of both yield and quality (Salman et al., 2003). After the granulation and baking stages the granola is passed through a Perspex built conveying rig. This rig is used to mimic industrial conveying during processing and packaging and is employed to better understand the degree and nature of breakage by the granola produced under various granulation processing conditions. The present dissertation thus aims to present a contribution to knowledge in the area of cereal granulation.

1.2 Research Objectives

The research objectives of the work are as follows:

- Conduct an experimental study on a laboratory scale high shear granulator. The study compromises a series of experiments, which investigate the influence of process parameters on key granule aggregate quality attributes such as granule size distribution and textural properties of granola in order to identify optimum process conditions.

- To do likewise during wet granulation using fluidised bed granulation by investigating the effect of various process parameters on key aggregate quality attributes.

- Investigate the effect product attrition in a post granulation conveying rig for different granulation stage processing parameters, different rig geometries and at different rig conveying velocities and different conveying times for granules manufactured by each of the above mentioned respective processes.

- Develop a simple model which incorporates key relevant processing parameters to predict breakage levels.

- Provide recommendations, based on the results obtained above, to improve final product quality. This can then act as the basis for optimization of granola manufacturing process.

1.3 Dissertation overview

Chapter 2 provides a general literature review. It begins by discussing the attributes of breakfast cereal in detail. Following this, a review of previous work in the field of high shear and fluidised bed granulation is presented. The granulation process of cereal granulation and operation conditions are discussed in detail. The advantages

and disadvantages of each of these operations are highlighted in this chapter. Finally, developments in pneumatic conveying of granular material are reviewed.

Chapter 3 investigates the effect of manufacturing process parameters on granule properties such as particle size distribution and textural properties of granola in high shear granulation. Various impeller speeds, binder addition rates, and wet massing times were respectively employed and their effects on effect on granule size, density, hardness and crispness were observed.

Chapter 4 investigates the effect of manufacturing process parameters (different nozzle air pressures and binder addition rates) on granule properties in fluidised bed granulation and proposes optimum operating conditions for the systems.

In chapter 5, the effect of manufacturing process parameters (granule history) and conveying rig parameters on attrition during pneumatic conveying of the granola were presented. Product attrition for different geometries and at different conveying velocities as a function of conveying time (number of passes through the rig) is presented. The effect of manufacturing process parameters during high shear granulation and fluidised bed granulation on conveying output parameters were also investigated.

Finally, chapter 6 presents the overall conclusions of this dissertation and makes some recommendations for future work in this field.

Chapter 2

Literature review

2.1 Breakfast cereal

The breakfast cereal industry has emerged as an important segment of the food industry. Remarkable progress has been achieved through a combination of knowledge gained in human nutrition, creative formulation, innovative progress in cereal processing technology and imaginative marketing. Breakfast cereal consumption increased significantly in continental Europe and in Japan during the 1990s, in both cases associated with growing indigenous manufacture and marketing. This has continued into the current century and is forecast to continue (Caldwell et al., 2004). The principal grains used in the manufacture of breakfast cereal are corn, rice, wheat, oats, and barley. Ready-to-eat cereals are produced from mixtures of grain components with other ingredients; they require extensive processing and are usually fortified with vitamins and minerals. They are specially packaged to protect their flavor, texture, and nutrition during storage as well as to display their contents in such a way as to visually appeal to and entice the consumer. In the case of granola, inulin is often used at about 5% with other carbohydrates to effectively bind the granola together, resulting in a higher-fiber, more crunchy cereal (Burrington, 2001). Honey is another ingredient widely used in granola manufacture. It is a commonly used food ingredient predominantly in the cereal, bakery and confectionery markets (LaGrange and Sanders, 1988) since consumers perceive it to be an all-natural ingredient thus adding value to products (LaGrange et al., 1991).

2.1.1 Textural properties of granola

Texture in ready-to-eat cereals is fundamental for product acceptance by consumers. Textural properties of food are used by consumers as key quality indicators that contribute to product acceptability (Szczesniak and Kahn, 1972; Szczesniak and Kleyn, 1963). Crispness is one such key indicator and is considered a primary textural attribute of breakfast cereal (Sauvageot & Blond, 1991; Martinez- Navarrete et al., 1998). Compression tests have been employed to evaluate crispness in ready-to-eat breakfast cereals (Nixon and Peleg, 1995). Cereal hardness is another force (Ferriola and Stone, 1998). Crisp and crunchy textures are generally expected by consumers and these can be imparted to raw materials in a number of ways (Solís-Morales et al., 2009) while Bourne (2002) reports that texture is an extremely important sensory acceptability factor of appearance.

Szczesniak (1990) claimed that crispness is the most important attribute affecting consumer acceptability. When it comes to measuring crunch, there is more than just eating the cereal (Burrington, 2001). If a crisp product does not produce the expected sound upon biting, then it is considered to be stale and of poor quality or has been produced using inappropriate ingredients or processes. It is important for scientists and technologists to have an understanding of the sensory perception of textural characteristics such as crispness in order to ensure that products are of optimal textural properties and exhibit characteristics of satisfaction to consumers. The mechanical properties of materials suggests that we should be able to predict the perceived texture of a food from the interaction between its mechanical properties and the processing conditions within the mouth (Vincent et al., 2002) .

2.2 Granulation

Granulation is a size enlargement process in which small particles are aggregated into larger, physically strong agglomerates, in which primary particles are still distinguishable (Benali et al., 2009). It is used in the dairy, food, chemical, agricultural and pharmaceutical industries and is a key step in many of these industries. Granulation can be desirable for many reasons. Gravity forces are increased faster than Van der Waals forces due to the size enlargement process resulting in improved flow properties. The amount of dust generated by powder handling is reduced, resulting in improved safety, since dust can cause explosions, and respiration of dust may cause health problems. It prevents the segregation of co-agglomerated components, resulting in an improvement in content uniformity. In addition, the compression and dissolution characteristics of the granules are improved. Finally, product attractiveness may be improved, which may be of importance in for example, the food or cosmetic industry (Hapgood et al., 2002; Knight, 2001; Lindberg, 1993). The process can involve either dry or wet methods. Dry granulation involves mechanical compaction (slugging or roller compaction) followed by a dry sizing process while a liquid binder is used in wet granulation processes. There are a number of wet granulation techniques: two of the most common are high shear granulation and fluidized bed granulation (Hegedus and Pintye-Hódi, 2007). Wet granulation is a complicated process, in which the resultant particulate properties (granule size and granule texture) is dependent on the process parameters and the physico-chemical properties of the materials used. The ultimate goal of research into wet granulation is to characterise the effects of process inputs on granule size and granule shape. To be able to manipulate granule size,

understanding of the granulation process and the effect of process parameters on the product is necessary.

2.2.1 Wet granulation

Wet granulation is a unit operation where fine primary particulate materials (powders, grains etc.) agglomerate due to a liquid binder to produce larger granules (Mackaplow et al., 2000). The process can be divided into several stages. After dry mixing of the primary particles ingredient the liquid binder is added to the dry mixture. This liquid can be sprayed or poured on the dry ingredients directly. The wetted particles bind other particles to form nuclei. Due to contact with other nuclei, the wall of the granulator, or other parts of the granulator (for instance impeller, chopper, or baffles) the nucleus may deform or densify (consolidate). Two nuclei can coalesce when the material is deformed. Consolidation enables liquid movement to the surface, which is then available for binding of other particles (Bouwman et al., 2005). Among all existing size enlargement operations, wet granulation in high shear mixers is particularly interesting as it allows one to obtain regular shaped granules with a high degree of compaction (Saleh et al., 2005). In wet granulation many techniques and different types of equipment can be used.

2.2.2 Granule growth

Granule growth in high shear wet granulation is a dynamic process in which granules are continuously forming and breaking down (Kristensen, 1988). The various mechanisms involved in wet granulation are known to be wetting and nucleation; consolidation and growth; and breakage and attrition (Iveson et al., 2001) as shown in Fig.2.1.

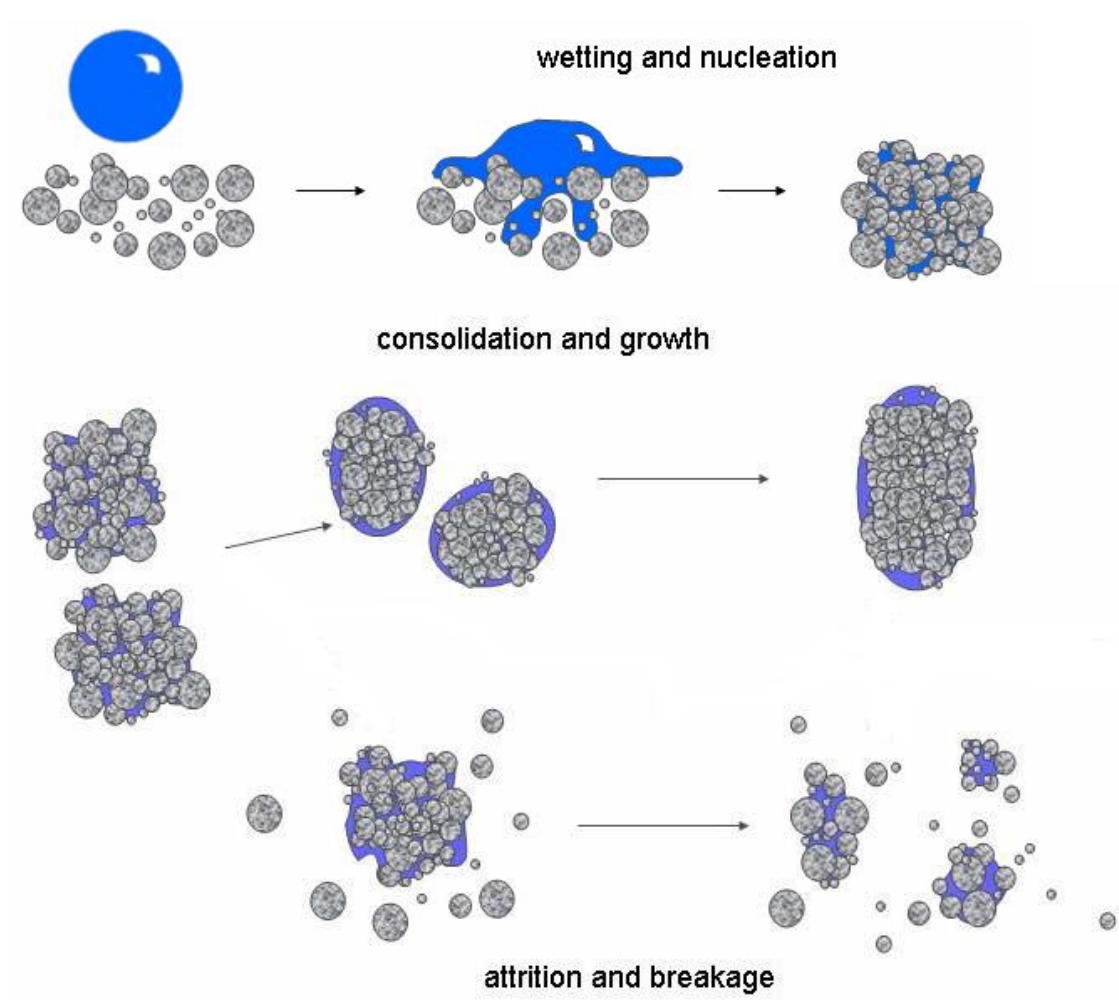


Figure 2.1: Schematic presentation of the different stages occurring during the wet granulation process (Litster et al., 2004)

1. Wetting and nucleation. Wetting occurs when the liquid drops are added and spread throughout the powder so that the air within the voids between powder particles is replaced by binder liquid. Wetting is a critical stage in determining the ultimate quality of the granules as poor wetting can result in much material being left ungranulated and requiring process recycling. Under good wetting conditions, the size distribution will be narrow and correlate closely with the liquid drop size distribution. Good wetting is also desirable as it ultimately leads to a narrower

granule size distribution. The nucleation phase of a wet agglomeration process is the initial phase where small agglomerates (nuclei) of loose structure are formed because of formation of liquid bridges between primary particles.

2. Consolidation and growth. Collisions between neighbouring granules, between granules and feed powder, or between a granule and the equipment lead to granule compaction and growth. The extent of the consolidation depends on the agitation in the granulator and the resistance of the granule to deformation. Consolidation results in the binder liquid being squeezed out from the pores of the granule to the surface and can thus only occur when the binder is still liquid. Granule consolidation determines the porosity and density of the final granules.

When two granule collide they may stick together to form a single large granule. This is growth due to coalescence. For successful coalescence (i) the energy of the impact must be absorbed during collision so that the granules do not rebound; and (ii) a strong bond must form at the contact between the colliding granules

3. Attrition and breakage. This refers to the inverse mechanism of coalescence where an agglomerate fractures into two or more similarly sized segments (Snow et al., 1997) where wet or dried granules break due to impact, wear or compaction in the granulator or during subsequent product handling of dried granules in the granulator, drier or in subsequent handling.

Breakage of wet granules will influence and may control the final granule size distribution, especially in high shear granulators. In some circumstances, breakage can be used to limit the maximum granule size or to help distribute a viscous binder. On the other hand, attrition of dry granules leads to the generation of dusty fines. As

the aim of most granulation processes is to remove fines, this is generally an undesirable situation to be avoided. Granule growth stops when the equilibrium between growth and breakage caused by impacts of particles, impeller, or wall is reached (Holm, 1997).

2.3 High shear granulation

High shear granulation is an effective means for turning powders into relatively dense granules. To create the granules, powders are added to a mixing bowl and the bowl is sealed (Fig.2.2). Dry ingredients are swept through the bowl due to the impact of a fast-rotating impeller. A large impeller spins at fairly slow speeds spinning the powders into a vortex. After the dry ingredients are blended together, liquid is added to be distributed over the powder particles. Primary particles stick together to form nuclei, which are consolidated by the impacts with the impeller and the wall. This consolidation of the nuclei pushes liquid binder to the outer surface of the granule, which can result in growth. An accompanying chopper tool located in the bowl spins at relatively high speeds shearing the granules and removing air. Large lumps are chopped into smaller pieces by the chopper, resulting in a smaller granule size distribution (Tardos et al., 2004).

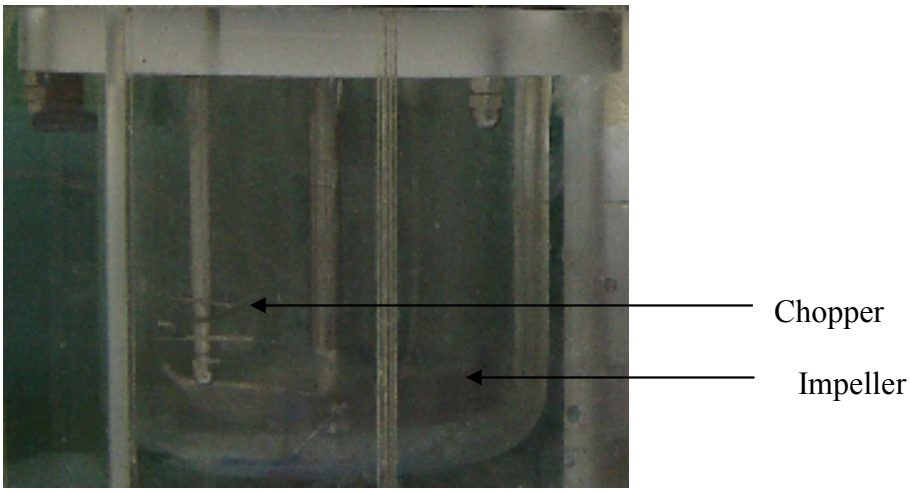


Figure 2.2: High shear granulator (4M8, Procept, Belgium)

Although the process is called high shear granulation, the high shear forces are actually not always 'high'. The mixer has the potential to produce high shear forces, but these forces are transmitted to the powder mass only if the powder is sufficiently cohesive or becomes sticky due to binder addition. Shear forces in the granulator are strongly dependent on the properties of the wet mass and they increase rapidly as soon as a 'granulation limit' is achieved; i.e. at the point where granules start to form a shearing powder mass (Litster et al., 2002). In high-shear granulation, an impeller is used to agitate the solid particles within a closed space. The binder solution is added or sprayed in from above. The impeller is used to exert high shear forces on the powder particles, which results in breakage, densification, and growth. The mixing continues until the desired end granule size and density is achieved. The process is ended before the granules begin to grow uncontrollably, which would result in the phenomenon known as "ball growth". In wet granulation, the amount of binder liquid used is usually defined as the ratio of binder liquid mass to the powder mass (Ennis, 1996). However, prediction of the appropriate amount of water to obtain the desired granule size is difficult, due to the fact that this amount is affected

by many variables. Examples of these variables are powder mixture properties, such as moisture content and particle size, and liquid characteristics, like viscosity (Knight, 1993). Knight et al. (2001) reported that an increase in (equilibrium) torque is found at increasing surface wetness of the granules. Increased wetness may be the result of increased impaction forces on the granules (e.g. a higher impeller speed) resulting in higher granule density or from an increase in the liquid content. The amount of liquid required is sensitive to the variations in the raw materials, complicating standardisation of the process. The end point is often determined via power consumption or torque on the main mixing tool drive. The granulated product may be dried in the mixing bowl, if equipped for vacuum drying, or discharged for fluidized bed drying. Therefore, high shear forces are only present from the moment when the powder is wetted and nuclei are formed (Litster et al., 2002).

The advantages of high shear granulation include the possibility of producing dense, spherical granules. Processing time is also short. Compared to fluidized bed granulators and conventional mixer granulators, wet granulation in high shear mixers is a robust process that can handle even very cohesive powders (Kristensen, 1996). Among all existing size enlargement operations, wet granulation in high shear granulator is particularly interesting as it allows one to obtain regular shaped granules with a high degree of compaction.

2.3.1 Parameters affecting high shear granulation

In high shear granulation, impeller speed and granulation time have been found to influence the following granule properties: size distribution (Biggs et al., 2003; Kokubo and Sunada, 1996a; Kristensen and Schaefer, 1987; Uribarri et al., 2003),

shape (Knight et al., 2000), porosity and friability (Badawy et al., 2000; Kiekens et al., 1999; Oulahna et al., 2003).

2.3.1.1 Impeller speed

The shearing forces acting on the powder mass depend on the inclination of the impeller blades, and on the rotation speed of the impeller. Schaefer et al. (1993) studied the effect of impeller shape on the granulation process. They found that curved impeller blades gave rise to smooth granules of spherical shape, whereas plane impeller blades caused product granules with irregular shapes. It was found that an increase in impeller speed improved granule homogeneity by increasing the extent of granule breakage for cases where the granule strength was low compared with the impact forces generated by the impeller. The mixing, densification and agglomeration of the wet material are performed by the impeller through the exertion of shearing and compacting forces. The shearing forces affect the critical agglomerate size that can be achieved while undergoing coalescence as well as breakage. Furthermore, the degree of consolidation of the aggregates, and consequently the liquid requirement, depend on the shearing forces. Changing impeller speed can have several effects. At prolonged granulation times, the granule diameter varies with the inverse square root of the impeller speed (Verkoeijen et al., 2002). However, this is not necessarily true. There is a balance between growth and breakage, both induced by the impeller. By changing impeller speed, this balance is changed as well. Increasing impeller speed can favour growth, thus leading to an increased granule size (Eliasen et al., 1999; Knight et al., 2000; Thies and Kleinebudde, 1999), or it can favour breakage, resulting in a decreased granule size (Seo and Schæfer, 2001). Impeller speed was also found to have an influence on

granule shape. Knight et al. (2000) observed that granules produced at high impeller speed for a short granulation time were less spherical than granules obtained at low impeller speed for a long granulation time. They concluded that the impeller speed and granulation time had to be optimized in order to obtain spherical granules.

2.3.1.2 Chopper speed

The shape of the chopper can also lead to breakage. The effects of impeller and chopper speeds on granule homogeneity were studied by van den Dries et al (2003). However, many authors reported that the speed of the chopper had no significant influence on granule properties (Holm et al., 1983; Holm et al., 1984; Westerhuis et al., 1997). Therefore, while the presence of a chopper could have an influence on granule properties, its speed is not likely to be of importance.

2.3.1.3 Wet massing time

Next to changing impeller speed, increasing wet massing time also has an effect on the granules produced. Increasing massing time also leads to more spherical granules (MacRitchie et al., 2002; Schæfer, 2001; Wan et al., 1994). For high impeller speed and/or long granulation time, granules are subjected to high-shear forces which leads to their densification, i.e., decrease of intragranular porosity and decrease in friability (Badawy et al., 2000; Kiekens et al., 1999; Oulahna et al., 2003; Tobyn et al., 1996).

Usually it is stated that the impeller is used to mix the powder and to make granules, and the chopper is used to chop large granules into smaller ones ingredients. The impeller and chopper are indeed designed for these purposes, however, both devices impact on the granules, so both devices can lead to growth and consolidation, or to

breakage. Therefore, the described effects of impeller speed below can also be read as effects of chopper speed.

For high impeller speed, the distribution of the binder solution in the powder or primary particle mix is improved. Consequently, a lower amount of binder solution is required to obtain granules with a narrow size distribution (Bardin et al., 2004; Holm et al., 1983; Holm et al., 1984; Knight et al., 2000; Schæfer et al., 1986). However, if the impeller speed becomes too high, fine particles can be generated from granule breakage due to mechanical stress. This can produce two major fractions in granule size distribution: a coarse agglomerated fraction on one side and a fine powder fraction on the other side.

Similarly, increasing granulation time increases granule mean size (Wang et al., 2008). However, after a certain time, breakage creates fine particles and leads to a wider size distribution (Kokubo & Sunada, 1996; Uribarri et al., 2003). When the speed of the impeller is too low, the inefficient distribution of the binder solution may lead to a wide particle size distribution and to the presence of ungranulated particles (Holm et al., 1983; Holm et al., 1984; Schæfer et al., 1986). Additionally, a short granulation time can be responsible for the poor distribution of the components. In particular, in the case of a dry binder at extremely short granulation times, the distribution of the solvent in the powder mass is not homogeneous and therefore these is poor binder dispersion (Kokubo and Sunada, 1996). Because of insufficient dispersion, weakly bonded particles are formed, thus increasing susceptibility to breakage. In addition, it is well recognized that the amount of granulating liquid affects granule growth; the greater the amount of granulating fluid,

the higher the granule mean size (Kokubo and Sunada, 1996; Konishi et al., 1996; Uribarri et al., 2003).

Based on these findings, impeller speed and granulation time must be concomitantly chosen to ensure to the resulting required properties of granules i.e., optimized sphericity, density, porosity, and size distribution.

2. 4 Fluidised bed granulation

Fluidised bed technology was developed in 1922 by Winkler for coal gasification, and since that time the technology has been extended into many areas of applications that require different constructions of fluidised bed apparatus (Mörl et al., 2007). Fluidised bed granulation is a widely used granulation process whereby granules are produced in a single piece of equipment by spraying a binder solution onto a fluidised powder bed (Fig. 2.3). The binder liquid is sprayed onto a bed of fluidised particles, whereupon having been wetted; the particles are bound together by liquid bridges. As the granules are dried in the same equipment without discharging, this process is sometimes classified as the “one-pot” system. The fluidised bed process enlargement has received considerable attention within the process industries, particularly food, agrochemical, dyestuffs, pharmaceutical, and other chemical industries, who have adopted this technology to address particle enlargement. Depending on the type of binder used, the liquid bridge will either solidify with cooling or dry with heating to form a solid bridge. The products manufactured in this way often have improved flowability and appearance, and sometimes have specific enhanced physical properties such as faster granule dissolution rates and higher strength (Tan et al., 2006).

Fluidised bed granulation is a complex process, influenced by several process variables (Rambali et al., 2001a). In order to develop a robust process, it is necessary to evaluate the effects of process variables at an early stage. Control of the fluidised bed agglomeration process is challenging as wetting, drying and mixing of particles all occur simultaneously in the bed. The different processes affect each other and are therefore difficult to control independently. It is necessary to understand the mechanisms involved and their relation to each other. Aulton and Banks (1981) classified several factors which influence the granulation process into three categories: equipment related, process related and product or formulation related. The following sub section will outline the relevant factors and their effect on fluid bed agglomeration; these can be used as a rule of thumb for designing an agglomeration process. Strict control of the fluidised bed granulation process is essential in order to get successful operation and desired end product quality in a reproducible way.

Figure 2.3: Laboratory scale of fluidised bed granulator employed in this study (Mini-Airpro, ProcepT, Belgium)

2.4.1 Process related variables

The process related factors which are used to control the fluid bed agglomeration process include fluidizing airflow rate (Gore et al., 1985; Smith and Nienow, 1983), inlet air temperature (Aulton and Banks, 1981; Ormos et al., 1973; Schaefer and Wørts, 1978a), relative humidity (Schaefer and Wørts, 1978a; Schaefer and Wørts, 1978c; Watano et al., 1995)and the nozzle operation (Ormos and Pataki, 1979; Waldie, 1991). It has also been shown that the choice of varying operating parameters such as inlet air temperature (Ehlers et al., 2009b; Faure et al., 2001; Jiménez et al., 2006; Rambali et al., 2001a), liquid feed rate (Bouffard et al., 2005; Ehlers et al., 2009; Faure et al., 2001; Hu et al., 2008; Jiménez et al., 2006) and atomizing pressure (Bouffard et al., 2005; Hemati et al., 2003; Jiménez et al., 2006)

have a significant effect on the quality of the end product, especially particle size distribution.

Charinpanitkul et al (2008) reported the influence of fluidizing air velocity, atomizing air temperature, atomizing air pressure and types of raw materials on granule physical properties. They found that the granules prepared at the lower fluidizing air velocity had larger average particle size. An increase in the atomizing air pressure resulted in an increase in amount of fine particles while the fluidizing air temperature has less effect.

2.4.1.1 Fluidising air velocity

The fluidizing air should not have a velocity too high, as fine powder will be blown out of the bed. This may result in clogging the filters. A fluidization velocity which is too low will result in defluidisation (wet quenching) of granules during agglomeration. It has been found that an increased fluidization velocity resulted in a smaller granule size due to increased evaporation and attrition (Aulton and Banks, 1981; Schaefer and Worts, 1978). In fact, the fluidization velocity, temperature and humidity have a direct effect on the moisture capacity of the fluidizing air (Gore et al., 1985) and through this on fluidization behaviour and granule growth. Agglomeration growth rate reduces with increased fluidizing air velocity (Aulton and Banks, 1978; Smith and Nienow, 1983; Tan et al., 2006). Tan et al. (2006) also showed that increased fluidizing air velocity tends to reduce the overall granule growth rate which might be due to several factors: (a) higher probability of particle rebound due to the increased impact kinetic energy, (b) increased liquid bridge rupture due to the increased agitation, (c) enhanced heat transfer that reduces the amount of binder available for agglomeration and (d) slower granule growth due to

the reduced amount of binder picked up per unit time at higher air velocity. The final granule size distribution was also seen to narrow with increased fluidizing air velocity and this is attributed to the increased powder renewal rate through the spray zone which allows a more uniform binder distribution. Tan et al (2005) reported that the higher air velocity formed stronger granules since a higher critical amount of binder is required to cause a successful particle aggregation in an environment with increased turbulence. An airflow rate which is too high results in result to attrition of the granules (Parikh, 1991).

2.4.1.2 Inlet air temperature

Higher inlet air temperature results in smaller granules because of the lower bed moisture content (Becher and Schlünder, 1998). The increase in fluidizing air temperature results in lower agglomerated moisture and a decrease in the wetting time (Dacanal and Menegalli, 2008). The effect of inlet air relative humidity on agglomerate growth and the particle size of the end product have been addressed by a number of authors (Hemati et al., 2003; Närvänen et al., 2008; Rambali et al., 2001a; Schaafsma et al., 1999), and the importance of this parameter has been frequently highlighted with the general understanding is that an increase in relative humidity of the inlet air yields larger granules.

A number of studies have shown that when the air relative humidity and/or liquid feed rate increased, larger granules were formed during granulation (Bouffard et al., 2005; Närvänen et al., 2008). Lipsanen et al (2007) studied the effect of dry and humid air on particle size. They reported that the batches of dry air had more small particles than granulations with high humidity, because in dry inlet air fewer liquid bridges are formed between particles.

2.4.1.3 Atomization air pressure

Granule growth was affected by atomization pressure through its effect on droplet size and spray pattern (Yamamoto et al., 2009). Atomization pressure of the binder during addition shows a linear relationship with granule size (Bouffard et al., 2005; Ehlers et al., 2009a; Jiménez et al., 2006). The increase in atomising air pressure decreases the binder droplet size and accordingly smaller granules are obtained (Bouffard et al., 2005; Hemati et al., 2003; Juslin et al., 1995a; Juslin et al., 1995b; Merkku et al., 1993; Yu et al., 1999).

Since liquid bridge formation among particles is the main mechanism for granule growth, it is reasonable to expect that spray rate and droplet size significantly impact on the wet granulation process, and influence granule size and size distribution. If the spray rate is too fast, rapid granule growth occurs, and the process could be sensitive to subtle changes of raw material and process conditions. Water-soluble components among the primary particles may be dissolved into the binder solution, thus accelerating granule growth. If the spray rate is too slow, it takes a long time to reach the desirable end point, and in worst cases this results in no granule growth. Large droplet size tends to produce larger and denser granules, while a fine mist may result in spray drying the binder solution. Spray pattern and atomizing air pressure influence binder droplet size. Distribution of the binder solution to the solid particle makes the solid particle surfaces sticky and deformable. When the solid particle size is greater than the binder droplet size, these sticky and deformable wet particles stick together, and this becomes the dominant nucleation mechanism. An increase in binder spray rate was found to result in significant increase in granule size (Bouffard et al., 2005; Gao et al., 2002; Lipps and Sakr, 1994).

2.4.1.4 Binder flow rate

The liquid flow rate of the nozzle should be balanced with the evaporation rate. A high liquid flow will result in overwetting the bed. A low liquid flow rate or very small droplets may result in spray drying of the binder liquid. Spraying a binder solution onto particles in a fluidised state at an appropriate rate can provide uniform wetting of the particles to generate wet granules (Roy et al., 2010).

Schaefer and Wørts (1978a) reported that a broad spray pattern creates insufficient air and liquid mixing, which causes droplet size to become larger. Small changes in air-to-liquid mass ratio influence droplet size changes. As a higher spray rate allows greater number of droplets to be sprayed onto the starting material per unit time, this resulted in an increased number of liquid bridges and hence larger granule size (Wan et al., 1995a). With a lower spray rate, binder solution evaporated more rapidly and binding of particles was reduced. In addition, the longer granulation time as a result of a low spray rate exposed granules to attrition forces resulting in smaller granules.

Abberger et al (2002) examined the effect of droplet size and powder material particle size on granule size. They concluded that the mechanism of agglomeration depended on the ratio of the binder droplet size to the particle size of the solid powder. When the solid powder particle size is smaller than the binder droplet size, immersion of the solid particle in the surface of the binder solution is the main mechanism of nucleation.

2.4.2 Equipment related variables

Fluidised-bed wet granulator performance depends on equipment related variables such as equipment size and shape, position of spray nozzle (top, bottom, tangential)

and number of spray guns, (Yamamoto et al., 2009). These variables need to be assessed during scale-up from lab or pilot scale to plant scale. The air distributor plate is responsible for the uniform distribution of fluidizing air throughout the bed. The pressure drop over the distributor plate should be at least one-third of the pressure drop over the bed to ensure uniform fluidization.

2.4.2.1 Air distributor plate

Ormos et al. (1973) reported the type and quality of the distributor plate had hardly any effect on granule properties such as mean granule size, size distribution or porosity. However Nienow et al. (1987) found that perforated plates and bubble caps minimize segregation (compared with porous plates) due to increased bubble size and a high local gas velocity.

2.4.2.2 Nozzle

The nozzle is an important instrument to distribute liquid onto the dry fluidised particles. Preferably, twin-fluid nozzles with external mixing are used to spray droplets, because the droplet size can be varied independently from the liquid flow and the chance of clogging is reduced by the extended liquid insert. The nozzle can be placed above, or in the bed, spraying sideways, upwards or downwards. Smith and Nienow (1983) used twin-fluid nozzles with internal mixing. They placed the nozzle in the bed at the bottom and were able to avoid caking of the nozzle by spraying upwards, although wet quenching occurred at lower liquid feed rates compared to top spraying. Davies and Gloor (1971) found that the decrease in the nozzle height increased the average granule size slightly and decreased the friability of the granules. This was explained by the binder's increased ability to wet and

penetrate the fluidised solids due to shorter distance. If the nozzle is located at too high a position, the risk of spray drying and wall wetting also increases (Hemati et al., 2003). In other studies, the height of atomizing nozzle or the nozzle diameter had only little or no effect at all on the fluidised bed granulation process (Cryer and Scherer, 2003; Rambali et al., 2001b). The nozzle determines the droplet size distribution, the spray rate and spray pattern upon the spray surface. The nozzle can be characterized independently of the granulation process itself, which makes it a good tool to control the process.

Agitators are also sometimes used to break up granules and thus establish an equilibrium between the growth rate of granules and the break-up rate, resulting in a narrow granule size distribution and higher granule density (Watano et al., 1995).

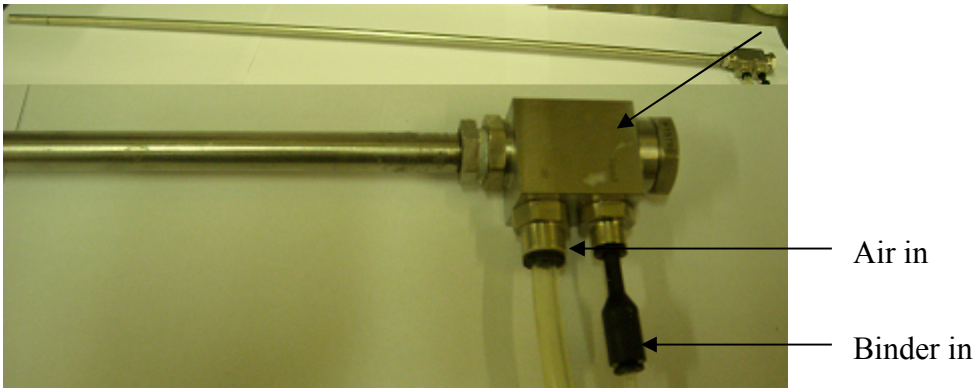


Figure 2.4: Nozzle used in the fluidised bed granulator employed in this study (Mini-Airpro, ProcepT, Belgium)

2.4.3 Product and formulation related variables

2.4.3.1 Starting material

The properties of the starting material have an important role in the fluidised bed granulation process. Since the wetting of the particles are essential in the formation of granules, the particle size of the starting materials affects the granulation. The fluidizing properties of the staring material (Schaefer and Wørts, 1978c), the wettability of the particles (Schaefer and Wørts, 1978a) in combination with the solvent, binder type and concentration (Schaefer and Wørts, 1978b; Schaefer and Wørts, 1978c) greatly affect agglomeration.

In fluidised bed granulation, the initial distribution of the binder among the bed of primary particles is critical. Wetting of particles is an essential phenomenon needed to form liquid and solid bonds between the particles that enable the growth of granules (Kristensen, 1988). However, if the moisture in the process is too high, granule growth can be excessive and the bed can even collapse due to the poor fluidizing capacity of the wetted mass (Schaafsma et al., 1999; Westrup, 1996). Owing to the much lower shear forces in fluidised beds, the liquid within the aggregates is less likely to be squeezed out, so coalescence will be limited. The initial wetting conditions are therefore very important in determining future granule size distribution (Faure et al., 2001). Schaefer and Wørts (1978c) investigated the influences of starting material property and its mixing ratio in the fluid-bed granulation process, using crystalline lactose and starch as raw materials. It was shown that the granule size depended on the ratio of lactose and starch, with higher lactose content generating larger granule size. It was also indicated that the higher

the surface area of starting materials, the smaller the granule size obtained after granulation.

2.4.3.2 Binder type

The wettability of the particles is important for the wet agglomeration process. Poor wettability results in smaller granules and these materials need higher spray rates to promote a slight overwetting of the material. Addition of surfactants premixed to the powder or to the binder liquid improves wetting (Aulton and Banks, 1981). The relationship between final granule size and binder concentration has been well established (Abberger, 2001b; Alkan and Yuksel, 1986; Bouffard et al., 2005; Davies and Gloor, 1971; Davies and Jr., 1972; Rajniak et al., 2007; Wan et al., 1992; Wan and Lim, 1991) and found a rise in granule size with increasing binder concentration. Schaefer and Worts (1977) have studied the relationship between binder viscosity, mean droplet size and mean granule size. The relationship between droplet size and granule size, under constant process conditions, is affected by the binder-induced mechanical strength of the wet granule liquid bridges. Wan & Lim (1989) reported that larger granule size and higher density is obtained when the binder is added as a powder, rather than by spraying as a solution.

The type of binders, and the binder concentration, could be critical variables when the binder solution is sprayed using a nozzle system. The binder type choice depends on properties which are required for agglomeration or for further processing. A binder is an essential component of the granulation process, as it provides binding ability in the formulation after drying. The binder can be added either as powder or sprayed as solution to the powder bed during the wet granulation process. In general, an increase in binder viscosity also results in increased aggregate size. The viscosity

of the binder liquid affects the droplet size, and thereby the size of the granule (Schæfer and Wørts, 1977). Gelatine for example has been found to form a portion of big granules even at low binder concentrations (Georgakopoulos et al., 1983; Rohera and Zahir, 1993).

The strength of a granule depends on the binder amount (concentration) and binder type. The binder should preferably be sufficient to withstand the rupture forces encountered in the fluidised bed. An increase of binder concentration was found to increase the granule size and reduce the number of smaller granules (Gore et al., 1985). The solvent of the binder solution can be either aqueous or organic, depending on the moisture sensitivity of the material to be granulated. Wan & Lim (1989) demonstrated that, after fluidised bed wet granulation, the granule size of lactose, a water-soluble excipient, is larger than the granule size of corn starch, a water-insoluble excipient.

The main advantage of a fluidised bed granulation is that it is a gentle, tunable and robust process during which many steps (pre-blending, granulation, drying) can be performed in the same piece of equipment. Usually, no milling is required after granulation. It is thus useful from a consistency perspective. Other advantages usually indicate the milder process conditions that prevail and product granules with higher porosity and narrower size distribution (Abberger, 2001; Boerefijn and Hounslow, 2005; Bouffard et al., 2005; Cryer and Scherer, 2003; Guignon et al., 2003; Panda et al., 2001; Pont et al., 2001). Although fluidised bed granulation is widely used to produce coarse solid particles in pharmaceutical, food and chemical industries, it continues to be one of the least understood processes among solid handling operations (Litster, 2003). These control parameters are all related to the

liquid concentration in the bed. The liquid concentration determines the degree of adhesion between particles and thus the aggregation process.

2.5 Pneumatic conveying of cereal granule

Pneumatic conveying is an important mechanism in the process industries for transportation of granular particles for example in conveying granulated material to packaging equipment. Typically, particles are transported along pipes using high speed air flow. There are two types of pneumatic transport systems: dilute phase transport, where particles occupy less than 5% of the line volume, and dense phase transport where particles may occupy up to 50% of the line volume. In dilute phase horizontal pneumatic transport, particles do not travel forward in a straight line, parallel to the pipe wall. Although gravity pulls the particles toward the bottom of the pipe, they are ideally kept in suspension by turbulent gas eddies. However, as the gas flow rate is reduced or the solids flow rate increased, particles eventually deposit on the bottom of the pipe (Fan and Zhu, 1998). In pneumatic conveying systems, particulate materials with a wide range of different sizes and shapes are used in industrial settings. Particle attrition is common during pneumatic conveying and usually considered a problem for particulate products. The degradation of particulate materials during pneumatic conveying is a very common phenomenon in process industries. An unavoidable occurrence in dilute phase pneumatic conveying systems, the attrition and breakage of the solid particles could lead to two major problems (Datta and Ratnayake, 2007):

1. Change in physical properties such as particle size and size distribution and consequently the problem of flowability and poor product quality.

2. Generation of fine particles leading to loss of valuable materials in the form of dust, with attendant environmental problems.

2.5.1 Granule attrition mechanisms

Granule attrition, where the particles suffer wear as a result of collisions and friction, can occur during the transport of materials. The attrition is caused mainly by impact or shear loads. The particles during pneumatic conveying experience extensive impact loads mainly at the bends because the flow direction is changing. Therefore, an analysis of the attrition at the bends is important. The particles could break in a single collision if the impact load is higher than its strength. The collision velocity, the angle of collision and the elasticity of the collision are significant factors in determining the impact load (Kalman, 1999). Particles can also break or be damaged while subject to lower impact loads than their strength, when collision occurs a number of times. This is known as fatigue. Parameters affecting the attrition rate can be divided practically into three categories: the particle strength, operating parameters, and pipeline and bend structure.

Frye and Peukert (2001) reported two basic stress modes leading to attrition: impact and friction. These mechanisms coexist in every conveying system, with their relative contribution depending on the prevailing mode of conveying. In dilute phase conveying for instance, where most of the particles are suspended in the gas stream, impact is the proposed dominant mechanism, whereas in plug flow conveying, friction will be dominant.

Pitchumani et al. (2003) identified three different causative mechanisms as being important for particle degradation during pneumatic conveying (1) Damage that

results from abrasion or attrition of the surface that generates new particles which are much smaller than the initial product. This process can result from particle to particle and particle to pipe wall contacts. (2) Chipping, the mechanism where larger fragments of the material are removed from the particle. Corners and edges on the particle are susceptible to this mode of damage, since they represent potential weak points (Taylor, 1998). (3) At higher conveying velocities the product can fracture, and this may result when the material is diverted e.g. in a right-angled bend. Such hard impacts can fracture the specimen into a few large and similarly sized particles and several smaller particles (Klinzing et al., 1997). Impact and friction are present in every conveying system. However, the contribution of each depends on the type of conveying: both impact and friction stresses are common in dilute phase conveying while friction stresses are dominant in dense phase conveying (Frye and Peukert, 2002). Particles may undergo different attrition mechanisms, such as erosion which produces dust with a slight change in the original particle size, chipping, or breakage of the particle into two or more particles of nearly sizes (Kalman, 2000).

2.5.2 Parameters affecting breakage and attrition during conveying

A large number of parameters control and influence the particle breakage process. Parameters which influence particle breakage and attrition in pneumatic conveying are the particle strength (a function of particle material, size and shape), operating parameters (particle velocity, concentration and loading ratio) and the configuration of the transport pipeline (radius of curvature, construction material, type of bends and the number of bends) (Kalman, 1999).

Kalman (2000) found that smaller particles result in lower levels of attrition since the strength increases. He also emphasized that the strength of the particles could change during the conveying process, not only due to size decrease but also due to other properties as shape and moisture content. In dilute phase pneumatic conveying particle-to-wall impacts are perhaps the predominant cause of degradation since the particles impinge on the wall surface at high velocities (Datta and Ratnayake, 2007).

2.5.2.1 Conveying velocity

The gas velocity and therefore the particle velocity before a collision, determine the momentum which is a function of the impact load during the collision. By increasing the gas velocity, the impact load increases. Particle concentration also has a large effect by reducing the particle velocity at higher concentrations (Kalman, 2000). Salman et al., (2002) suggested that there exists a threshold velocity, below which particle breakage does not occur. Therefore, to avoid particle breakage it is essential that the velocity remains below this critical value. Many products cannot be conveyed in dense phase and at low velocity. Therefore, the conveying air velocity must be as close as possible to the minimum transport velocity.

Bell et al (1996) presented attrition experiments using salt in which the size distribution was measured online. They showed that the air velocity has the principal effect on the attrition rate, although the loading ratio and the bend structure also have some effects. Aarseth (2004) found that conveying velocity and the impact angle of the particles against bends and fittings in the conveying system are principal causes of damage to feed pellets during transportation while Taylor (1998) expresses particle attrition as proportional to the square of impact velocity, but also considers material-specific parameters such as particle hardness and fracture toughness.

Datta and Ratnayake (2007) studied degradation of maize starch during pneumatic conveying. Their work indicated that for a given air temperature condition, the variation of attrition rate was a complex function of air velocity and solids loading ratio. Also the attrition rate was found to an increase substantially with an increase in air temperature. Salman et al. (2002) reported that impact angle was an important variable in determining the extent of particle fragmentation. They found negligible breakage in the horizontal pipe due to very low impact angle against the wall. Long radius bends, therefore which give a lower impact angle, can be superior to those with short radius.

2.5.2.2 Pipe geometry

Bends and elbows play vital roles in giving pneumatic conveying systems considerable flexibility by allowing routing and distribution (Akilli et al., 2001). Kalman (2000) suggests that attrition is lowest in long radius elbows, since small particles can follow the air stream easily through the bend. Salman et al (1995) determined that damage caused by oblique impacts is a maximum at an angle of approximately 40°. The flow of particles through a bend can exhibit a sliding bed or a bouncing motion. With each impact in the bend, the velocity of the particle is reduced, so significantly that an acceleration zone is required to reaccelerate the particles exiting the bend (Klinzing et al., 1997).

During pneumatic conveying, particles experience impact stresses at bends, due to the change in the flow direction (Kalman, 1999). Kalman (2000) reported that significant damage is caused by the normal component and increasing the bend radius decreases the angle of collision. It is generally recognized that during pneumatic conveying the particles experience extensive impact loads at the bends

because the flow direction is changing. Frye and Peukert (2002) also found that by changing the ratio of bend radius to pipe diameter, particle attrition is reduced since normal stresses decrease, which outweighs the increasing tangential stress. In addition, Frye and Peukert (2005) suggest that particles with large diameters have high inertial forces and such particles do not follow the gas stream through the bend, but directly impact the outer wall of the bend.

Much research work has been carried out on bend design with the objective of prolonging the service life of the bend and reducing particle damage and degradation as a result of impacts in the bend (Wypych and Arnold, 1993). In addition to particle velocity, the rigidity of the bend and the angle of impact determine the impact load in an elbow. Chen and Soo (1982) revealed reduced attrition of wheat grains by suspending a copper plate with spring action in the elbows. Moreover, flexible (made of rubber) long radius bends give much lower attrition compared to rigid long radius bends made of steel (Kalman and Goder, 1998). Mason and Smith (1972) determined that the bend material affected the erosion rate and materials respond differently to angles of impact. Brittle pipe material has the maximum erosion for normal impacts, whereas ductile materials have the most erosion at 20° and little erosion at 90°. Kalman (2000) suggested that flexible walls can absorb more energy and therefore should result in lower attrition rates.

Yilmaz and Levy (2001) measured velocity of solid and mass concentration using a fiber optic probe in two different 90° pipe bends with bend radius to pipe diameter ratio of 1.5 and 3.0. Their investigations suggested a continuous rope-like structure that formed within the elbow and disintegrated further downstream into large and discontinuous clusters. A continuous stream of particles was observed near the post-

bend region, while the rope dissolves into loose, agglomerated particles further downstream.

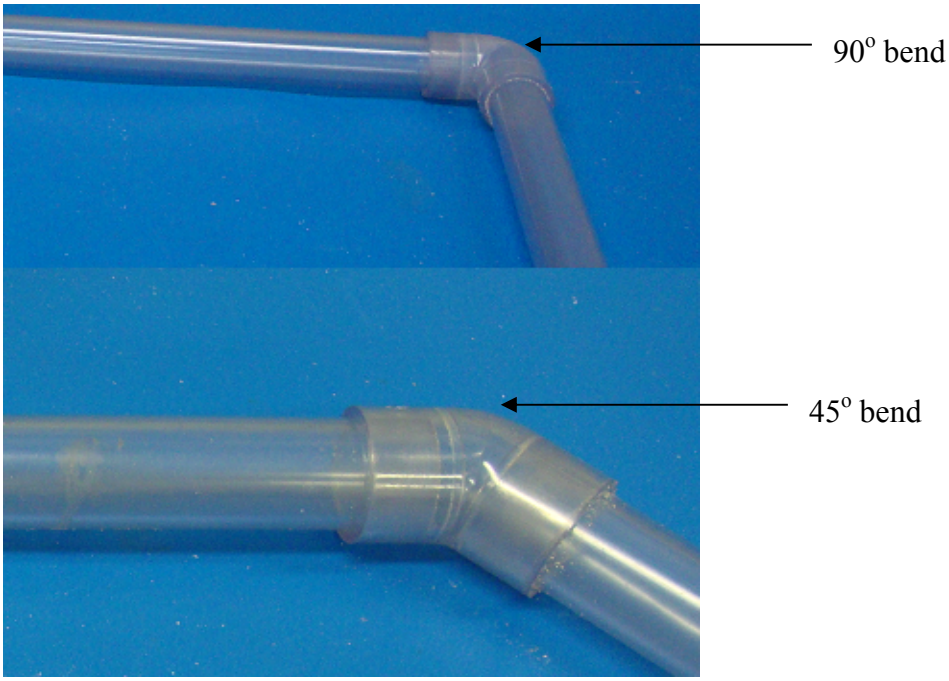


Figure 2.5: Bend geometries used in pneumatic conveying

Subero and Ghadiri (2001) investigated the effect of velocity and macro-voids in the particle on the breakage pattern in agglomerates. They reported that failure of the agglomerate is due to both cohesive and adhesive failure between individual particles in the specimen. During pneumatic transportation, particles are exposed to repeated impacts which can result in progressive weakening of the product as a result of macro-void or crack propagation (Tavares and King, 2002).

In terms of exposure time some researchers (Han et al., 2003; Heffernan et al., 2005; Kalman, 1999; Kalman, 2000; Salman et al., 1998; Zumaeta et al., 2005) have concluded that attrition rate decreases with time and tends towards some constant rate after a certain period of time. The high early rates of breakage are presumed to be due to the preferential breakage of the weaker particles at the start of the attrition

process. Once weak particles have shattered an almost constant rate process, mainly abrasion, occurs in which the particles are smoothed and rounded. Zumaeta et al. (2006) studied the breakage behaviour of whey protein aggregates formed under different agitation intensities and reported that particles formed at lower agitation intensity remain larger along the pipeline even after long exposure time to turbulent conditions than those particles formed at higher agitation rates.

In pneumatic conveying systems, mass flow rate is also an important parameter to measure. Mass flow rate is influenced by several factors that are reasonably predictable, however, the influence of the material properties is not fully understood at present (Mills et al., 2004). Arakaki et al., (2009) investigated the changes in dextrose monohydrate particles during repetitive pneumatic conveying pneumatically conveyed repetitively in a pilot plant–scale rig. The effect of the changes in shape and size of the particles on the mass flow rate in the system was studied. The mass flow rate measured in this system was not affected considerably by changes in size and shape.

The transport phenomenon of the conveying process in gas–solid system is not fully understood despite numerous studies, both experimental and numerical, which have been conducted on different pneumatic conveying systems to characterize the flow profiles of the solids in the pipes of different sizes and for different pipe bends. The breakage of particles in many pneumatic conveying systems can represent a major problem for particulate products. In some cases, due to conveying, the size distribution and appearance of the particles may change so significantly that the product no longer meets required specifications. Experimental and theoretical studies to understand the mechanical impacts of conveying upon granules is needed so that

formulations, the processing parameters and the pneumatic conveying systems can be optimized to avoid problems at the large scale.

2.6 Breakage prediction

The role of particle breakage is significant in many cases in terms of yield and quality of product, whether the desire is to cause breakage or avoid it. Considerable effort has been made to model particle and granule attrition by a number of researchers (Frye and Peukert, 2002; 2004; Han et al., 2003; Moreno-Atanasio and Ghadiri, 2006).

Frye and Peukert (2002; 2005) suggest that a model can be developed to predict the amount of attrition occurring in a pneumatic conveying system through process functions (conveying conditions) and material functions (material properties). Process functions are calculated using computational fluid dynamics whereas the material functions are determined through single particle experiments.

Aarseth (2004) conducted experiments on pneumatic conveying-attrition in order to establish an attrition model. This model could adequately describe the physical degradation of three heterogeneous materials that differ widely in susceptibility to attrition.

Ghadiri and Zhang (2002) developed a mechanistic model of impact attrition of particulate solids, having a semi-brittle failure mode. A dimensionless attrition propensity parameter (η) is derived from the above approach, whereby the extent of breakage is related to the material properties and impact conditions.

$$\eta = \frac{\rho v^2 l_i H}{K_c^2} \qquad (2.1)$$

where ρ is the particle density, v is the impact velocity, l_i is a characteristic particle size, H is the hardness and K_c is the fracture toughness.

Salman and other workers have conducted much research on the breakage rate of particles due to impact under various angles (Salman and Gorham, 2000; Salman et al., 1995). They found a simple function that is based on a Weibull distribution and provides a very close fit to most of their experimental data.

$$\xi = 100 - 100\exp\left[-\left(\frac{v}{c}\right)^m\right] \qquad (2.2)$$

where ξ is the selection function, v is the impact velocity, and c and m are correlation parameters.

Peukert and Vogel (2001; 2002) used an approach based on the work done by Weichert (1992) to define the selection function (Eq. 2.3). However, Eq. (2.3) can be related to Eq. (2.2) by defining the breakage ratio as a function of the specific energy (kinetic energy per mass) = $v^2/2$ that is multiplied by the number of impacts, n. The constant 'c' in Eq. (2.2) can be defined as the multiplication of an empirical material function and the particle size, $(1/c)^m = f_{\mathrm{Mat}} d$.

$$\xi = 100 - 100\exp\left[-f_{Mat}\, d\left(n\frac{v^2}{c}\right)^{m/2}\right] \qquad (2.3)$$

Petukhov and Kalman (2004) found the following function to fit well to experimental data for the selection function as a function of the impact velocity

$$\xi = \xi_f + \frac{\xi_i - \xi_f}{1 + \left(v/v_{50}\right)^p} \qquad (2.4)$$

where ξ is the selection function, ξ_i is the initial breakage ratio, ξ_f is the final breakage ratio, v is the impact velocity, v_{50} is the median velocity (the velocity that causes 50% of the population to break), and p is the breakage ratio distribution.

Zumaeta et al (2005) studied the effect of flow intensity and exposure time on particle breakage by recycling the particle dispersion through the 100mm pipe for 30 cycles. A power law model fitted the experimental data well.

$$d = C_1(t + C_2)^n \qquad (2.5)$$

where d is mean particle size, t is exposure time, n calculated for each flow rate. The constant C_1 and C_2 calculated as $C_1 = 2.5(d_0/v)$ and $C_2 = 1/v^{2.9}$ and d_0 is the initial particle size, v is the mean flow rate.

Zumaeta et al (2006) proposed a breakage model to determine breakage of the whey protein precipitate particles flowing through a turbulent pipe with sharp pipe diameter contraction.

$$\Delta D_{50} = k\varphi\left(\frac{1}{\varepsilon_{th}}\right)^n (\varepsilon_i - \varepsilon_{th})\Delta t \qquad (2.6)$$

where ΔD_{50} is particle breakage, (ε_i) local turbulent eddy dissipation rate, (ε_{th}) threshold turbulent eddy dissipation rate, Δt is exposure time, n is power law coefficient, $(\varphi \approx \exp(G))$ is aggregate restructuring coefficient and G is average shear rate.

Bradley (1999) suggested that particle attrition increases with increasing particle impact velocity. This relates directly to the deceleration forces experienced by the

particles when impacting against pipeline bends, alignments, etc. The degree of degradation or wear depends on the mechanical properties, shape and size of the particles and the mechanical properties and geometry of the pipe wall. The general trend is well documented as:

$$E = K(v_p)^n \tag{2.7}$$

where E is particle attrition, v_p is particle velocity, K is a constant and n is the exponent, which is related to the mechanical properties of the material.

Taylor (1998) expresses particle attrition as proportional to the square of impact velocity, but also considers material-specific parameters such as particle hardness and fracture toughness.

Konami et al (2002) investigated the changes of granules in the attrition process during repeated pneumatic transport. Eq. (2.8) describes the relationship between the fine particle increment rate and the mass median diameter of coarse granules.

$$F = \frac{X_a - X_b}{1 - X_b} \tag{2.8}$$

In Eq. (2.8) the fine particle increment rate F is defined as the rate of increase of the fine particle ratio (mass fraction) before and after transport, X_a is the fine particle ratio after transport and X_b is the fine particle ratio before transport. The fine particle ratio is defined as the ratio of the mass passed through the 150 μm sieve to the overall mass. Also they presented the effect of granule size on granule attrition. The relative granule diameter S was defined as the ratio of the mass median diameter of coarse granules before transport to that of coarse original granules. Coarse granules

refer to granules of over 150 μm in diameter. The relationship between S and F was investigated.

$$S_i - S_{i-1} = k_e (1 - F_i)^{1/3} \qquad (2.9)$$

where $k_e = (n_{i-1}/n_i)^{1/3}$ and i shows number of transport repetitions (-).

Gwyn proposed a means of describing attrition and stated that the attrition of initially monodispersed particles could be described empirically by Gwyn (1969). The initial rate is a function of initial diameter, whereas the decrease in attrition rate of a given size depends only on time. This equation has been verified by laboratory and commercial data.

$$x = Kt^m \qquad (2.10)$$

where x is the fraction of the initial feed that has undergone attrition at time t and m is an empirical constant.; K is another constant, which Gwyn suggested is a function of initial particle size.

Chapter 3

Effect of manufacturing process parameters on granule properties in high shear granulation

3.1 Introduction

Granola is an aggregated baked food product often used as a breakfast cereal containing ingredients such as oats, nuts and honey. It generally has a high degree of friability. Processing of granola involves mixing of dry ingredients followed by the addition of binder usually containing honey, water, molasses and/or oil.

Granulation is a size enlargement process widely used throughout the chemical and process industries. Among size enlargement operations wet granulation in high shear mixers is of particularly interest as it results in regular shaped granules with a high degree of compaction. It is therefore an effective means for turning particulate materials into dense granules. The process uses a liquid agent, called the binder, to hold particles together while they are mixed by mechanical agitation. In high-shear granulation, an impeller is used to agitate the solid particles within a closed space. The binder solution is added or sprayed from above. The mixing, densification and agglomeration of the wet material are performed by the impeller through the exertion of shear and compaction forces.

Granule growth in high shear wet granulation is a dynamic process in which granules are continuously forming and breaking down (Kristensen, 1988). The various mechanisms involved in wet granulation are known to be wetting and nucleation; consolidation and growth; and breakage and attrition (Iveson et al., 2001).

1. Wetting and nucleation, where the liquid binder is brought into contact with a dry powder bed, and is distributed through the bed to give a distribution of nuclei granules;

2. Consolidation and growth, where collisions between two granules, granules and feed powder, or a granule and the equipment lead to granule compaction and growth; and

3. Attrition and breakage, where wet or dried granules break due to impact, wear or compaction in the granulator or during subsequent product handling.

Several studies have investigated granulation parameters in high-shear mixers (Bajdik et al., 2008; Benali et al., 2009; Rahmanian et al., 2008; Tu et al., 2008) showing the effects of processing parameters on the growth rate of granules in the high-shear wet granulation process. Granule agglomeration tends to increase with an increase in liquid saturation of the granules, which is enhanced by the continuous addition of granulating solution or by granule consolidation during processing, while high impeller speed and long wet massing time tend to decrease granule porosity (Badawy et al., 2000).

Various tests have been developed to measure textural analysis of cereal based and snack food products. It is generally accepted that crispness, which is perceived through a combination of tactile, kinesthetic, visual and auditory sensations, represents the key texture attributes of dry snack products such as breakfast cereals or extruded crisp rice (Dacremont, 1995; Szczesniak, 1990). Breakfast cereals are products whose crunchiness or crispness is considered a primary textural attribute (Sauvageot and Blond, 1991). As outlined by Vincent (1998), crispness is associated

with a rapid drop of force during mastication which, in turn, is based on fracture propagation in brittle materials. The maximum force reached can be equated with hardness; the higher the force, the harder the material.

Granule growth rate is influenced by impeller speed, wet massing time and the amount of binder added (Badawy et al., 2000). Granulation parameters must be controlled in order to ensure the manufacture of a granulation with the desired particle size. The aim of this study, therefore, was to evaluate the effects of different processing parameters during wet granulation using a high shear mixer on key granule aggregate quality parameters such as size, hardness and crispness.

3.2 Materials and methods

The dry ingredients (Table 3.1) were mixed in high-shear granulator (4M8, Procept, Belgium) (Fig. 3.1) over a 2 minute mixing period at impeller rotation speeds of 150 rpm (tip speed 1.35 m/s), 200 rpm (tip speed 1.80 m/s) and 300 rpm (tip speed 2.70 m/s) and at 500 rpm chopper speed. Batch size was 100 g. This was followed by the binder addition stage (at the same respective impeller and chopper speeds), whereby 26 g of binder was added via a funnel onto the rotating ingredient bed at flow rates of 0.22, 0.33 and 0.65 g/sec respectively. A honey-water mixture (95:5) was used as binder. After adding the binder, the mixture was wet massed for a period of 6, 9 or 12 minutes at the respective impeller speeds. Each granulation experiment was conducted in triplicate. The wet cereal granules were then dried in an oven at 160°C for 10 minutes followed by a period of cooling in a desiccator.

Table 3.1: Granola ingredients

Ingredients	**Percentage**
Jumbo Oat flakes	25.39
Corn flakes	3.97
Puffed Rice	3.97
Malted buckwheat (milled)	3.06
Malted barley (milled)	3.06
Brown sugar	6.73
Oil	6.42
Honey	24.70
Water	1.30
Oat beta glucan	13.30
Wheat germ	2.75
Inulin	5.35

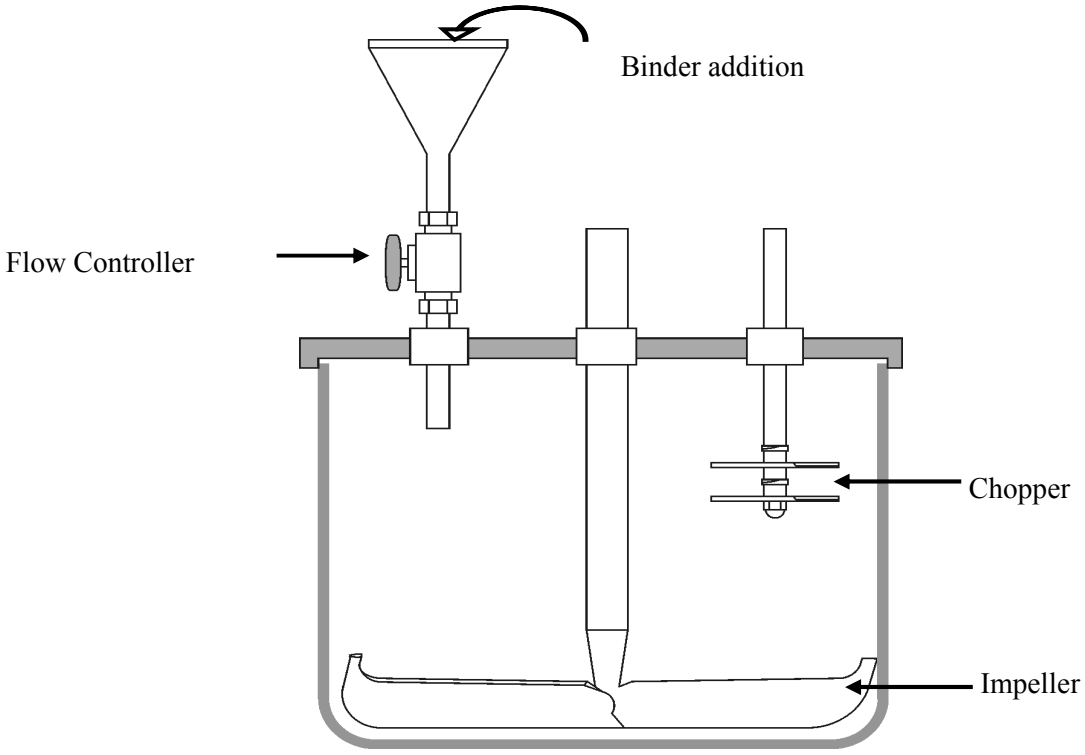


Figure 3.1: A schematic diagram of the high-shear granulator used for the production of granola

The granulation experimental parameters outlined above were chosen after preliminary granulation trials showed these were within the operating limits for the formation of cereal granules for the system under consideration. Fig. 3.2 shows an example of granola produced by this operation at impeller speed 300 rpm, wet massing time 9 minute and binder addition rate 0.33 g/s.

Figure 3.2: Granola produced by high shear granulation

3.2.1 Measurement of granola properties

Granule mass was measured using a digital balance (Sartorious ED4202S, Germany) with a sensitivity of 0.01 g. A Camsizer (Retsch, Germany) digital image analyzer (Fig. 3.3) was used for measuring granule size distributions and density (Heinrich et al., 2009). The measurement range for the Camsizer is from 30 µm to 30 mm. This unit works as a digital image processor. A sample is transported to the measurement field via a vibratory feeder where the particles drop by gravity between an extended light source and two CCD (Charge-Coupled Device) cameras. The two CCD cameras include a basic camera (CCD-B) which records large particles and a zoom camera (CCD-Z) which records smaller ones. There are three possible options for measurement: measurements using CCD-B, measurements using CCD-Z and measurements using both cameras. The measurement range of the CCD-B goes from approximately 400 µm to 30 mm. It is claimed (by the manufacturer) that the

measurement range of the CCD-Z ranges from 30 µm up to approximately 3 mm. The projected particle shadows (areas) are recorded at a rate of 60 images per second and analyzed to determine equivalent diameters (length).

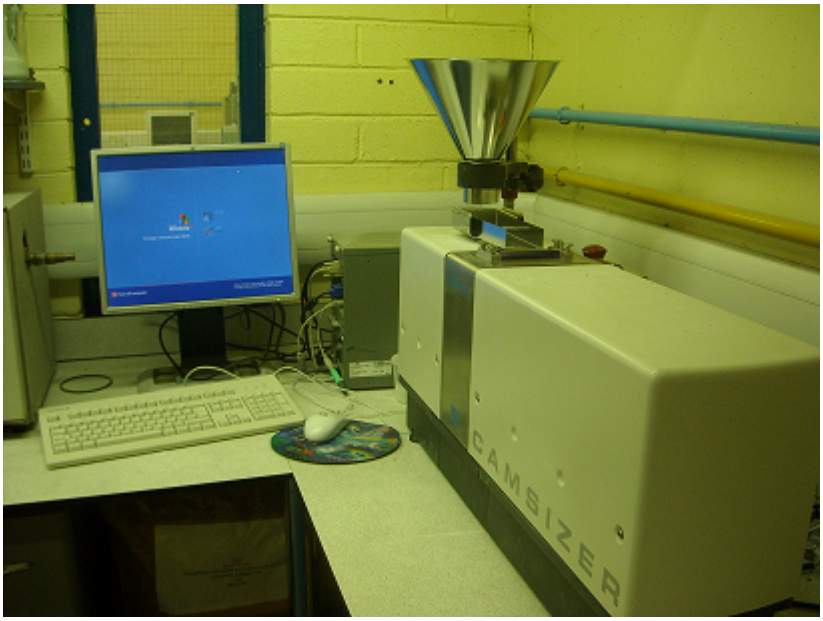


Figure 3.3: Camsizer digital image analyzer

Textural properties of samples were measured by using a TA-XT2i texture analyzer (Stable Microsystems Ltd, UK) equipped with an SMS P/75 compression platen (d = 75 mm) with 250 kg load cell. A flat-ended cylindrical aluminium plate of 75 mm diameter was used for compression. A representative sample is placed on the lower plate of the instrument and the flat-ended cylindrical plate moves downwards until it reaches a distance of 15 mm at a constant speed of 1.0 mm/s, a speed suitable for breakfast cereal experiments (Ferriola and Stone, 1998; Heidenreich et al., 2004). Each sample was measured in fifteen replicates. During each test run, the resistance of the sample was recorded and plotted on a force (N) versus distance (mm) graph (Fig. 3.4). The hardness (N) of the granola is a function of the highest peak and crispness is a function of the degree of plot fluctuations. The 'linear distance' (LL)

of the graph can be equated with crispness (Bajaj and Singhal, 2007). Linear distance is determined on a plot of force (*F*) versus distance (*l*) using the 'Linear Distance' function. This function calculates the line length by summing the lengths calculated between consecutive data points using the Pythagoras equation (Gregson and Lee, 2003; Heidenreich et al., 2004; Norton et al., 1998):

$$LL = \sum \sqrt{\Delta F^2 + \Delta l^2} \qquad (3.1)$$

Although a length is calculated, distance units cannot be used as one axis has units of force, and thus linear distance (or crispness) is considered dimensionless in terms of units used (Gregson and Lee, 2003). A highly jagged line, i.e. lots of fluctuations (peaks) in force due to many fracture events, is produced from products that are perceived as crispy or crunchy. The total length of this line is much longer for a crispy product if compared to the smoother line resulting from the testing of a softer, less crispy product.

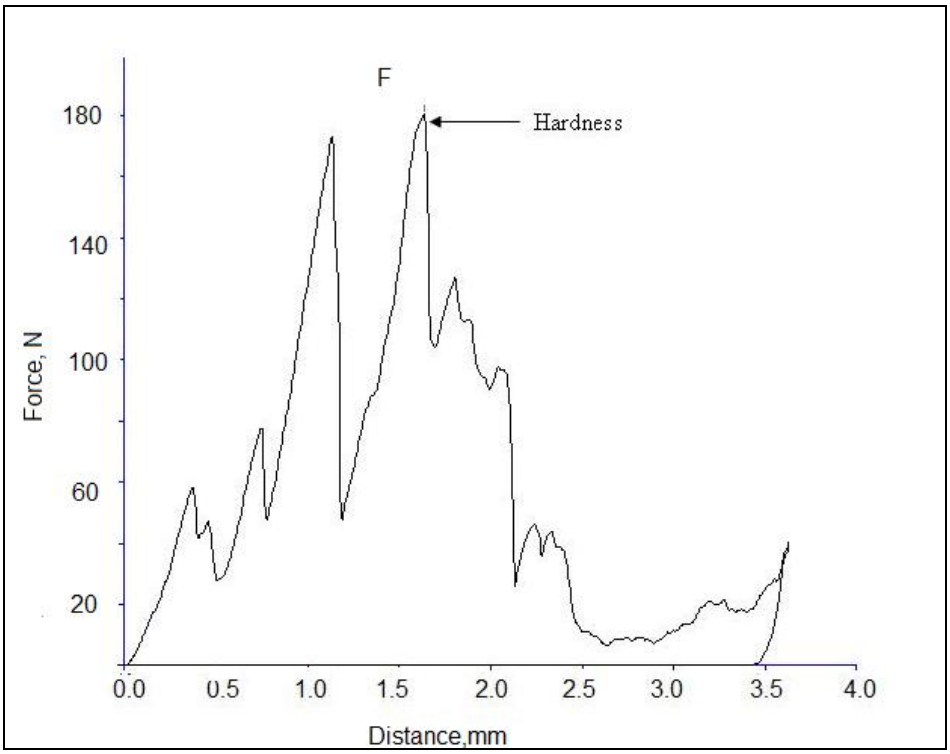


Figure 3.4: Typical force–distance deformation curves during compression of a granola particle

Data analysis was performed using Microsoft Windows Excel 2003 and Statistica 7.1 (StatSoft Inc., USA). Pareto charts were drawn to express visually the statistical significance of each factor and two level interactions between factors. The Pareto chart shows the standardized effects, which are equal to the effects divided by the respective standard error. In this way, statistical significance can be judged by comparing all standardized effects with the t-student value for the confidence level (95%) and degree of freedom. Linear effects are equal to the difference between the average of all data obtained at the two extreme settings of the factor. Quadratic effects are equal to the difference between the linear effect between intermediate and the low level and the linear effect between the high and the intermediate level. Interactive effects are half of the difference between the linear effect of one factor at one setting of another factor and the linear effect of the first factor at another setting

of the second factor. The vertical dashed line represents the critical *t*-value at the P = 0.05 significance level (see Fig. 3.9). The effects above this line are regarded as significant. The effects that fall below this line are not considered significant (Hill and Lewicki, 2006; Toutenburg and Nittner, 2002).

3.3 Results and discussion

3.3.1 Effect of processing parameters on granule size distribution

The process variables investigated were impeller rotation speed, wet massing time and binder addition rate. These process parameters are interdependent and a greater understanding of the relationship between parameters can result in the production of a more desirable granola product. The median granule size (d_{50}) of the granola produced in the study ranged from 6.3 to 15.9 mm. Typical granule size distribution is shown in Fig. 3.4. The influence of impeller speed (150, 200 and 300 rpm) on granule size distribution was investigated. Increasing the impeller speed to 300 rpm resulted in larger, coarser granules with a reduced proportion of fines (Fig. 3.5). The distribution span measures the width of the particle size distribution (Korhonen et al., 2002). Table 3.2 shows the distribution span for different processing parameter. A small span value indicates a narrow size distribution. A combination of high wet massing time (12 minutes) and high impeller speed (300 rpm) are the best combinations for achieving a narrow size distribution. Binder addition rate does not appear to have a significant effect on distribution span however.

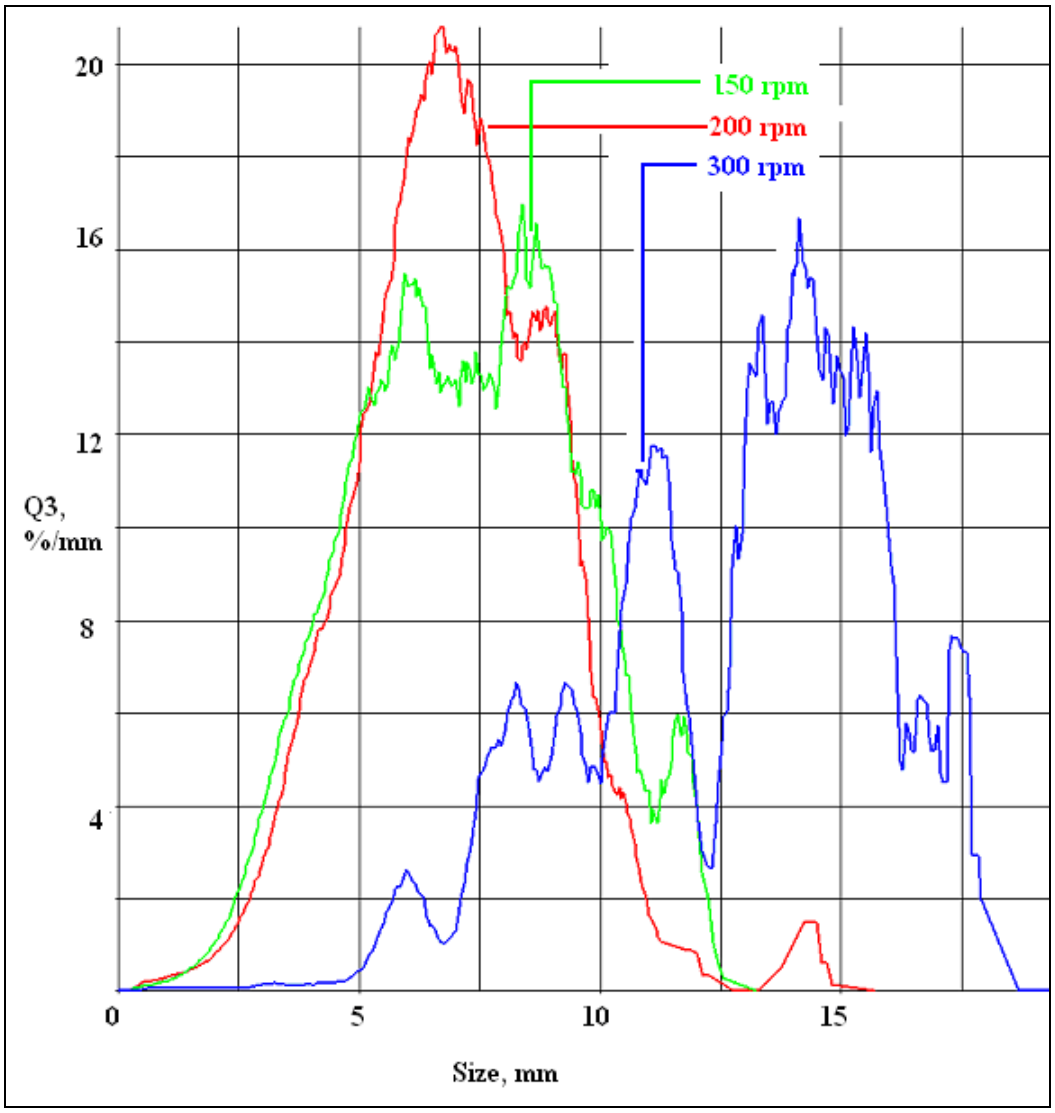


Figure 3.5: Granule size distribution at different impeller speeds at wet massing time 6 min and binder addition rate 0.22 g/s

Table 3.2: Distribution span

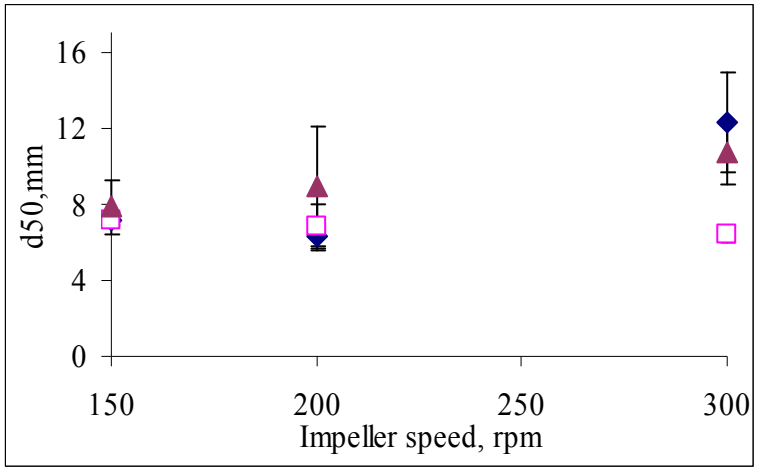
Impeller speed, rpm	**Binder addition rate,** g/s	**Wet massing time,** min	d_{50}**, mm**	**Distribution span,** $\frac{d_{90}-d_{10}}{d_{50}}$
150	0.22	6	7.1	0.86
200	0.22	6	6.3	0.78
300	0.22	6	12.2	0.67
150	0.33	6	7.2	0.95
200	0.33	6	6.8	0.89
300	0.33	6	6.4	0.86
150	0.65	6	7.9	0.99
200	0.65	6	8.9	0.74
300	0.65	6	10.7	0.68
150	0.22	9	7.8	0.78
200	0.22	9	6.8	0.98
300	0.22	9	14.1	0.54
150	0.33	9	7.5	0.80
200	0.33	9	6.8	0.97
300	0.33	9	6.4	0.72
150	0.65	9	7.2	0.89
200	0.65	9	9.0	0.73
300	0.65	9	15.1	0.55
150	0.22	12	8.5	0.74
200	0.22	12	6.4	0.85
300	0.22	12	15.9	0.51
150	0.33	12	8.4	0.78
200	0.33	12	7.6	0.85
300	0.33	12	11.4	0.54
150	0.65	12	7.5	0.77
200	0.65	12	8.1	0.73
300	0.65	12	14.3	0.48

3.3.1.1 Effect of impeller speed on median granule size

The effect of impeller speed on median granule size is shown in Fig.3.6. The highest impeller speed of 300 rpm results in larger particle sizes across almost all binder addition rates and wet massing times. The larger d_{50} is probably caused by the increased number of collisions resulting from high impeller speed, particularly in the case here where a very viscous binder is employed (i.e. low Stoke's number). With adequate binder rate and wet massing time present, the growth rate and final mean size can be controlled by the impeller speed. For high impeller speed, the distribution of the binder solution in the powder mix is improved. The size distribution becomes narrower at prolonged impeller speed (Table 3.2) and the narrowest size distribution is obtained at a combination of highest impeller speed, binder addition rate and wet massing time. This finding agrees with that of Schaefer et al. (1990) who found narrower granule size distribution obtained at higher impeller speed. This combination of operating parameters also leads to aggregates which are among the largest. Impeller speed affects the quality of the mixing between the particles and the binder, and the collisions between the particles or between the particles and equipment. Increasing the impeller speed leads to granule compaction and good binder dispersion which infers uniform wetting and improved granulation rate.

As might therefore be expected, lower impeller speed result in smaller granule size. The influence of the different impeller speeds on the size of the granules was more pronounced between 200 and 300 rpm than between 150 and 200 rpm. Overall, granule growth was faster at higher speeds and led to larger granules than at lower speeds. This finding is an agreement with that by Tu et al. (2009) who studied effect of impeller speed on granulation behavior of MCC 102 and found that the increasing

impeller speed leads to increased rate of granule growth and ultimately larger particles.



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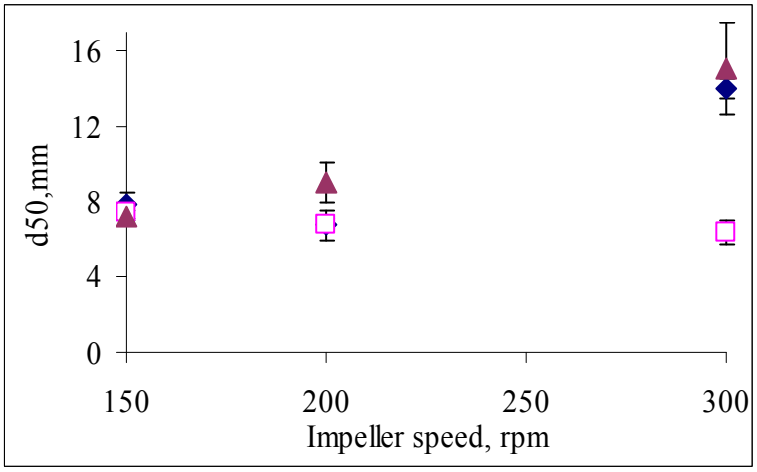

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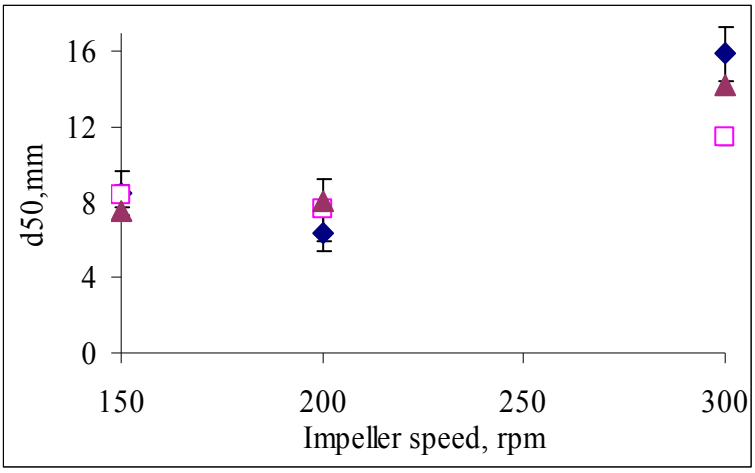


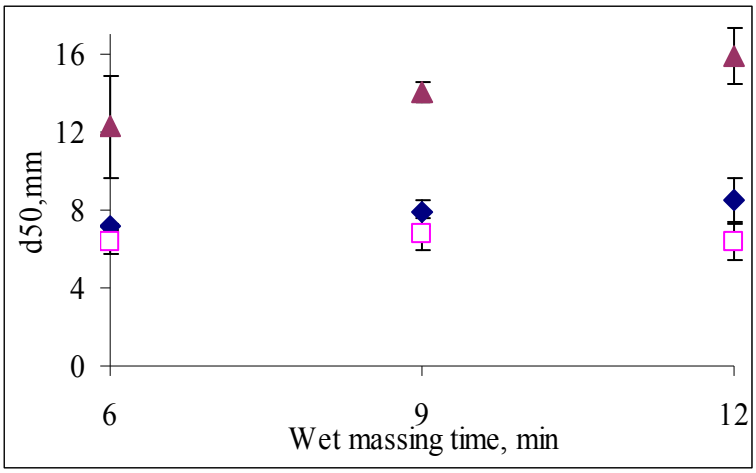
(c)

Figure 3.6: Effect of impeller speed during granola manufacture on d_{50} at binder addition rate of ◆: 0.22 g/s; □:0.33 g/s and ▲:0.65g/s: (a) 6 min, (b) 9 min and (c) 12 min

3.3.1.2 Effect of wet massing time on median granule size

With increasing wet massing time (Fig.3.7), there is a trend towards higher median diameters at high impeller speeds (300 rpm) particularly at lower binder addition rates (up to 0.33 g/s). This effect is not evident however at lower impeller speeds (up to and including 200 rpm). It appears that the combination of slow binder addition rate and high impeller speed leads to a good distribution of binder, and a high number of particle-particle impacts which may combine to result in aggregate growth of strong particles over time with corresponding low levels of breakage. This effect may be explained by the fact that a long wet massing period offers more chances of colliding which is necessary for granule growth (Bock and Kraas, 2001; Wang et al., 2008). Overall higher wet massing time and impeller speed results in higher granule size. Increasing the granulation time resulted in a shift in the particle size distribution towards coarser particles. This was attributed to improved wetting of the wet mass during the granulation process (Bock and Kraas, 2001). Conversely

granules produced over all binder addition rates (0.22, 0.33 and 0.65 g/s) and at relatively low impeller speeds (150 and 200 rpm) show no significant variation in d_{50} value over respective wet massing times.



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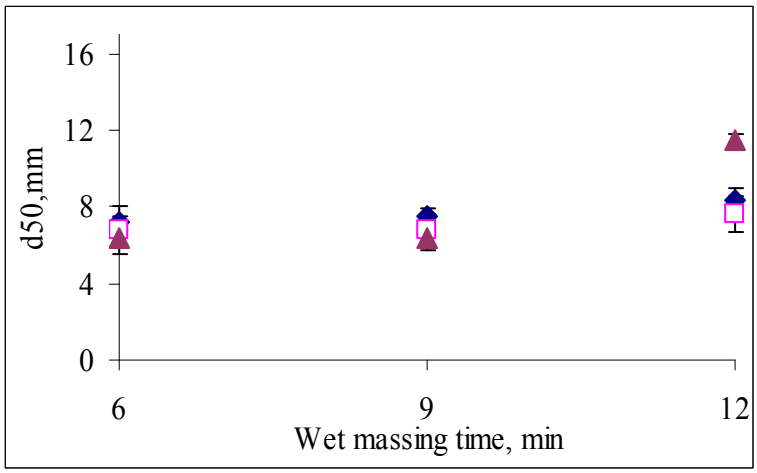

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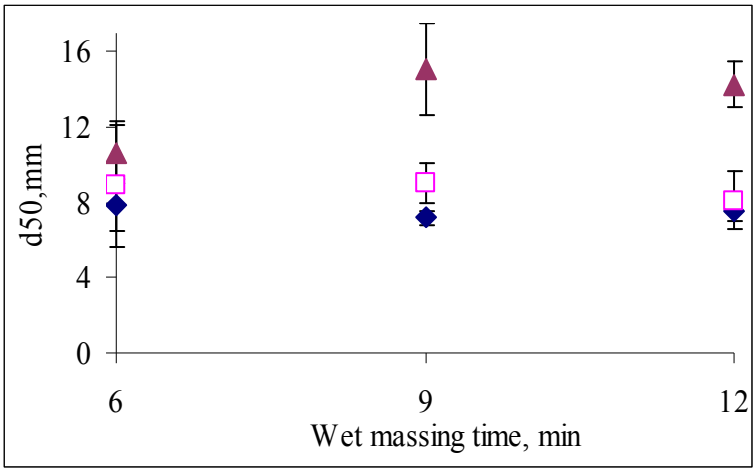

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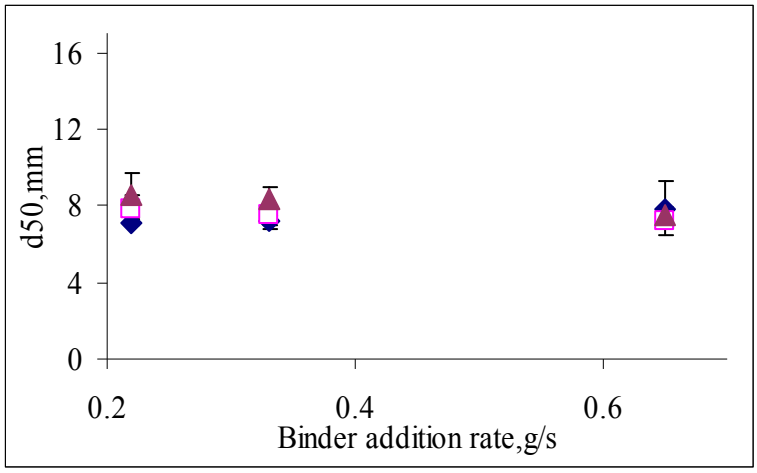
Figure 3.7: Effect of wet massing time during granola manufacture on d_{50} at impeller speed of ◆: 150 rpm; □:200 rpm and ▲:300 rpm: (a) 0.22 g/s, (b) 0.33 g/s and (c) 0.65 g/s

3.1.1.3 Effect of binder addition rate on median granule size

At the lower binder addition rate of 0.22 g/s the resultant d_{50} lies in the range 6.3 – 15.9 mm. At a binder addition rate of 0.33 g/s, this falls to between 6.4 – 11.4 mm and median granule size lies between 7.2 – 15.1 mm for highest binder addition rate of 0.65 g/s (Fig. 3.8). Unlike the effect of wet massing time and impeller speed, the binder addition rate does not appear to alter the median aggregate size appreciably over all processing input conditions studied.



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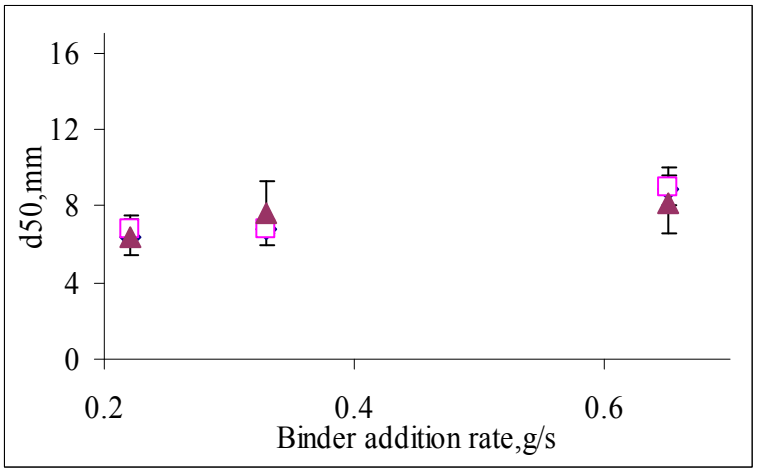

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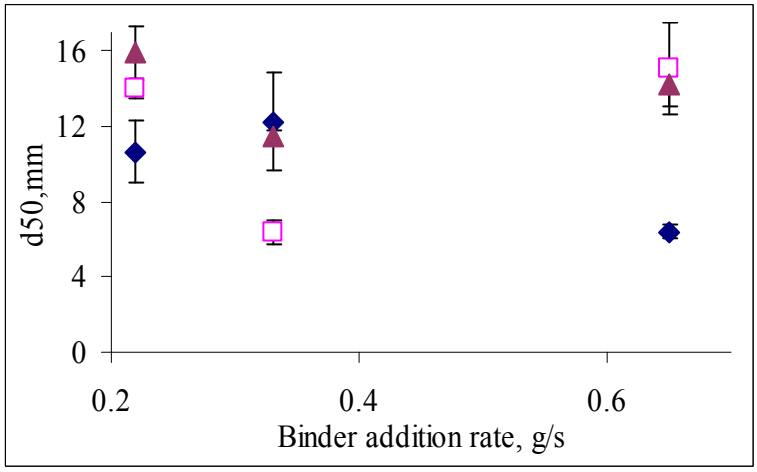


(c)

Figure 3.8: Effect of binder addition rate during granola manufacture on d_{50} at wet massing time of ◆:6 min; □:9 min and ▲:12 min: (a) 150 rpm, (b) 200 rpm and (c) 300 rpm

3.3.1.4 Statistical effects on median granule size

Statistical analysis was carried out to determine the most significant processing parameters, and from this Pareto chart was constructed (Figs. 3.9 – 3.11). The vertical line on the Pareto chart indicates the effects that are statistically significant at the 95% level. All three process parameters, impeller speed, binder addition rate and wet massing time were found to be statistically significant in term of granule median size (d_{50}) (Fig. 3.10). Similar trends were found at the d_{10} and d_{90} levels (Figs. 3.9 and 3.11). From the Pareto charts, d_{50} is indicative because of similar plots and representative of particle size of granola since the effect of processing parameters on d_{10} and d_{90} values broadly track d_{50} values in high shear granulation process. The interactive effects of process parameters have significant effect on granule size. A relationship between impeller speed and granule size has been shown before by several authors (Knight, 1993; Knight et al., 2000; Mangwandi et al., 2010; Schæfer, 2001; Schæfer and Mathiesen, 1996; Zhou et al., 1997). A higher impeller speed increases the rate of mixing and the number of effective collisions. During the granulation process in a high-speed mixer, primary particles or small granules agglomerate due to coalescence, and larger granules are broken due to the mechanical forces acting on them (Schaefer et al., 1986). These two effects counteract each other and the net effect depends on several factors such as the nature of the formulation, impeller speed and to a lesser extent, binder addition rate and wet massing time.

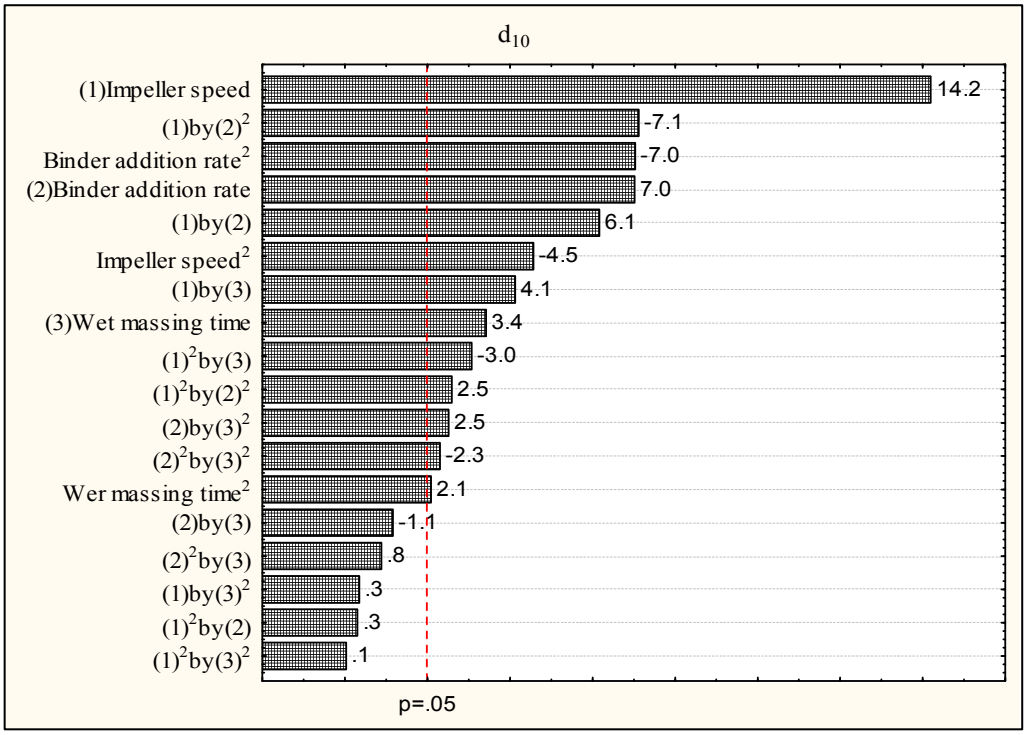


Figure 3.9: Pareto chart of standardized effects for d_{10}

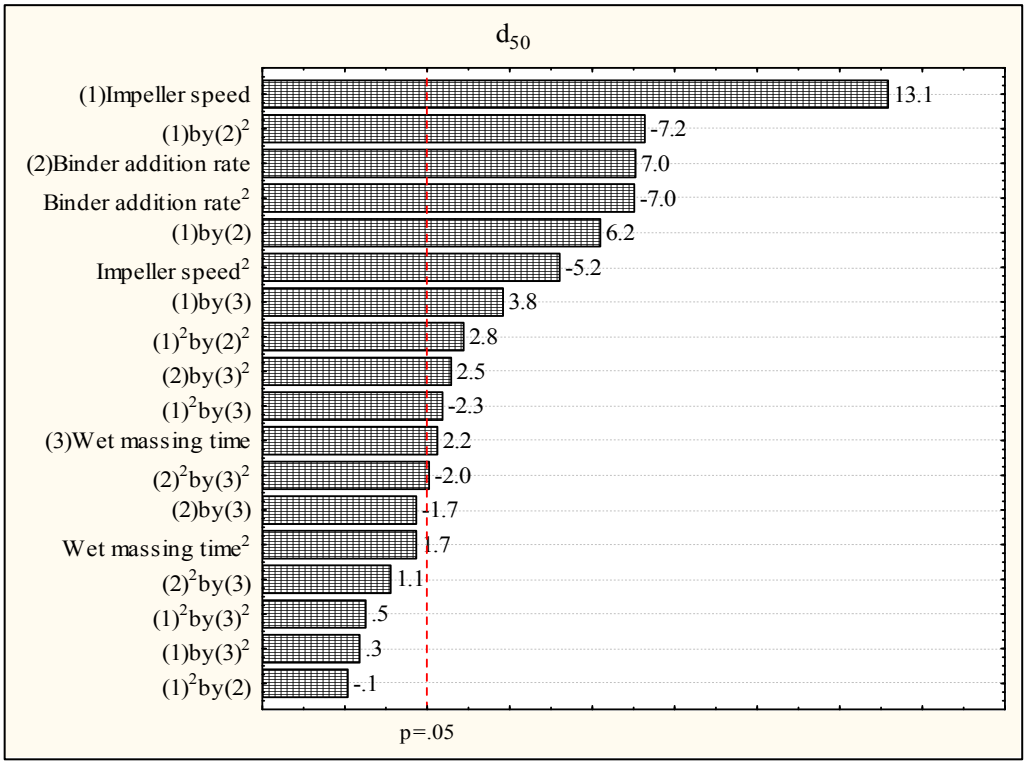


Figure 3.10: Pareto chart of standardized effects for d_{50}

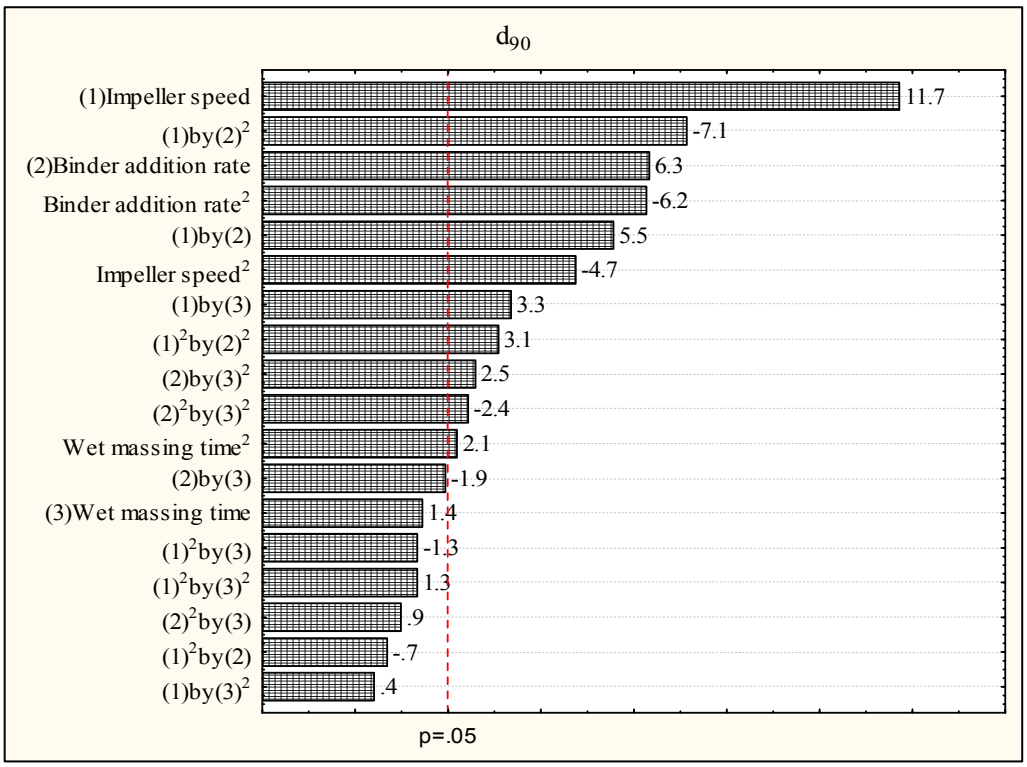


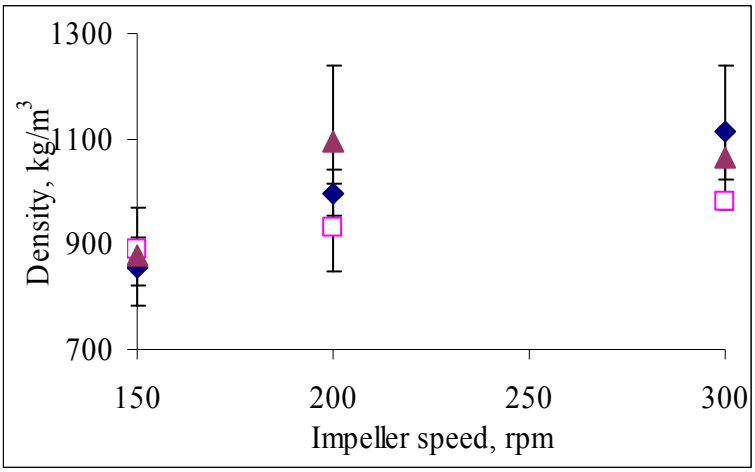
Figure 3.11: Pareto chart of standardized effects for d_{90}

3.3.2 Effect of processing parameters on granule density

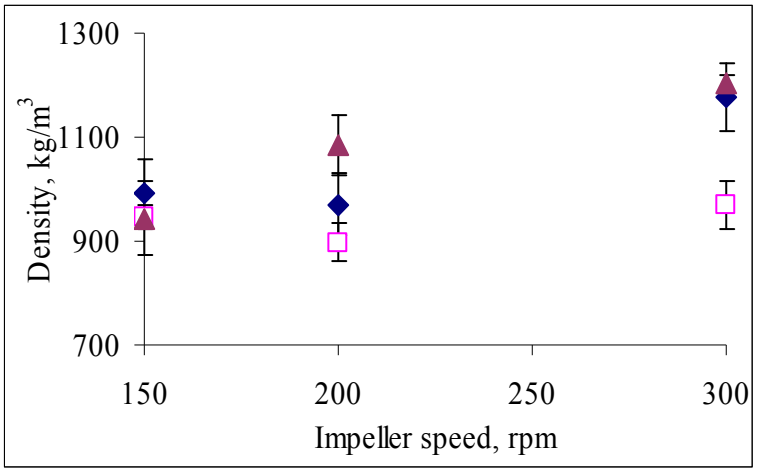
The effect of various processing parameters on granule density is shown in Figs. 3.12 – 3.14. Product density ranges from 923 to 1478 kg/m^3. The highest impeller speed of 300 rpm is shown to promote an increase in granule density with time, though this effect is less evident at lower impeller speeds (Fig. 3.12). The higher impeller speed would appear to enable the formation of dense, strongly rigid granules as a result of enhanced levels of contact between the respective particles bound by a high viscosity binder (Fig. 3.12). This concurs with previous studies (Badawy et al., 2000; Benali et al., 2009; Schæfer, 2001) which imply that high impeller speeds and long wet massing times can result in decreased granulation porosity and increased levels of consolidation and density by subjecting the granules to high-shear forces for longer periods of time. Consolidation is a very important rate

mechanism in granulation, as it controls not only the amount of air inside a particle but also controls the rate that binder is eventually forced out of pores. Impeller speed is a critical parameter which affects the granule density. Normally a low intragranular porosity is expected at a higher impeller speed. The lowest impeller speed of 150 rpm shows lowest density values in the range 856 – 998 kg/m^3 .This is as expected. The impeller speed of 200 rpm also results in similarly low densities, except at the highest binder addition rate where there is an increase in density evident with impeller speed across the full range of impeller speeds studied.

The consistently low density at low wet massing time and low impeller speed associated with all binder addition rates (Fig. 3.13) would appear to be as a result of poor mixing of the viscous binder at these lower operating conditions. This is particularly likely due to the viscous nature of the honey based binder. Wettability may also be a factor with this binder at these conditions. Davies and Gloor (1971) also reported that denser granules formed due to greater wetting ability of binder solution.



(a)



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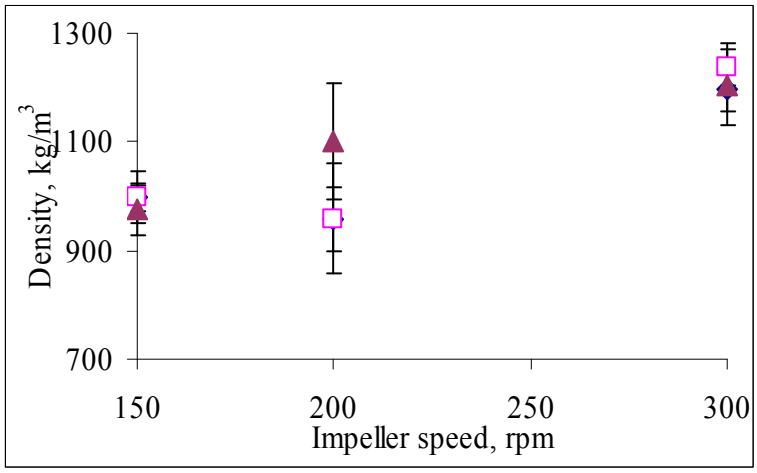
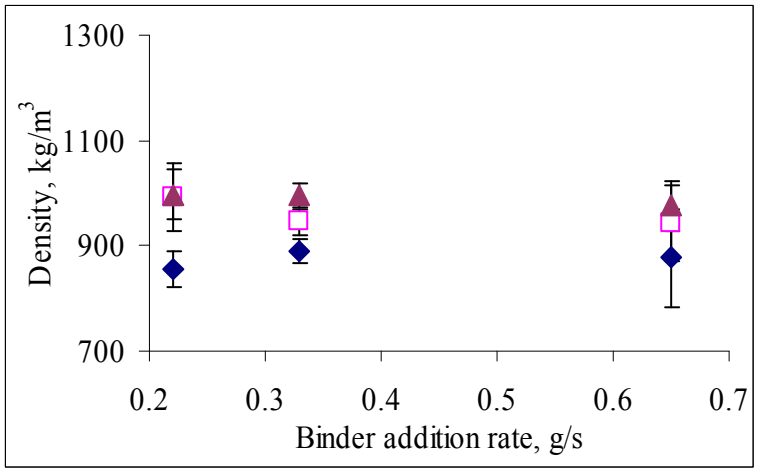


(c)

Figure 3.12: Effect of impeller speed during granola manufacture on density at binder addition rate of ◆: 0.22 g/s; □:0.33 g/s and ▲:0.65g/s: (a) 6 min, (b) 9 min and (c) 12 min



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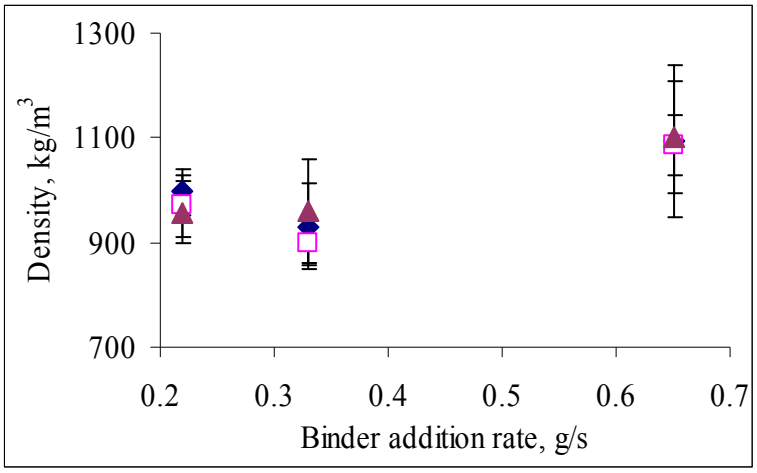

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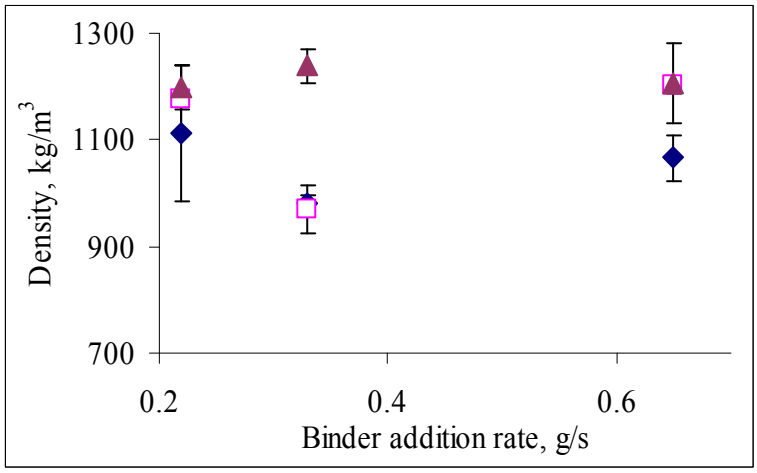
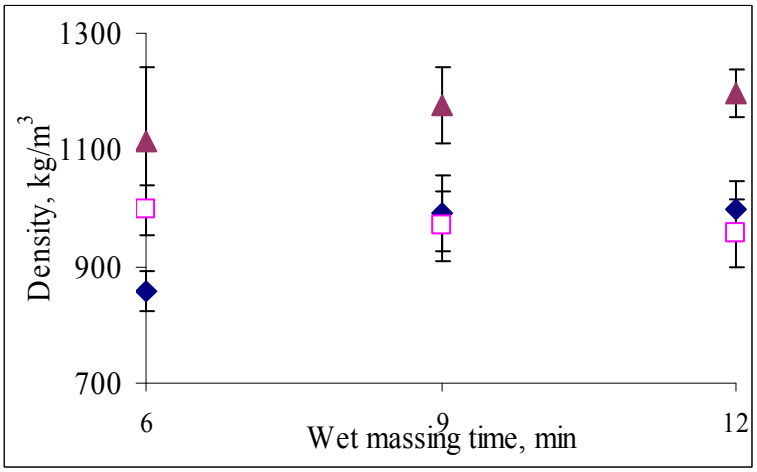
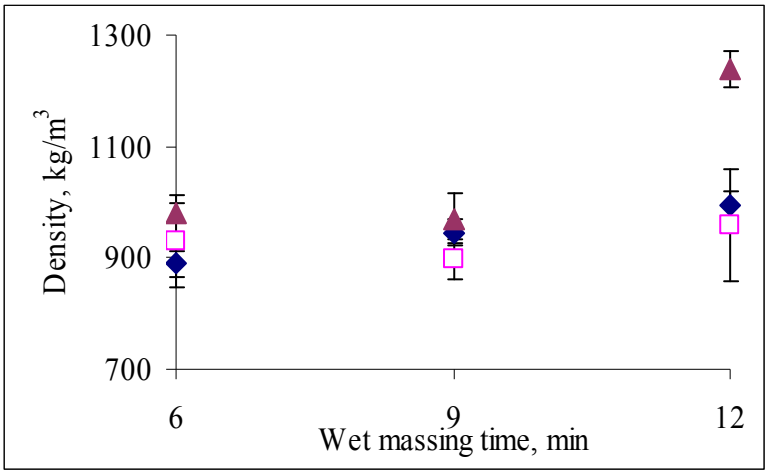


(c)

Figure 3.13: Effect of binder addition rate during granola manufacture on density at wet massing time of ◆ : 6 min; □: 9 min and ▲:12 min: (a) 150 rpm, (b) 200 rpm and (c) 300 rpm



(a)



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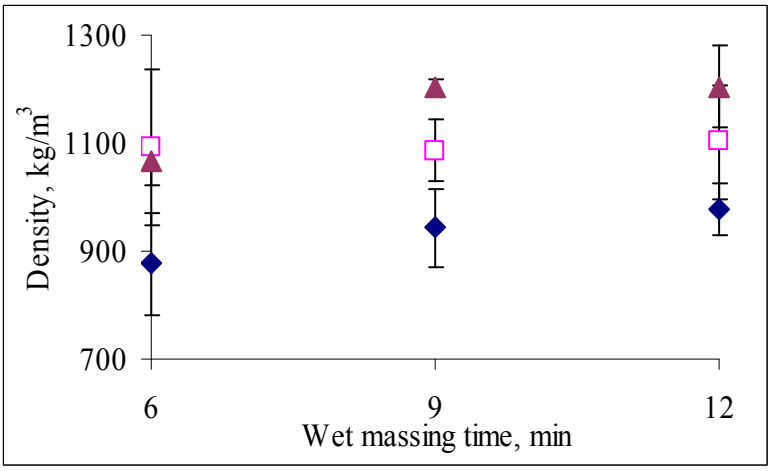


(c)

Figure 3.14: Effect of wet massing time during granola manufacture on density at impeller speed of ◆ : 150 rpm; □:200 rpm and ▲:300 rpm: (a) 0.22 g/s, (b) 0.33 g/s and (c) 0.65 g/s

The effects of wet massing time on density are shown are shown in Fig. 3.14. In general there appears to be a slight increase in density with wet massing time across all impeller speeds and binder addition rates. The effect of high impeller speeds may be due to more intimate mixing due to the high shear, which leads to an efficient distribution of the binder in the mix. Consequently, more air spaces are displaced, resulting in the formation of more dense granules (Tobyn et al., 1996). Moreover, the impeller speed and granulation time can modify granules porosity. For high impeller speed and/or long granulation time, granules are submitted to high-shear forces,

which leads to their densification, i.e. decrease of intragranular porosity and decrease of friability (Badawy et al., 2000; Kiekens et al., 1999; Oulahna et al., 2003; Tobyn et al., 1996).

Fig. 3.15 shows a Pareto chart created to investigate the significance of manufacturing process parameters on density. The Pareto analysis clearly shows that impeller speed is the most significant factor on determining granule density. Although the binder addition rate and wet massing time were also considered as significant factors, they are nowhere near as influential as the impeller speed. *P*-values calculated by the ANOVA represent the level of significance. Sensitive process parameters are those, whose *P*-values are small ($P < 0.05$). *P*-values of the impeller speed, binder addition rate and wet massing time obtained from ANOVA were 0.000001, 0.000053 and 0.030834.

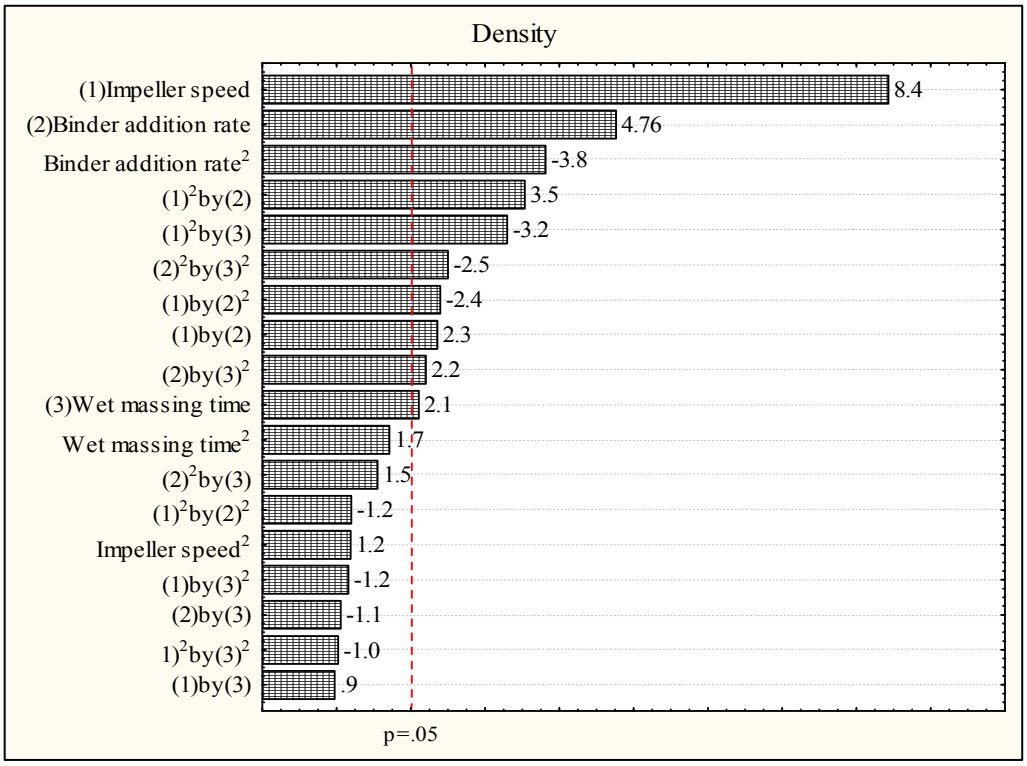


Figure 3.15: Pareto chart of standardized effects for density

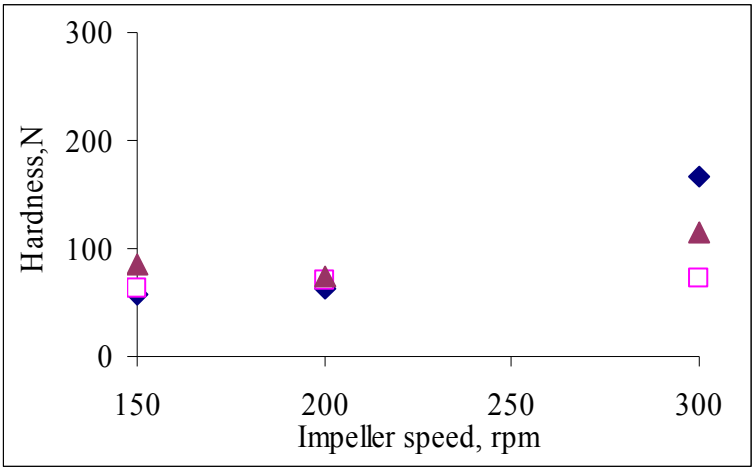
3.3.3 Effect of processing parameters on hardness

Textural parameters were also evaluated in this study. Figs. 3.16 – 3.19 shows the effect of processing parameters on hardness. The actual values for hardness ranged from 56 N to 284 N. An increase in impeller speed leads to increased granule hardness. This trend of the variation of the hardness with the impeller speed is in agreement with crushing strength measurements made by Rahmanian et al. (2008). As the higher impeller speeds cause more intensive mixing and compaction of the granules and, hence higher aggregate strength. At 300 rpm there is a significant increase in hardness relative to lower impeller speeds for all binder addition rates though in particular at lower rates (Fig. 3.16). Fig. 3.16 also shows that there is no significant variation in hardness value produced at impeller speeds of 150 and 200 rpm. However at the higher impeller speed a lower hardness is evident at the middle binder addition rate of 0.33 g/s (Fig. 3.18). Higher hardness at the lower binder addition rate and higher impeller speed would appear to be result of well mixing of the viscous binder at these extreme conditions (Fig. 3.18). Hardness also increases with wet massing time (Fig. 3.17).

It is noteworthy that density shows a correlation with hardness; the highest maximum hardness was found among granules which had been subject to higher impeller speed, and these granules also demonstrated the highest degree of density (Figs. 3.12 and 3.16). Increasing the impeller speed will increase the agitation intensity and leading to faster consolidation. As they consolidate they become stronger and less deformable. From a consumer perspective, hardness levels towards the lower end of this range would be deemed more acceptable, and consumer preference tests associated with this work showed a preference for levels below 100

N. Typical hardness value for commercially available granola ranged between 14 N to 70 N (see Appendix A).

Fig. 3.18 shows the Pareto chart resulting from the associated statistical analysis for hardness. As expected, impeller speed is the most important parameter. Wet massing time and binder addition rate are also shown to have significant effects on granule hardness with positive and negative correlations respectively though these parameters are for less important than impeller speed.



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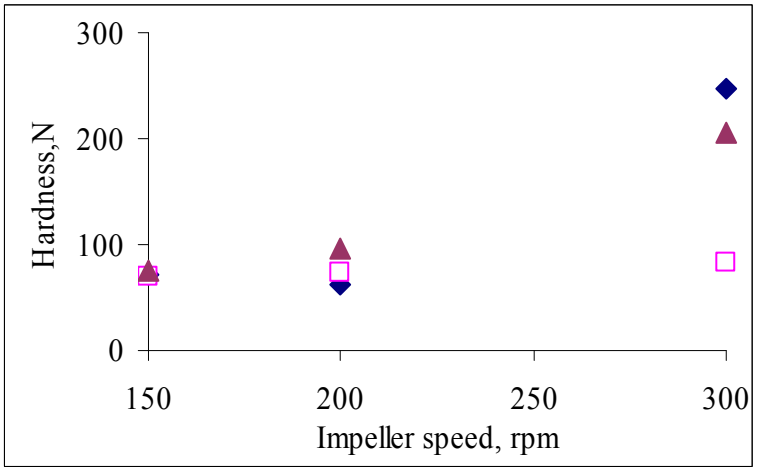

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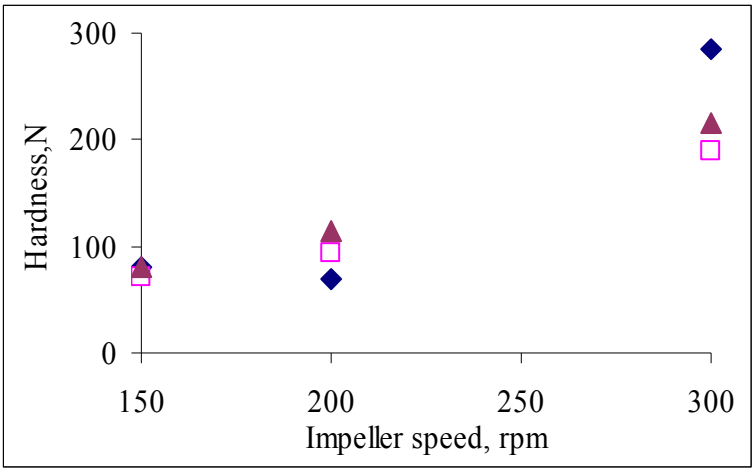
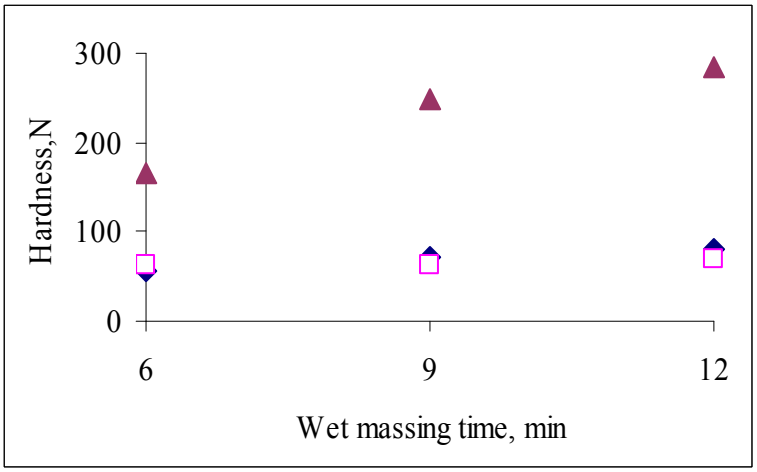


(c)

Figure 3.16: Effect of impeller speed during granola manufacture on hardness at binder addition rate of ◆: 0.22 g/s; □:0.33 g/s and ▲:0.65g/s: (a) 6 min, (b) 9 min and (c) 12 min



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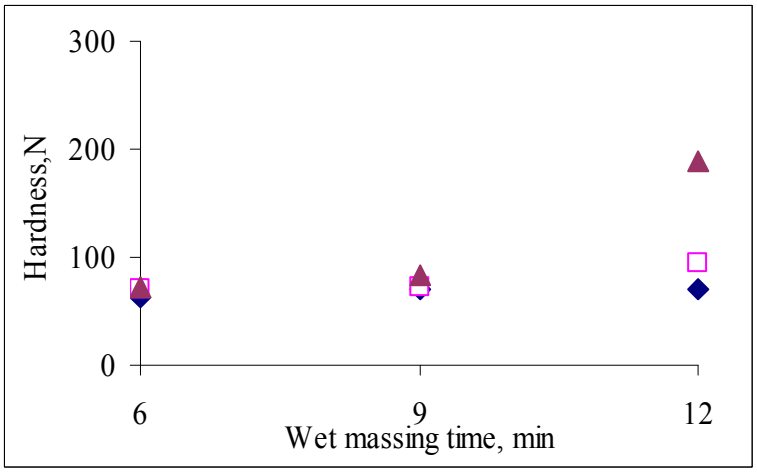

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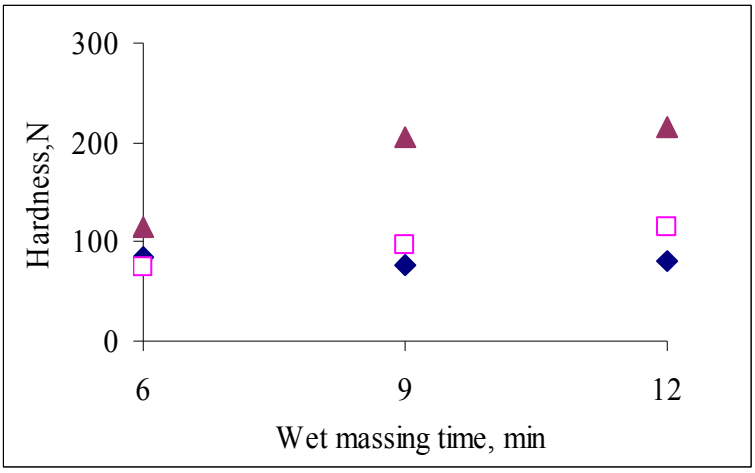
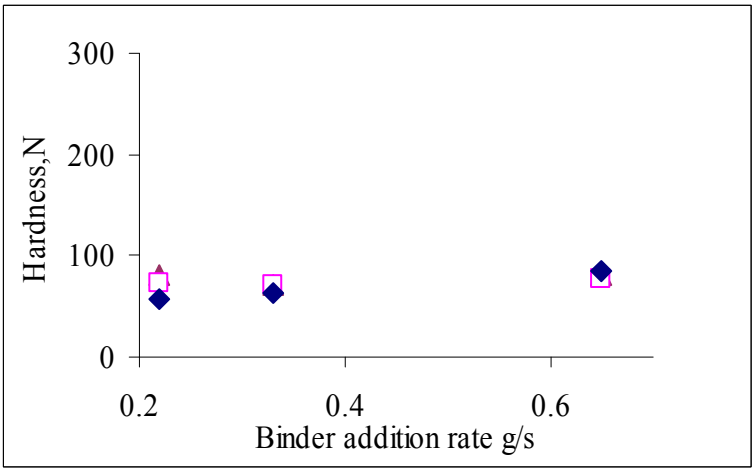


(c)

Figure 3.17: Effect of wet massing time during granola manufacture on hardness at impeller speed of ◆: 150 rpm; □:200 rpm and ▲:300 rpm: (a) 0.22 g/s, (b) 0.33 g/s and (c) 0.65 g/s



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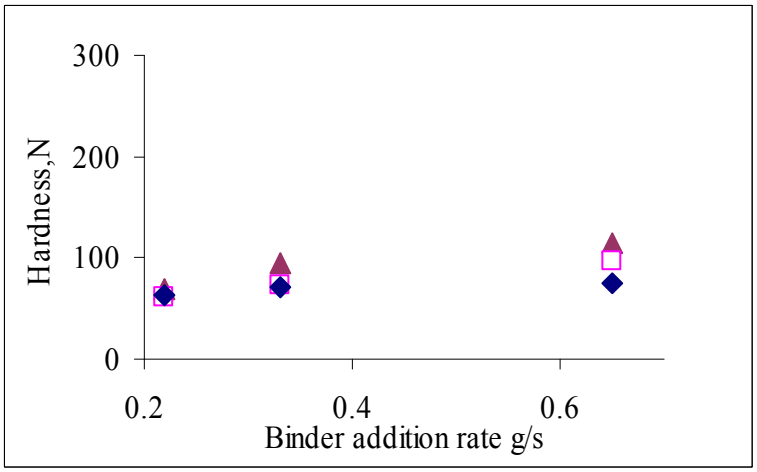

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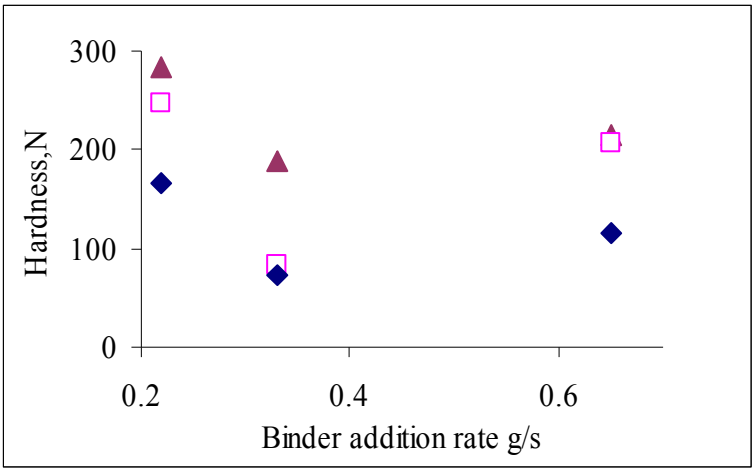


(c)

Figure 3.18: Effect of binder addition rate during granola manufacture on hardness at wet massing time of ◆: 6 min; □:9 min and ▲:12 min: (a) 150 rpm, (b) 200 rpm and (c) 300 rpm

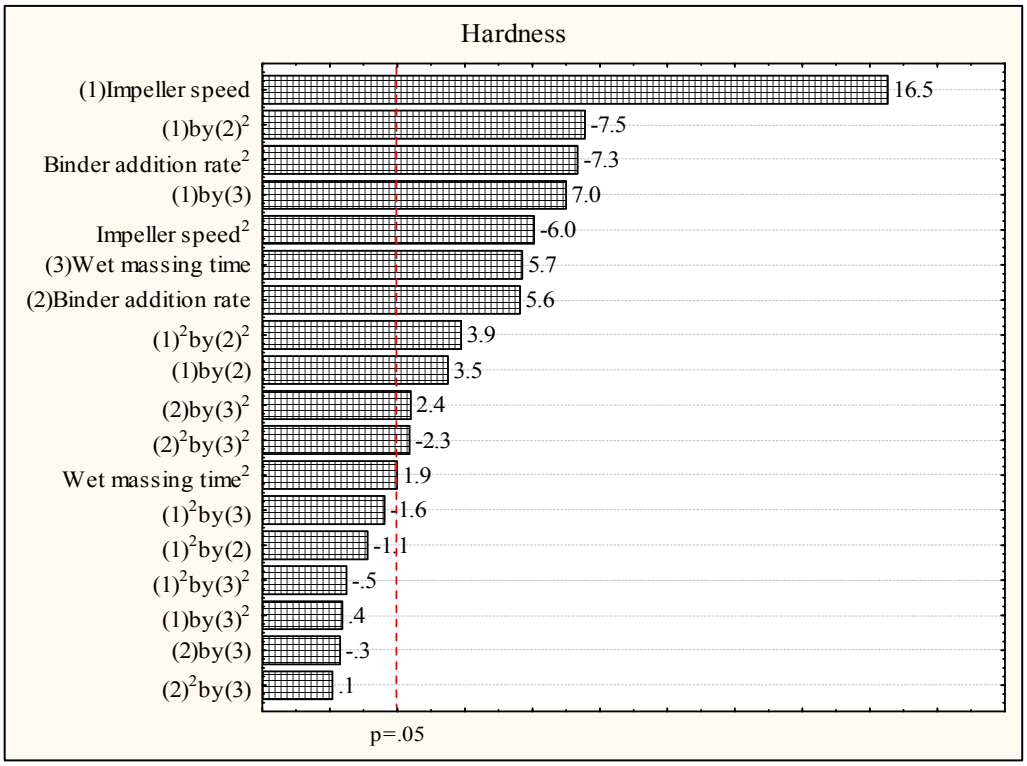
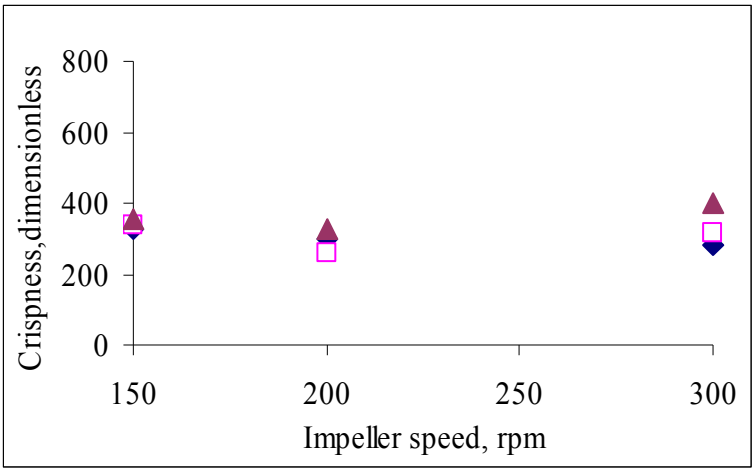


Figure 3.19: Pareto chart of standardized effects for hardness

3.3.4 Effect of processing parameters on crispness

Crispness is a desirable property of breakfast cereal granules. Crispness is associated with a rapid drop of force during mastication which, in turn, is based on fracture propagation in brittle materials (Vincent, 1998). Definitions for the "crispy" attributes include a combination of a snap clean break, a light texture and sound. Positive attributes were also associated with freshness, moistness and brittleness for some consumers (Fillion and Kilcast, 2002). The values of crispness ranged between 258 and 755. An impeller speed at 300 rpm (Fig. 3.20) appears to promote a higher value for crispness, particularly when related with a low binder addition rate. Crispness increases with speed above 200 rpm, though 150 rpm appears to show slightly higher values. Crispness values also increases with wet massing time particularly at high impeller speed (Fig. 3.21). Binder addition rate shows no significant effect for impeller speed of 150 and 200 rpm (Fig. 3.22) though aggregates formed at higher impeller speeds (300 rpm) over longer wet massing times (in particular 12 minutes, but also to a lesser extent 9 minutes) with low binder addition rates demonstrate the greatest level of crispness.



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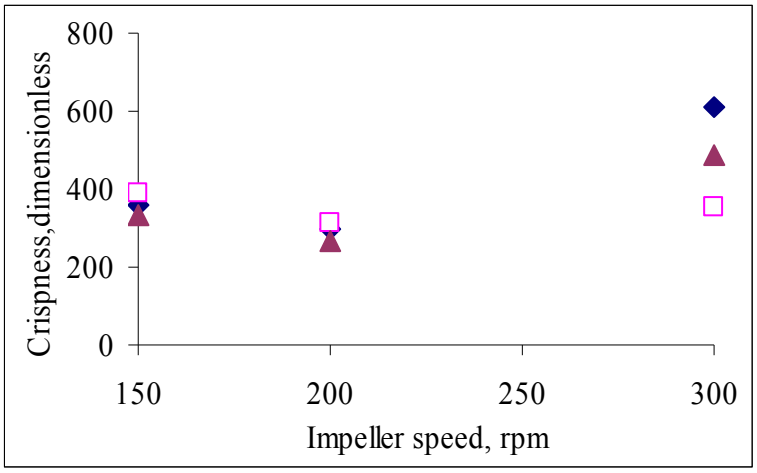

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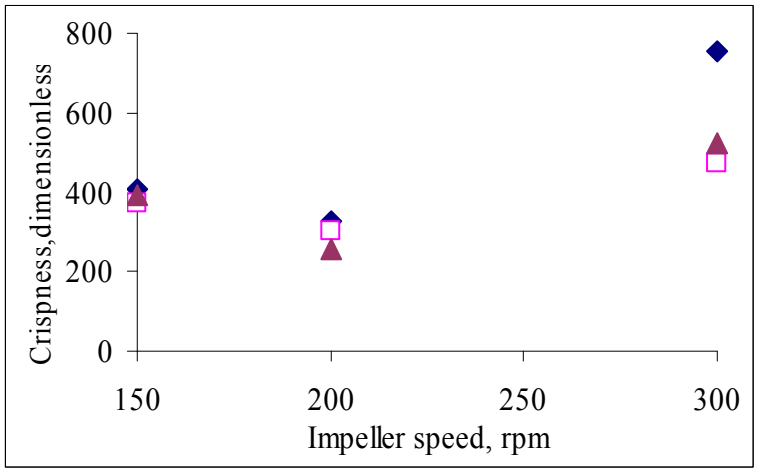
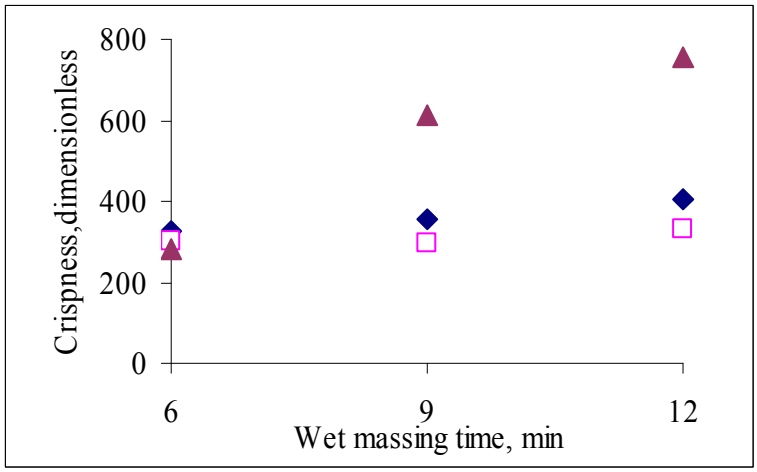


(c)

Figure 3.20: Effect of impeller speed during granola manufacture on crispness at binder addition rate of ◆: 0.22 g/s; □:0.33 g/s and ▲:0.65g/s: (a) 6 min, (b) 9 min and (c) 12 min



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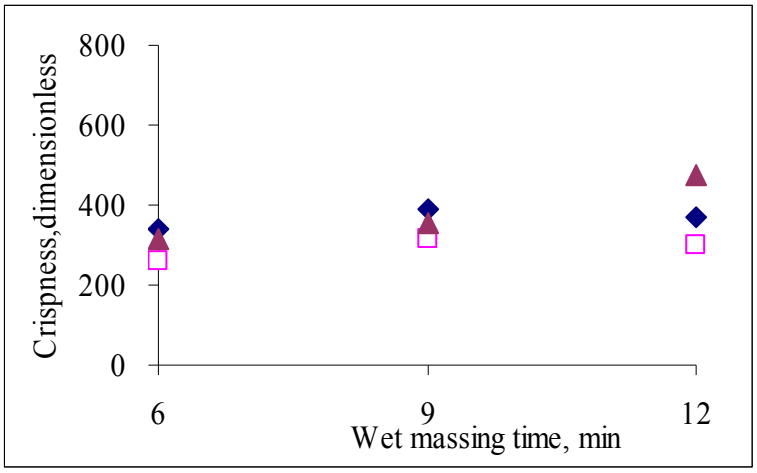

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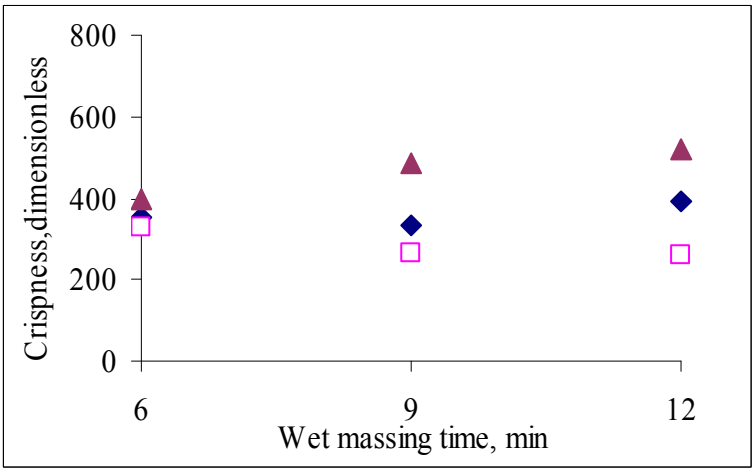
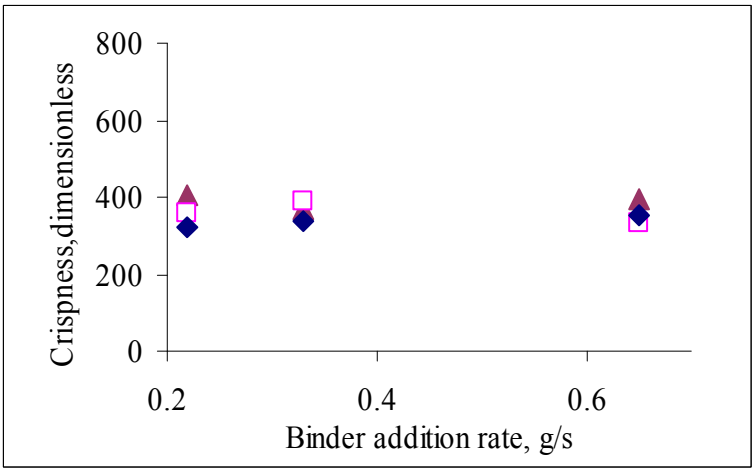


(c)

Figure 3.21: Effect of wet massing time during granola manufacture on crispness at impeller speed of ◆: 150 rpm; □:200 rpm and ▲:300 rpm: (a) 0.22 g/s, (b) 0.33 g/s and (c) 0.65 g/s



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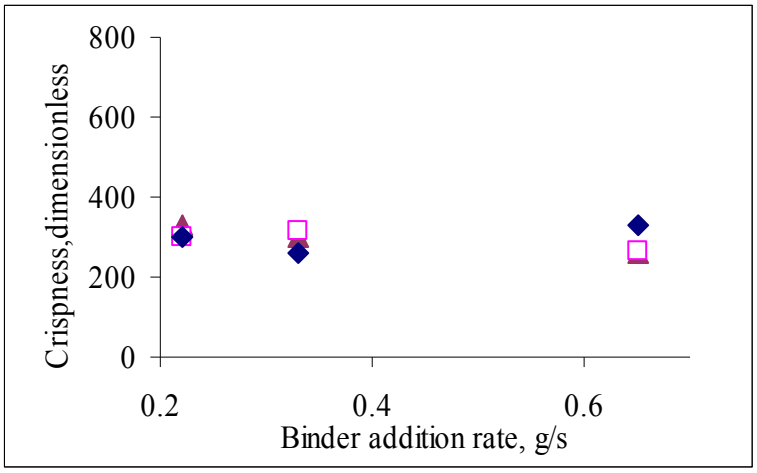

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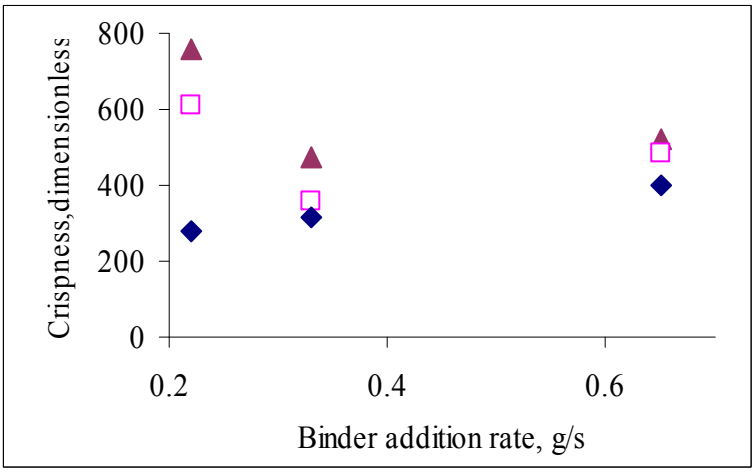


(c)

Figure 3.22: Effect of binder addition rate during granola manufacture on crispness at wet massing time of ◆: 6 min; □:9 min and ▲:12 min: (a) 150 rpm, (b) 200 rpm and (c) 300rpm

Given that crispness is associated with high level of porosity and hence low density, it is surprising that the denser aggregates formed at 300 rpm have higher levels of crispness. Perhaps at higher impeller speeds and extended wet massing times, the resultant fragmentation leads to the fine granule particles being layered onto the surface of other granules, this forming a loose structure on the surface which produces crisp granules. This appears to be borne out by visual inspection of these aggregates. By means of comparison, typical values for commercial available granola found to be in the range 68 to 572 (Appendix A).

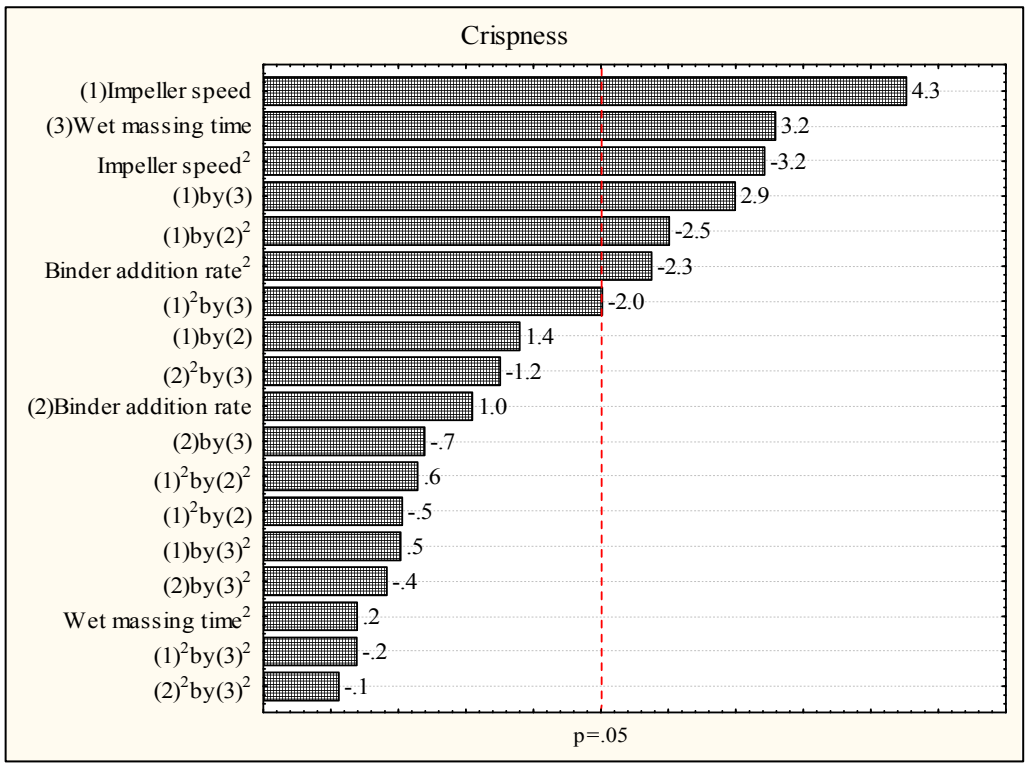


Figure 3.23: Pareto chart of standardized effects for crispness.

Fig. 3.23 shows the Pareto chart resulting from the associated statistical analysis for crispness. Impeller speed and wet massing time are shown to be the most influential factors in determining crispness from Pareto analysis. Binder addition rate is shown only to demonstrate a quadratic influence on crispness.

3.4 Conclusions

This study focused on the effect of different processing parameters on granule size and textural properties of a granola cereal product manufactured in a high shear granulator. The choice of processing parameters is critical in developing a manufacturing process that produces cereal granulation with acceptable properties for this natural product with significant inherent natural variation. The experimental results show that the impeller rotational speed is the single most important process

parameter which influences granola physical properties. After that binder addition rate and wet massing time also show significant impacts on granule properties. Increasing the impeller speed and wet massing time increases the median granule size while also presenting a positive correlation with density. The key quality parameters measured relating to texture varied significantly with impeller speed. Granola breakfast cereal produced in a high shear granulator shows higher hardness value 56 N to 284 N compared with commercially available granola which were found to be in the range 14 N to 70 N. Crispness values for commercially available granola were found to be in the range 68 to 572, whereas the granola produced in the high shear granulator ranged between 258 and 755. The combination of the highest selected impeller speed of 300 rpm and lowest binder addition rate resulted in granules with the highest levels of hardness and crispness. The insights gained from this work can aid in developing a process for the production of granola or other similar products via a high shear granulation process to match with consumer and manufacture expectations and requirements. These results when combined with the results obtained from chapter 5 on conveying can aid the study of the process will assist towards selecting suitable operational / process parameters.

Chapter 4

Effect of manufacturing process parameters on granule properties in fluidised bed granulation

4.1 Introduction

Fluidised bed granulation is a widely used granulation process whereby granules are produced in a single piece of equipment by spraying a binder solution onto a fluidised powder bed. Fluidised bed granulation is a complex process, influenced by several variables such as spray rate, inlet airflow rate, inlet temperature, inlet air humidity, nozzle air pressure and nozzle height (Rambali et al., 2001b). Agglomeration occurs in the fluidised bed due to the wetting of the particle's surface by the sprayed droplets. As a result of collisions and coalescence between the surface-wetted powder particles, liquid bridges are formed and nucleation of particles occurs leading to the growth of granules. The growth mechanism involved depends on the operating parameters: fluidizing air temperature and flow rate; nature, concentration and feed rate of the sprayed solution; spraying system and settings; and primary particles properties (Heinrich and Mörl, 1999; Jiménez et al., 2006).

The binder droplet distribution onto the fluidized bed is of great importance in terms of aggregate growth rate, as the droplet size influences the agglomerate size directly (Ax et al., 2008; Schaafsma et al., 2000). The droplet size depends mainly on the spray rate and the nozzle air pressure (Rambali et al., 2001a). Wan et al. (1995b) investigated the effect of atomizing air pressure on spheroid size and found that the size of the droplets sprayed does not seem to have a significant effect on spheroid

size. Aulton and Banks (1981) reported that atomizing air pressure, nozzle type and position of nozzle all influence the physical properties of granules. They found that using a higher feed rate of liquid binder results in a larger average granule size.

The spray rate, a critical parameter to the formulation, affects granule size, bed moisture and product quality (Hu et al., 2008). Many researchers (Davies and Gloor, 1971; Rankell et al., 1964; Schaefer and Wørts, 1978a) found that the mean granule size increases with increased binder spray rate. However, Hemati et al. (2003) reported that the spray rate had no effect on ultimate granule size when it was lower than some critical value. AbuBaker et al. (2003) proposed that the binder delivery rate/atomization air pressure ratio was the most important factor that controlled the granule growth rate and properties. Jiménez et al. (2006) found that the binder spray rate and nozzle air pressure influences the angle of the sprayed jet and the droplet size.

The objective of this study was to determine the effect of binder spray rate and nozzle air pressure on the size (geometric mean diameter) and texture properties of granola breakfast cereal prepared by fluidized bed granulation. It was not feasible to vary fluidizing air temperature or flow rate on the unit used so these setting were set as constant values throughout.

4.2 Materials and methods

Granule aggregates were produced in the fluidised bed granulator (Mini-Airpro, ProcepT, Belgium). The fluidised bed granulator has a glass container with the lower portion being conical in shape and a cylindrical upper portion. The entire container is 0.73m in height while the upper cylindrical portion has an inner diameter of 0.2m

and is 0.28m in height. A schematic drawing of the experimental set up is shown in Fig. 4.1. Spray from the nozzle is in a downward direction, counter current to the fluidising air flow. The granules are produced in the process column by spraying the binder solution from the top in counter current air flow and under pressure conditions. A pulse back system is used for cleaning filters. The fluidising air was first preheated by an electrical heater (80°C) for 10 minutes to achieve steady state inlet temperature conditions of 40°C and its flow rate is subsequently measured before entering the bed. The air used to fluidise the product bed is normally heated so that the heat transfer process is very rapid. Hot inlet air is supplied during the process as heat is transferred to the particles in order to help evaporate the liquid and mass is transferred as vapour into the moving air. The monitoring of the bed temperature was achieved by controlling the inlet fluidising air temperature and regular monitoring of the outlet air temperature.

The ingredients were initially placed in the unit and were mixed by using air of 40°C was employed at a flow rate at 0.95 m^{3}/min (the maximum operating limit for the equipment for this product). Product composition is presented in Table 4.1. The granola ingredient composition is not same as that used in high shear granulation (Table 3.1). In the fluidised bed granulation process a lower liquid binder content is used as otherwise wet heavy agglomerates ensue which cannot be lifted adequately to permit the formation of a fluidised bed for the operating parameters and unit employed in the present study. Moreover, due the extremely hygroscopic nature of inulin, which causes it to becomes very hard in the presence of the blowing inlet air, it is not used as an ingredient in fluidised bed granulation process. A honey-water mixture (85:15) used as binder. The binder solution was then sprayed on the fluidizing powder bed using a peristaltic pump (to adjust the spray rate). The

granulating liquid is drawn up by peristaltic pump from a reservoir, set up on balance, to the spray nozzle. The binder flow rate is controlled by the pump revolution while the reservoir weight is continuously monitored. Three peristaltic pump speeds of 5, 10 and 15 rpm were used resulting in binder flow rates of 0.4, 0.8 and 1.2 g/min respectively. This corresponded with binder addition times of 46 min, 23 min and 15 min respectively. Nozzle air pressure of 2, 3 and 4 bar was used. Each experiment is conducted in triplicate. Spraying continued until all the binder solution was used. Afterwards 0.5 litres of water was sprayed in order to rinse the tubes. The granola was then taken from the unit and dried in on an oven at 160˚C for 10 minutes.

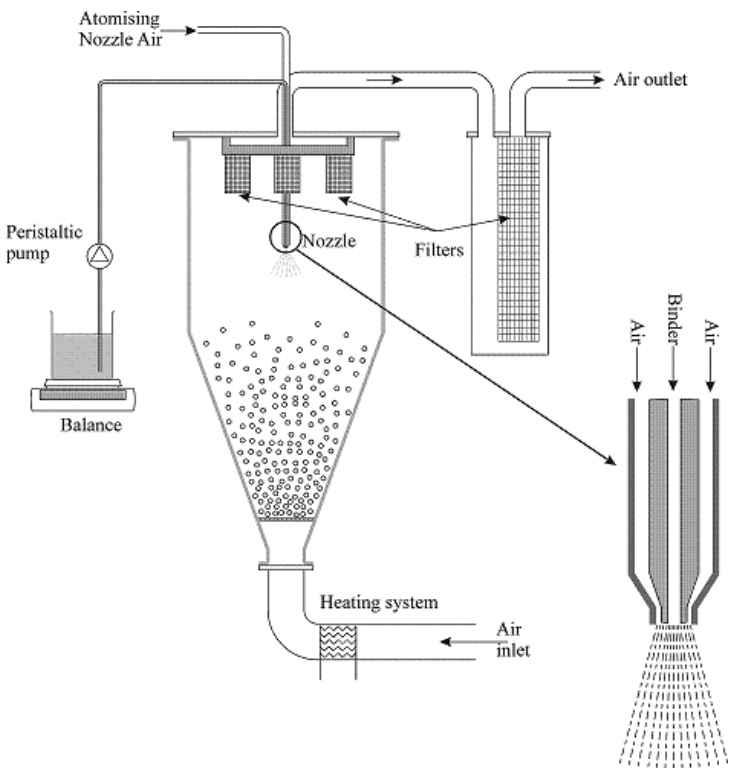


Figure 4.1: Schematic sketch of fluidised bed granulator (Mini-Airpro, ProcepT, Belgium)

Table 4.1: Granola ingredients in fluidised bed unit

Ingredients	Percentage
Jumbo oat flakes	33.4
Corn flakes	5.2
Puffed Rice	5.2
Malted buckwheat (milled)	4.0
Malted barley (milled)	4.0
Brown sugar	8.8
Honey	15.6
Water	2.8
Oat beta glucan	17.4
Wheat germ	3.6

A Camsizer (Retsch, Germany) digital image analyzer was used for measuring particle size distribution and density (Heinrich et al., 2009). Particle size distributions were characterized by d_{10} (tenth percentile), d_{50} (median size) and d_{90} (ninetieth percentile). Granule mass was measured by using a digital balance (Sartorious ED4202S, Germany) with sensitivity of 0.01 g. Textural properties measurement and data analysis are explained in chapter 3.

4.3 Results and discussion

4.3.1 Effects of processing parameters on aggregate size

The process variables investigated were binder spray rate and nozzle air pressure. These process parameters are interdependent and a greater understanding of the

relationship between parameters can result in the production of a more desirable granola product. Typical granule size distribution is shown in Fig. 4.2 for the applied nozzle air pressures (2, 3 and 4 bar). Nozzle air pressure at 2 bar resulted in slightly coarser granules compared with other nozzle pressure (Fig. 4.2). Table 4.2 shows the distribution span for different processing parameters. The distribution span measures the width of the particle size distribution (Korhonen et al., 2002). A small span value indicates a narrow size distribution. In general, it appears that combination of low nozzle air pressure and low binder spray rate results in the lowest distribution span and thus the tightest particle size distribution.

Table 4.2: Effect of nozzle air pressure and binder spray rate on particle size distribution span and granule density.

Nozzle air pressure, bar	**Binder spray rate, g/min**	**Distribution span,** $\frac{d_{90} - d_{10}}{d_{50}}$	**Granule density, kg/m^3**
2	0.4	1.3	655 ± 57
2	0.8	1.4	555 ± 52
2	1.2	1.5	544 ± 36
3	0.4	1.6	614 ± 47
3	0.8	1.7	586 ± 13
3	1.2	1.6	588 ± 30
4	0.4	1.5	531 ± 58
4	0.8	1.5	566 ± 82
4	1.2	1.8	558 ± 78

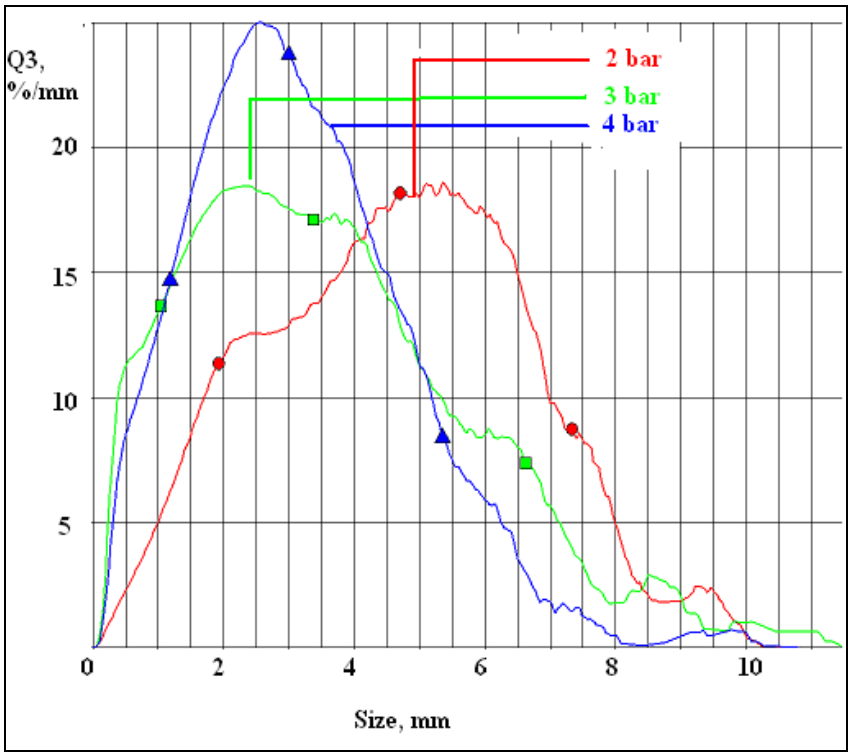


Figure 4.2: Granule size distribution at different nozzle air pressure at binder addition rate 0.4 g/min

Granule median size, d_{50} values of the granola breakfast cereal ranged 3.2 – 4.1 mm. Fig. 4.3 shows the effect of binder spray rate and nozzle air pressure on median granule size, (d_{50}). A nozzle air pressure 2 bar produces slightly larger median aggregate size compared with 3 and 4 bar nozzle air pressure for all binder spray rates. The higher the spraying air pressure, the smaller the jet angle and the liquid droplets diameter and the higher the droplets speed (Jiménez et al., 2006). Granule size does not show much variation at nozzle air pressure 3 to 4 bar. A combination of lower binder spray rate and nozzle air pressure shows higher d_{50} values as well as tighter distribution (see Table 4.2). However at binder spray rate 0.8 g/min, there appears to be no significant variation in d_{50} values at various nozzle air pressures. At other binder spray rates granule size decreases as nozzle air pressure increase. This study appears to confirm that the nozzle air pressure clearly affects granule size, in

accordance with findings by other authors (Bouffard et al., 2005; Jiménez et al., 2006). This study found, in line with Bouffard et al. (2005) that an increase in atomizing pressure lowers the droplet size, reducing the droplet penetration of the bed which leads to lower moisture content of the bed and a smaller granule size.

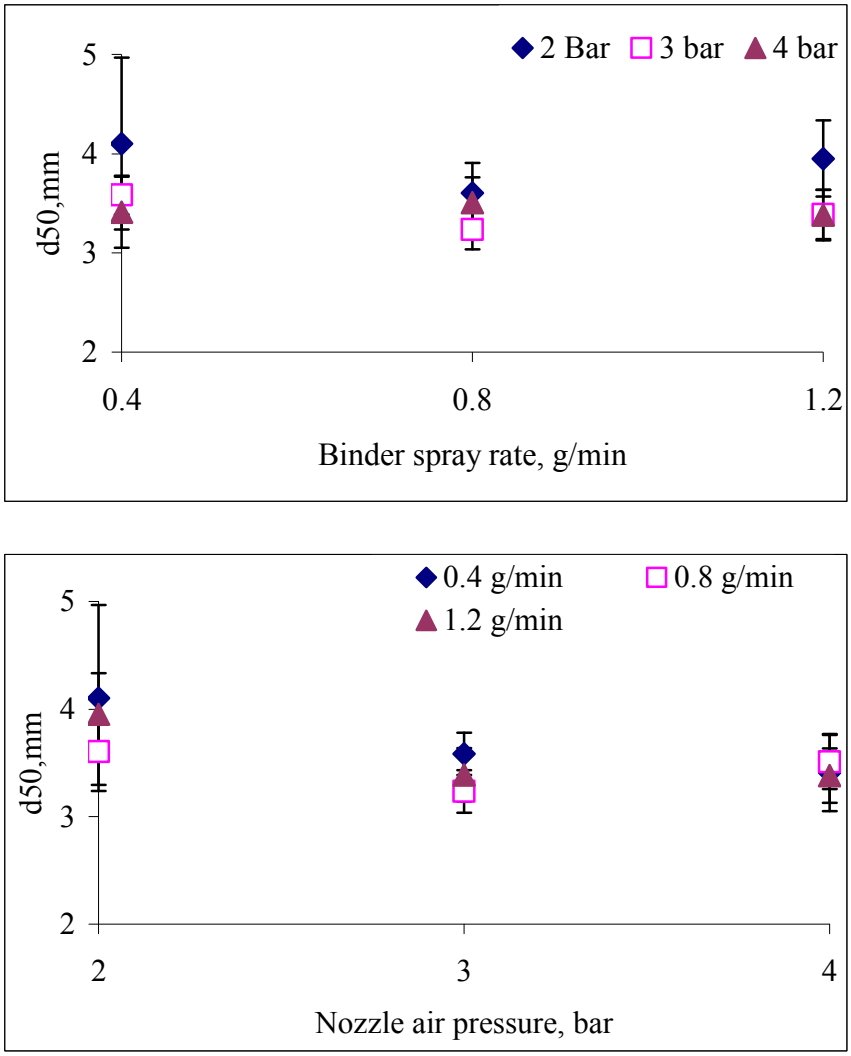


Figure 4.3: Effect of binder spray rate and nozzle pressure during granola manufacture on d_{50}

Statistical analysis was carried out to determine the most significant parameters, and from this a Pareto chart was constructed (Fig. 4.4). The effect of nozzle air pressure on granule size was statistically significant ($P < 0.05$). As seen in Fig. 4.4 binder

spray rate is shown to have no significant effect on d_{50} values. The increasing atomizing pressure lead to a decrease in the droplet size, resulting in smaller granule sizes as a result of a large reduction of droplet-particle collisions (Lin and Peck, 1995). The effect of liquid flow rate might be due to a change in droplet size (Iveson et al., 2001). This finding is in agreement with that by Hemati et al (2003) who studied the influence of liquid flow rate and atomising pressure on droplet size. It was observed that increased atomising air pressure produces a smaller droplet size, while there is negligible effect of binder spray rate on the droplet size for the operating regimes reported. Their experimental results revealed no significant influence on the growth rate at increased binder spray rate and this was attributed to the negligible differences in the mean droplet size. The overall final granule size is observed to increase with lower nozzle air pressure. In fluidised bed granulation, particle growth follows a nucleative process. Particle ingredients, when wetted, form nuclei that are held together by liquid bonds. The formation of these nuclei is influenced by the size of the droplets sprayed. Larger droplets form larger nuclei because they are able to bind more particles. The size of the droplets sprayed can be changed by varying the atomizing air pressure. An increase of the atomizing air pressure decreases the size of the droplets and consequently the size of the granules produced (Merkku and Yliruusi, 1993). The atomizing pressure also increases the air shear forces within the granulation chamber adding to the breakage of formed granules, which contributes to size reduction of the granules (Bouffard et al., 2005).

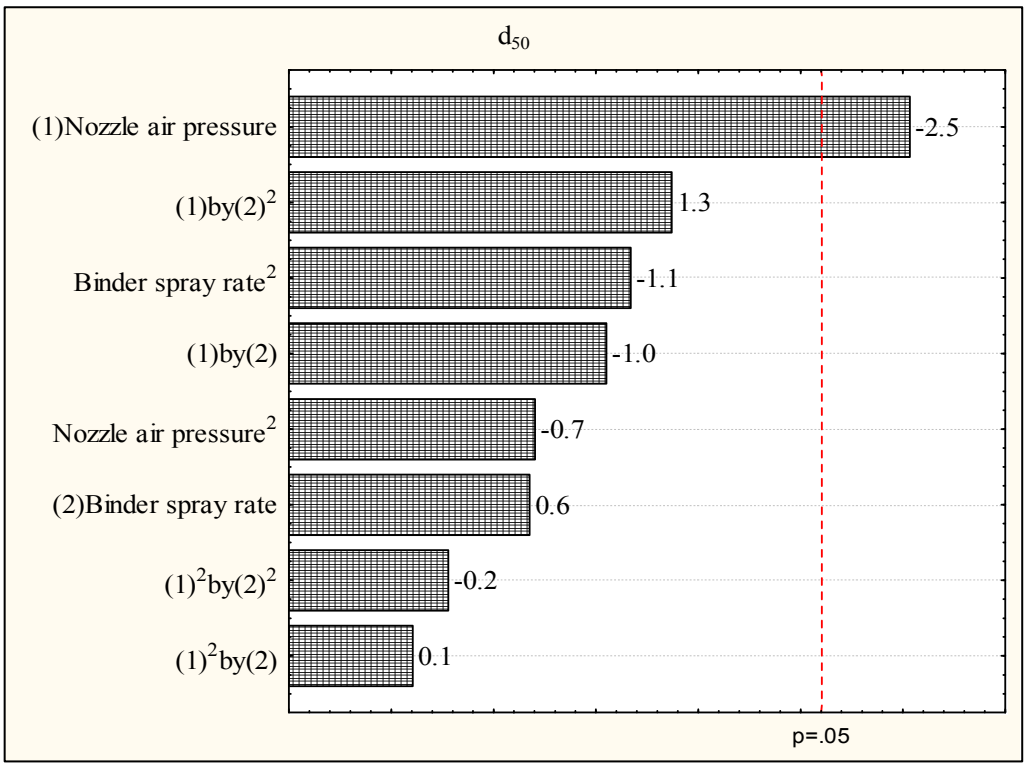


Figure 4.4: Pareto chart of standardized effects for d_{50}

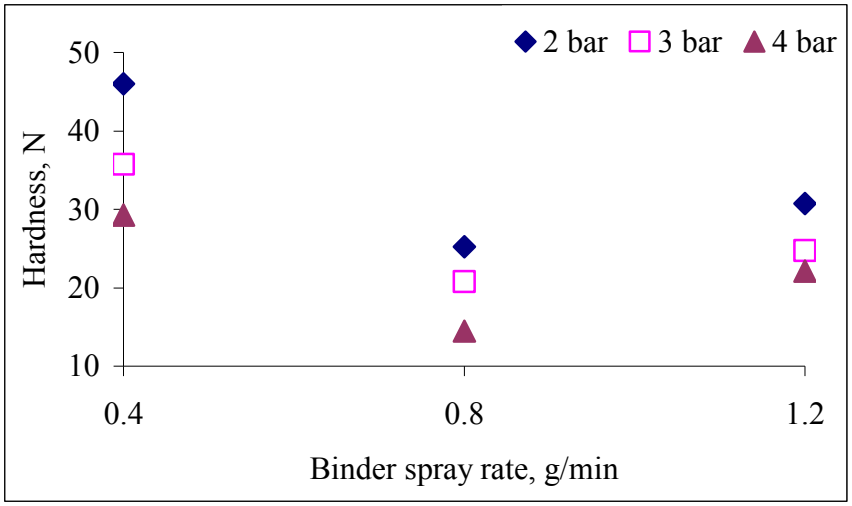
4.3.2 Effect of processing parameters on granule density

The effect of the processing parameters binder spray rate and nozzle air pressure on granule density is shown in Table 4.2. Average product density ranges from 531 to 655 kg/m^3. The highest density value was found at a combination of lower nozzle air pressure and lower binder spray rate. It is noteworthy that higher density shows a correlation with granule size; the highest maximum density was found among granules which had been subject to lower nozzle air pressure and lower binder spray rate, and these granules also demonstrated the highest d_{50} value (Fig. 4.3 and Table 4.2). Davies and Gloor (1971) also reported that lower nozzle pressure increases the granule size and resulted in higher density. At binder rate of 0.4 g/min, granule density decreased significantly as nozzle air pressure increased. At binder rate 0.8

g/min and 1.2 g/min, there was no significant difference found on granule density with respective nozzle air pressure.

4.3.3 Effect of processing parameter on hardness

Textural parameters were also evaluated in this study. Fig. 4.5 shows the effect of processing parameters on hardness. A binder spray rate of 0.4 g/min displays the highest values of hardness while the binder spray rate of 0.8 g/min shows the lowest hardness value for all nozzle air pressures. Moreover granules formed at the lowest nozzle pressure of 2 bar show highest hardness values. This corresponds with aggregates displaying the highest density and size (see Figs. 4.3, 4.5 and Table 4.2). Menon et al. (2002) also reported that the atomization pressure affects the granule particle size distribution, which influences its hardness. Granule hardness decreases as nozzle air pressure increases. A combination of lower nozzle pressure and lower binder spray rate produces the highest values of hardness. The actual values for hardness ranged from 14 N to 46 N. This range would appear to be deemed acceptable, as consumer preference tests associated with this work showed a preference for levels below 100 N.



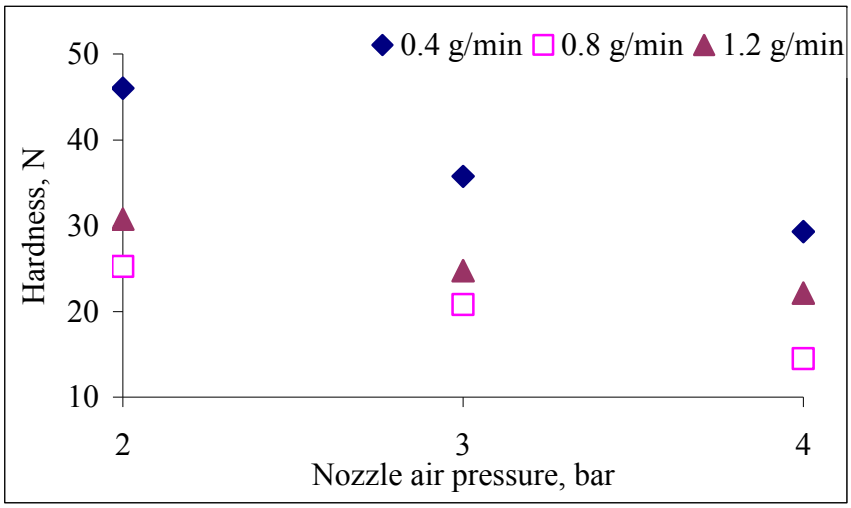


Figure 4.5: Effect of binder spray rate and nozzle pressure during granola manufacture on hardness

A corresponding Pareto chart shows that nozzle air pressure had a significant effect on granule hardness (Fig. 4.6). The nozzle air pressure decrease and subsequent increase in average granule size resulted in a less friable granulation (Davies and Gloor, 1971). Binder spray rate is shown to have no significant effect on hardness values. This finding is an agreement with that by Menon et al. (2002) who increased operating parameter for pilot trials to increase product output and found that granule hardness was not affected by binder spray rate.

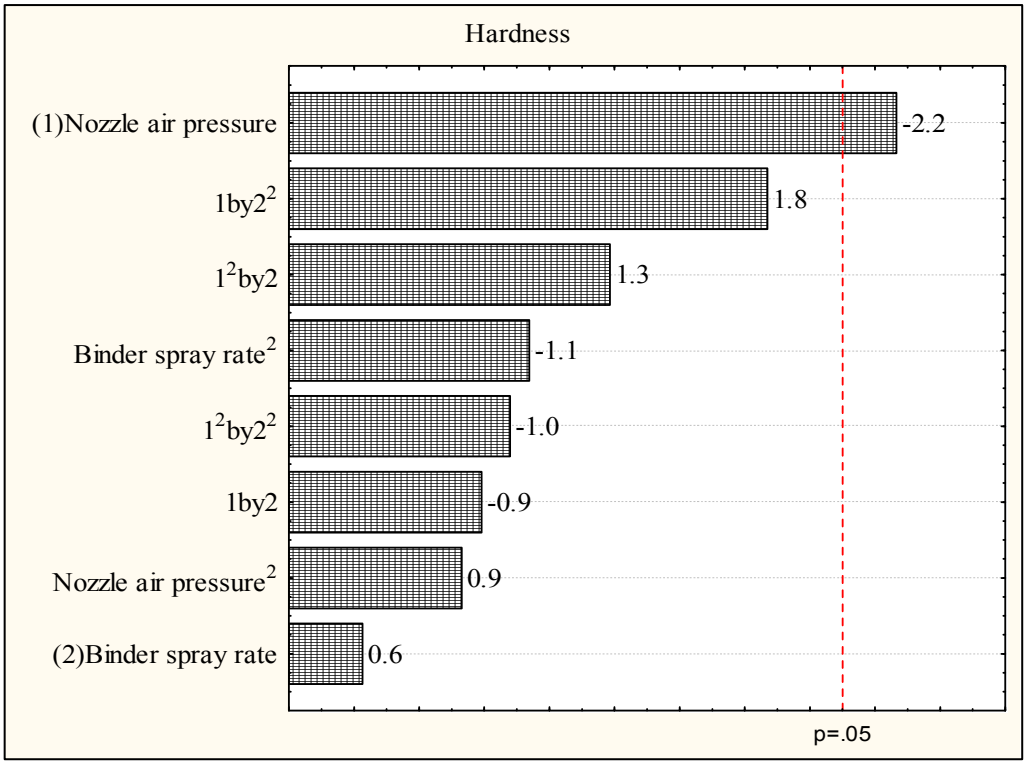
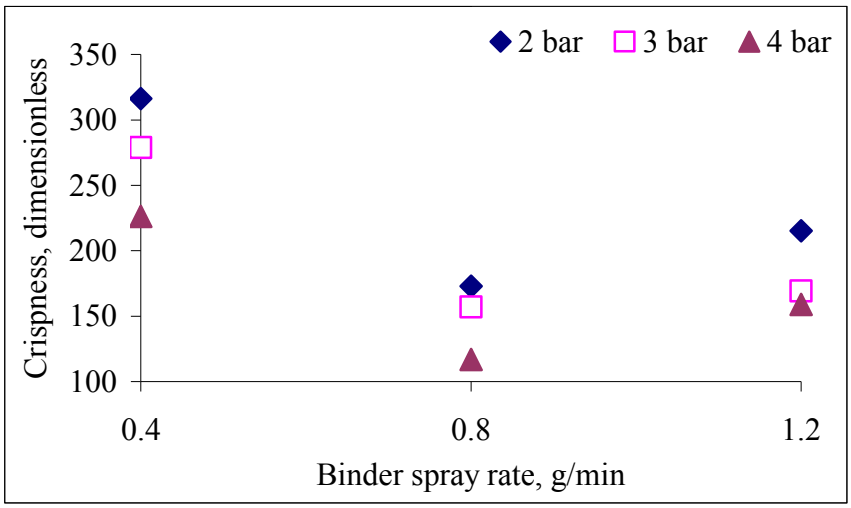


Figure 4.6: Pareto chart of standardized effects for hardness

4.3.4 Effect of processing parameter on crispness

Crispness is a desirable textural attribute of low moisture snack foods (Arimi et al., 2010; Roudaut et al., 2002) such as breakfast cereals (Sauvageot and Blond, 1991). Crispness is perceived through a combination of the noise produced and the force required to break down a product during biting (Duizer et al., 1998). The values of crispness ranged between 117 and 316. Fig. 4.7 shows that higher crispness values were associated with the lowest binder addition rate of 0.4 g/min while crispness reduces at the binder addition rate of 0.8 g/min. The value of crispness also decreased as nozzle air pressure increases. Nozzle pressure 2 bar shows the highest crispness values at all binder addition rates. As with hardness lower nozzle air pressure and lower binder spray rate resulted in higher crispness values. A nozzle air pressure 4 bar and binder spray rate 0.8 g/min produces the lower crispness value.

These findings are consistent with the hardness values (Figs. 4.5 and 4.7) while Vickers (1988) shows the inverse relationship between crispness and force deformation parameter. A Pareto chart of effects is shown in Fig. 4.8. According to this, neither nozzle air pressure nor binder spray rate show any significant effect on granule crispness.



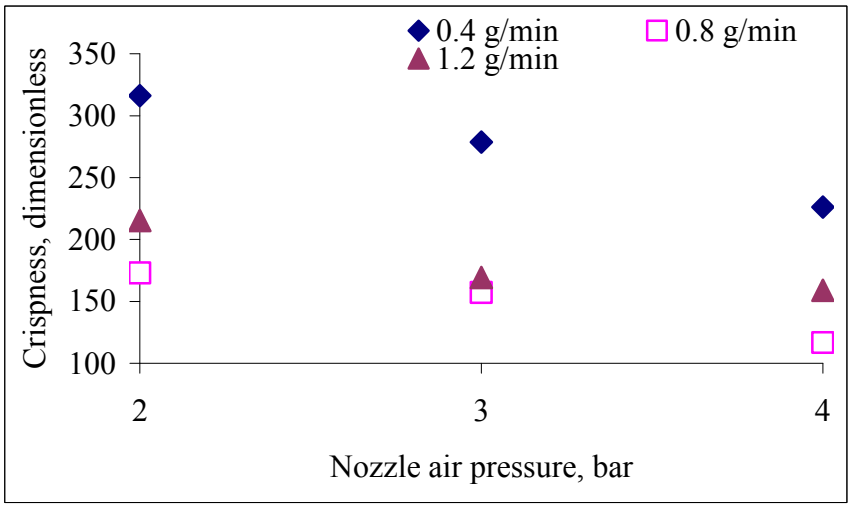


Figure 4.7: Effect of binder spray rate and nozzle air pressure during granola manufacture on crispness

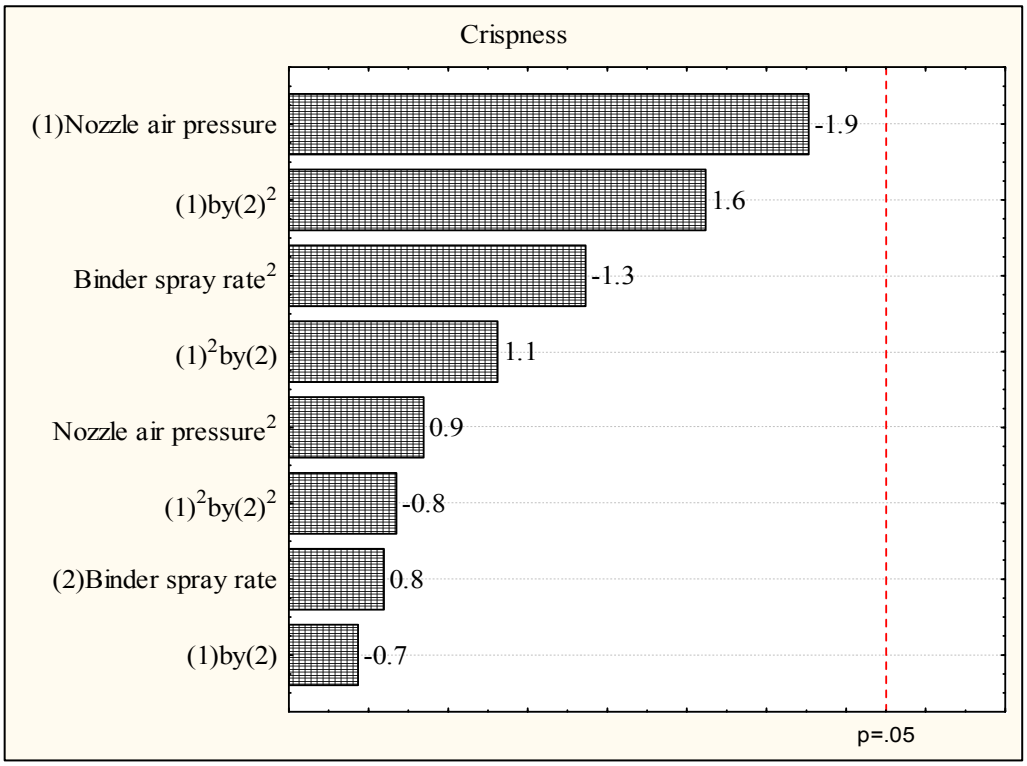


Figure 4.8: Pareto chart of standardized effects for crispness

4.4 Conclusions

The effects of the manufacturing process parameters on granola breakfast cereal properties in fluidised bed granulator are investigated. The experimental results show that the nozzle air pressure has a great importance on granule properties. In general a decrease in nozzle air pressure leads to larger in mean granule size. Other granule properties are a function of size. However nozzle air pressures had significant effect on granule hardness. Both hardness values and crispness values of the granola breakfast cereal produced in fluidised bed granulator shows good agreement with commercially available granola (see Appendix A). The combination of lowest nozzle air pressure and lowest binder spray rate results in granules with the highest levels of hardness and crispness. Further studies on effects of processing conditions for the production of granola breakfast cereal or other similar food products may be

considered an opportunity for applied research. These results when combined with the results obtained from chapter 5 on conveying can aid the study of the process will assist towards selecting suitable operational / process parameters.

Chapter 5

Effect of manufacturing process parameters during granulation on attrition during pneumatic conveying of granola

5.1 Introduction

Pneumatic conveying systems are used in food processing and other process industries to transport granular materials. The breakage of particles in many pneumatic conveying systems can represent a major problem for particulate products. In some cases the size distribution and appearance of the particles may change so significantly due to conveying that the product no longer meets required specifications. During pneumatic conveying erosion of particle surfaces can result in a high level of fines and reduce the functionality of coated particles. Furthermore, particle breakage may influence the performance of transport and processing operations (Byrne et al., 2002).

Pneumatic conveying systems are a key unit operation in the transportation of particulate material. Attrition is caused mainly by impact or shear loads, which can occur during the transport of materials. Conveying velocity is one of the main parameters influencing the product attrition (Kalman, 1999; Klinzing et al., 1997; Klinzing et al., 2001). Other important parameters are the loading ratio, or the mass flow of product relative to the mass flow of air (Kalman, 1999) and bend design (Chen and Soo, 1982; Kalman, 2000; Salman et al., 2002). Particles can break in a single collision if the impact load is higher than their strength. The collision velocity, the angle of collision and the elasticity of the collision significantly affect the impact load. The particles during pneumatic conveying experience extensive impact loads

mainly at the bends since the flow is changing direction. Particle breakage of aggregated granola can occur during conveying as product is transferred as part of the production process on its way to packaging. Such breakage occurs as a result of particle-particle and particle-wall collisions with the conveying equipment (Bas et al., 2010).

Flexible (rubber) long radius bends have been shown to result in much lower attrition compared to rigid long radius bends made of steel (Kalman and Goder, 1998). It has been shown that the vibration of bends affects the attrition of granules (Kalman, 2000), and that long bends are not always useful to reduce such attrition (Aked et al., 1997). From the engineering point of view, the bends in a pneumatic conveying pipeline are one of the major critical devices. They contribute a major part of the pressure drop and hence energy consumption for the conveying system. They can also cause great damage to the particles which is sometimes desirable but usually undesirable. They tend to wear and can cause blockage to the flow due to product buildup (Kalman, 2000).

The attrition of particles or granulated materials during pneumatic transport is an issue of considerable industrial concern. The objective of the present research was to study the effect of manufacturing and conveying parameters on product attrition of granola breakfast cereal produced by high shear granulation and fluidised bed granulation processes.

5.2 Materials and methods

5.2.1 Granules produced by high shear granulation process

The aggregation of the granola ingredients (Chapter 3, Table 3.1) took place a high shear granulator (Procept, 4M8, Belgium) subject to impeller agitation at 150 rpm, 200 rpm and 300rpm for 6, 9 and 12 minutes with binder flow rates of 0.22 g/sec, 0.33 g/sec and 0.65 g/sec. The aggregates were then baked in an oven for 10 minutes at 160°C.

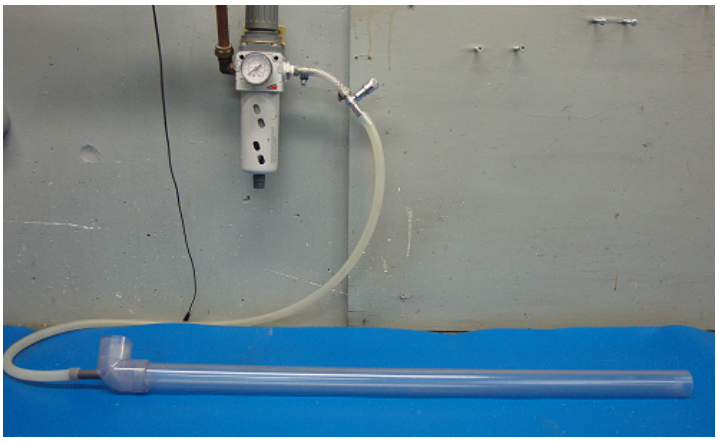
5.2.2 Granules produced by fluidised bed granulation process

Granola was produced in a fluidised bed granulator (Mini-Airpro, ProcepT, Belgium) as shown in Fig. 4.1 (Chapter 4). A granola ingredient is displayed in Table 4.1 (Chapter 4). The detail granola manufacturing process described in section 4.2 (Chapter 4). Three peristaltic pump speeds of 5, 10 and 15 rpm used which resulting binder flow rate of 0.4, 0.8 and 1.2 g/min respectively. Nozzle air pressure of 2, 3 and 4 bar was used. The granola was then baked on an oven at 160˚C for 10 minutes.

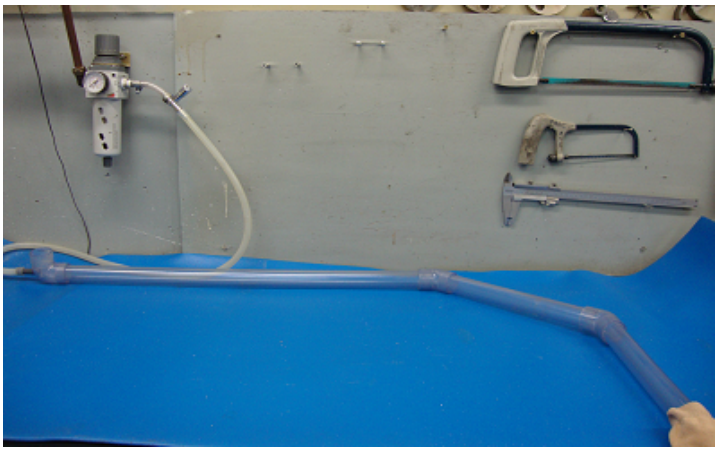
5.2.3 Pneumatic conveying of granola product

The pneumatic conveying rig comprised a horizontal pipe of internal diameter 25 mm with different bend configurations (straight pipe, two 45° bend and 90° bend). Fig. 5.1 shows a photograph of the pneumatic conveying rig used in the present study. The outer diameter of the large Perspex pipeline is 33.5 mm, giving a wall thickness of 4.25 mm. All sections of pipeline are made of transparent PMMA (Perspex), so that granola breakage and flow behaviour can be observed. The particle

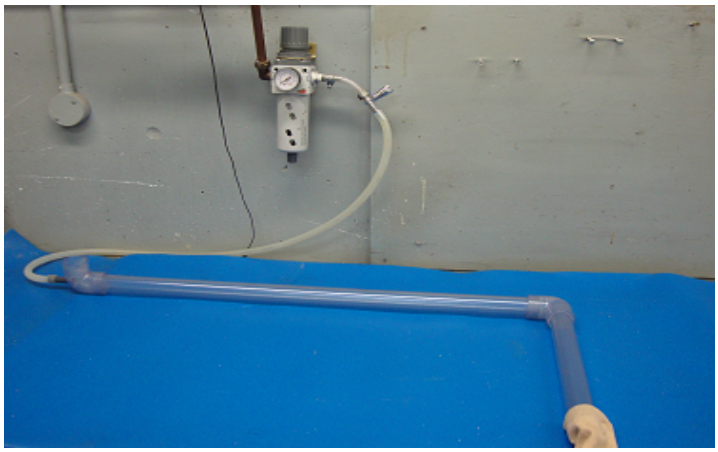
size distribution of the granola was measured after passage through the each rig configuration at each air pressure after various numbers of cycles through the rig.



(a) A straight pipe rig



(b) A rig comprising two 45° bends



(c) A rig comprising one 90° bend

Figure 5.1: Photograph of the pneumatic conveying rigs

A straight pipe was used as the first conveying rig. The straight pipeline has 1.3 m length in horizontal axis with 25 mm internal diameter (Fig. 5.2). Air is supplied by the compressor and regulated to deliver the pressures of 2 bar, 3 bar and 4 bar. The granola is fed into the apparatus by hand at the location indicated on the diagram. A soft, fine-mesh piece of cloth is used to capture the granola at the end of the rig for measurement of its particle size distribution.

A rig comprising two 45° bends, comprised an initial section 0.70 m long and two short sections of 0.30 m length with the same internal diameter of 25mm (Fig. 5.3). The granola is fed into the apparatus by hand at the location indicated on the diagram. A soft, fine-mesh piece of fabric is used to capture the granola at the end of the rig for measurement of its particle size distribution.

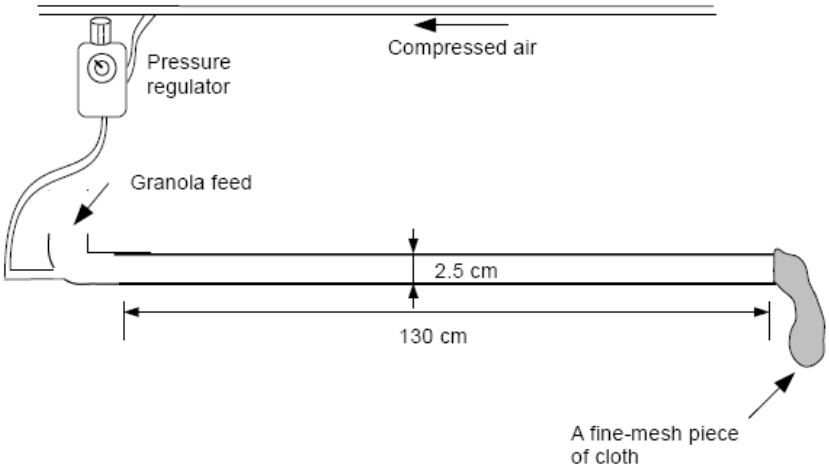


Figure 5.2: A schematic diagram of the straight pipeline used for pneumatic conveying of granola

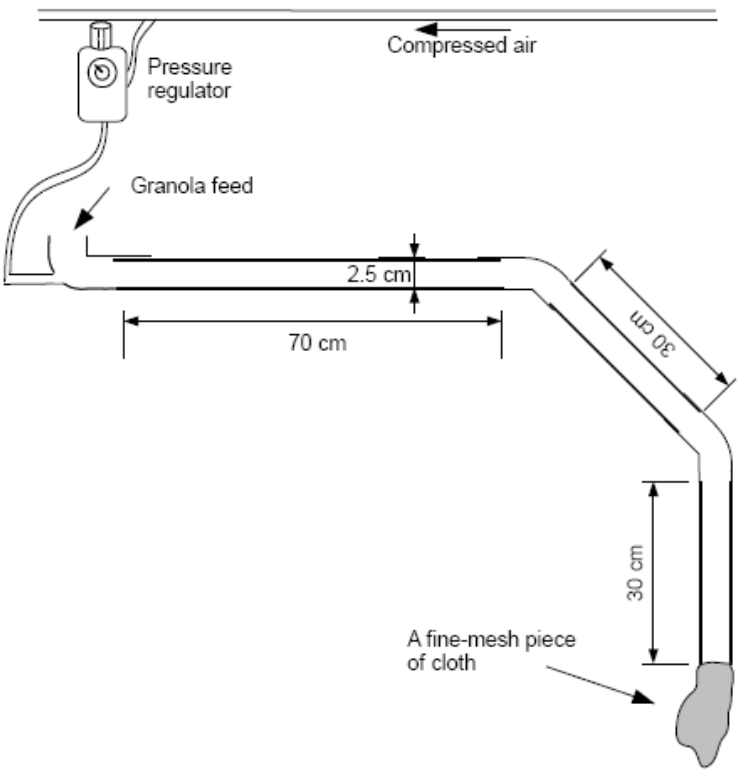


Figure 5.3: A schematic diagram of the 45° bend pipeline used for pneumatic conveying of granola

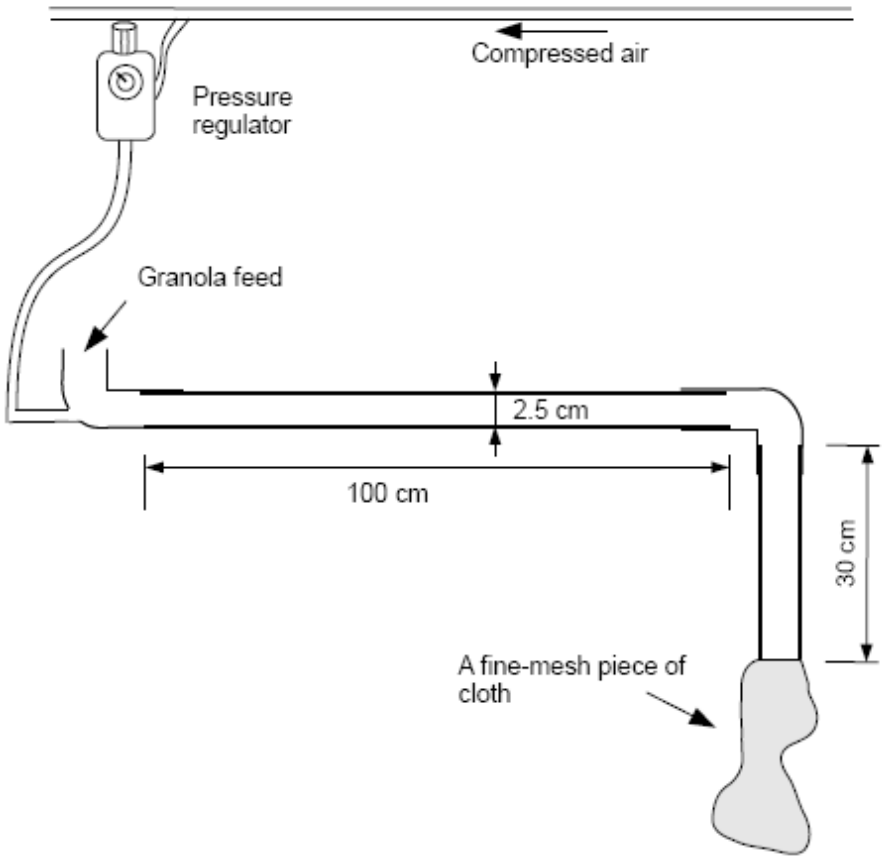


Figure 5.4: A schematic diagram of the 90° bend pipeline used for pneumatic conveying of granola

Finally, a 90° bend conveying rig was also used for propelling the granola. The initial section of the pipeline is 1 m long and a short 0.30 m length of pipeline with the same diameter as the initial section was attached to the 90° bend (Fig. 5.4). The granola is fed into the apparatus by hand at the location indicated on the diagram. A soft, fine-mesh piece of fabric is used to capture the granola at the end of the rig for measurement of its particle size distribution. For each of the configuration applied compressed air pressures of 2 bar, 3 bar and 4 bar were used.

Moreover, the transparent PMMA (Perspex) pipeline enabled observation of the breakage behaviour of particles. Particle conveying through the pipeline was recorded with a high speed camera (AOS, X-Motion, Switzerland) and visual inspection of particle flows appeared to suggest the type of breakage mechanism. A Camsizer (Retsch, Germany) digital image analyzer was used for measuring particle size distributions of the resultant granola before and after passage through a conveying rig where aggregates are transferred by compressed air at a number of different flow rates and for a number of passes.

5.3 Results and discussion

5.3.1 Breakage of granola produced in high shear granulator

Breakage of granola breakfast cereal has been studied at three different geometries namely a straight pipeline, a pipeline with two 45° bends and a pipeline with one 90° bend. Three different air pressures were applied to each configuration; 2 bar, 3 bar and 4 bar. The aggregated granola which was passed through the rig had been produced subject to impeller agitation at 150 rpm, 200 rpm and 300 rpm for 6, 9 and 12 minutes with binder addition rates of 0.22 g/sec, 0.33 g/sec and 0.65 g/sec. The effects of flow geometry, applied air pressures and manufacturing process history on granola breakage will be examined in turn. Typical granule size distribution of the granola breakfast cereal was measured after one, five, ten and twenty passages (cycles) through the conveying rig as shown in Fig. 5.5.

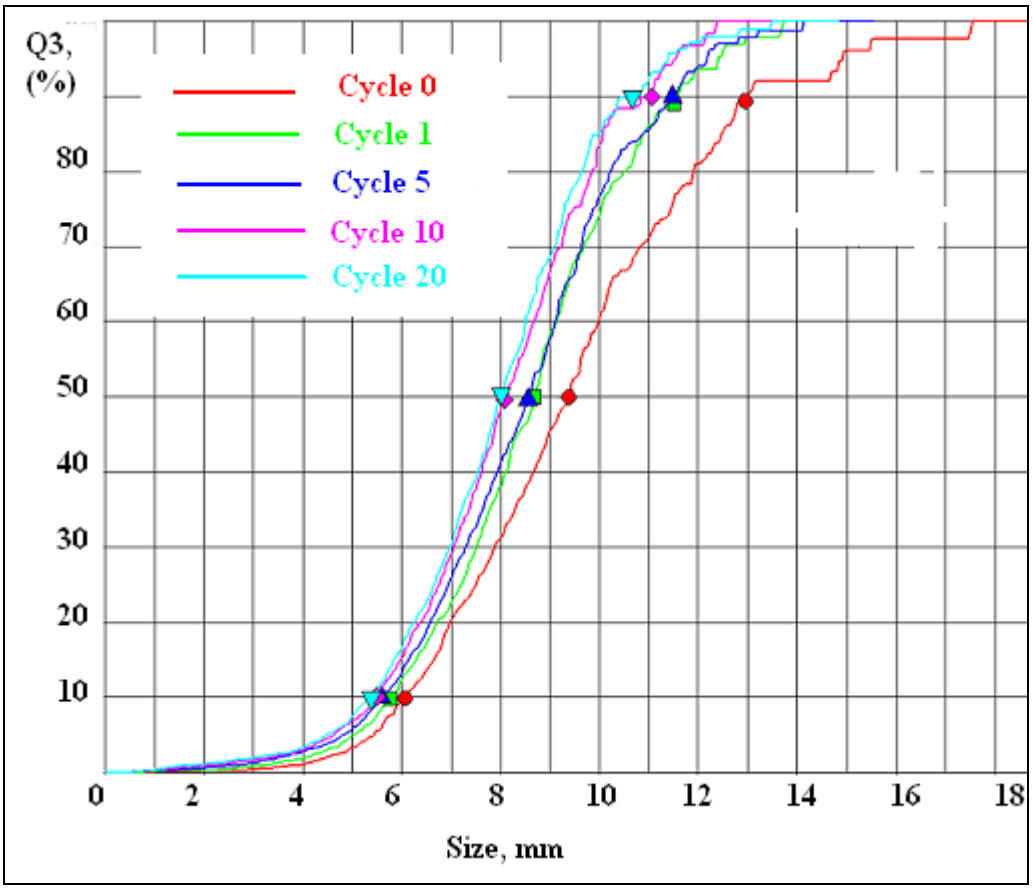


Figure 5.5: Typical granule size distribution for granola subject to various numbers of cycles through the 90° rig conveying passes (for granola produced in the high shear granulator)

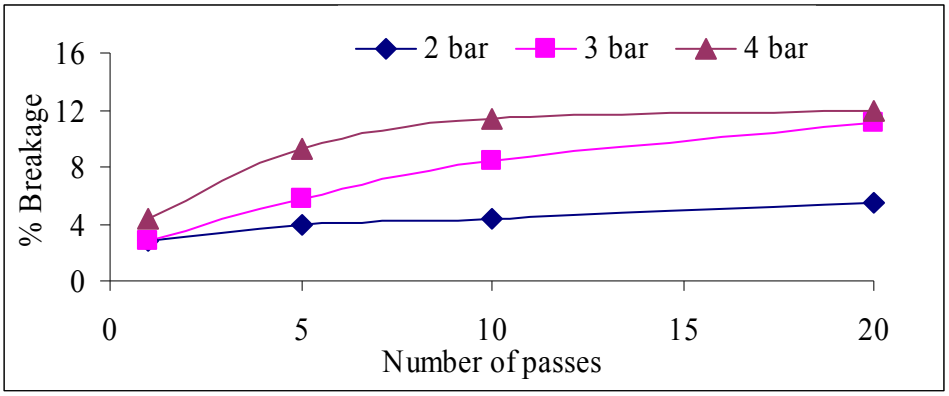
5.3.2 Effect of air flow velocity

In the conveying rig, the air velocity defines the momentum that turns into the impact load during collision. Fig. 5.6 shows the effect of air velocity (based on applied pressure) on percentage breakage. Percentage breakage ($\%B_r$) of granola breakfast cereal for a number of conveying passes was calculated as

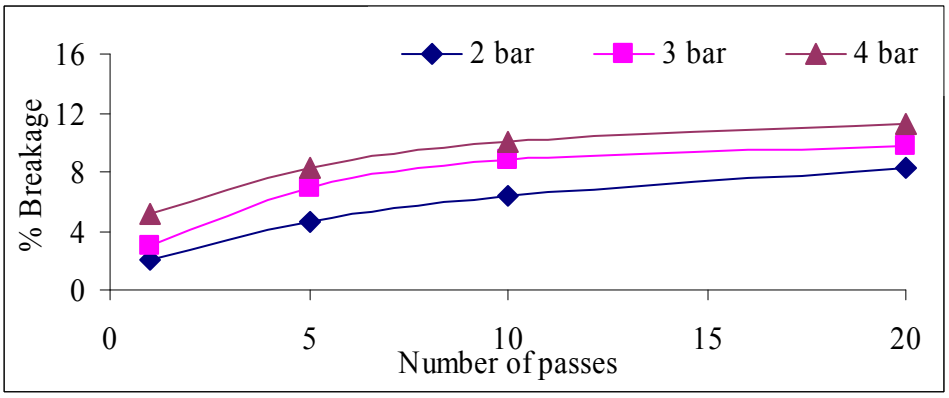
$$\%B_r = \frac{(d_0 - d_T)}{d_0} \times 100 \qquad (5.1)$$

where d_0 is initial granule size, d_T is the granule size after conveying passes.

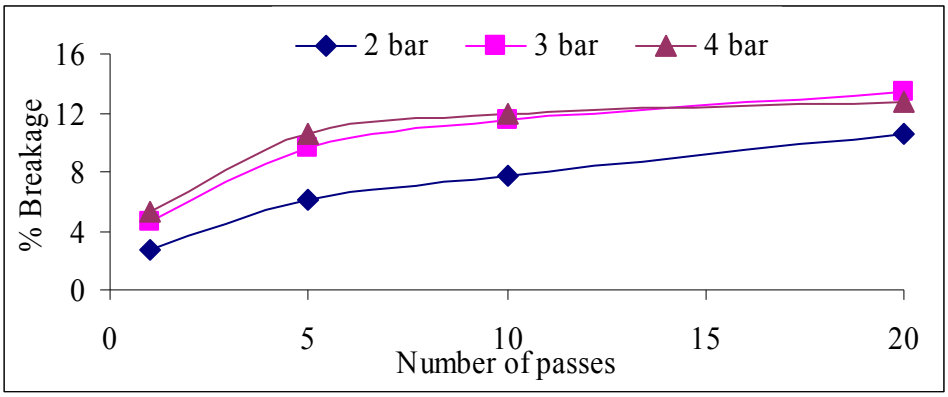
Percentage breakage increased from 2.7% to 5.3% when air pressure increased from 2 bar to 4 bar for 90° bend. Similar trends were found for other pipe geometries as breakage increases with conveying air pressure resulting in higher air flow velocity. As the air velocity becomes higher, the impact load increases. During conveying, the particles collide with one another and with the walls of the bend. As the velocity increases, the attrition was increasing significantly. It is clear from result that by increasing the air velocity and consequently the particle velocity, the attrition increases significantly.



(a) A straight pipeline



(b) Two 45° bend pipeline



(c) A 90° bend pipeline

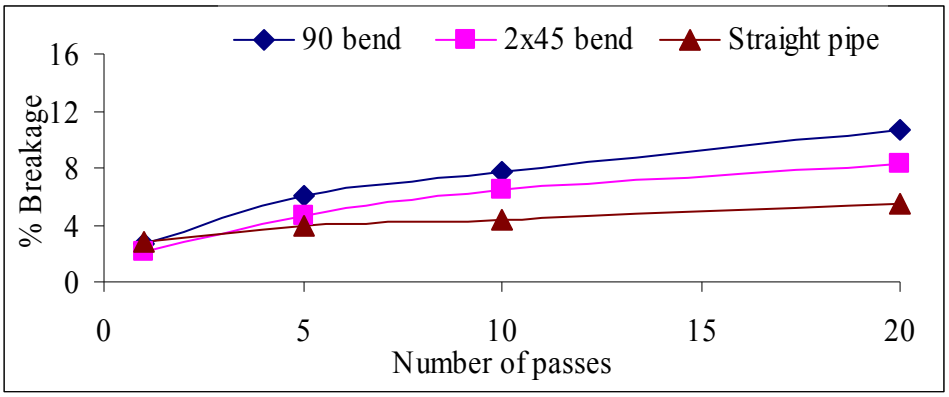
Figure 5.6: Percentage breakage of granola breakfast cereal manufactured at impeller speed 300 rpm, binder addition rate 0.33 g/sec and wet massing time 9 min as a function of air velocity with respective applied air pressure for three pipe geometries.

5.3.3 Granule breakage during pneumatic conveying at different pipe geometries

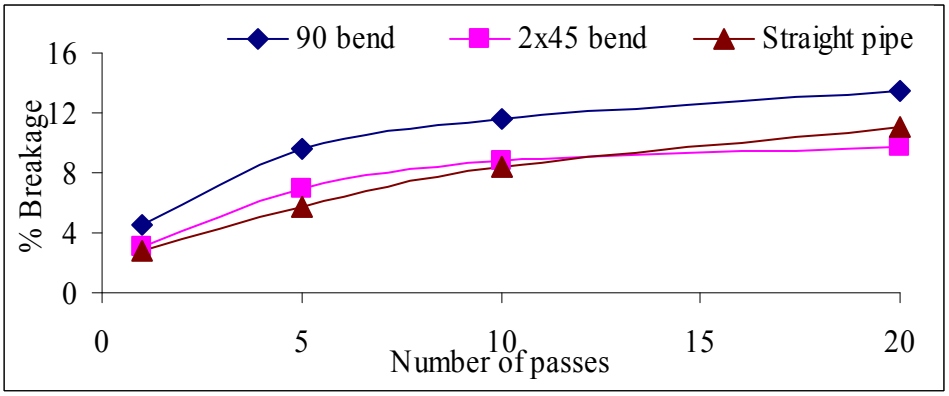
The effect of the shape of the pipe geometries on granule breakage was compared by performing tests with the following geometries: straight pipe, two 45° bend and 90° bend. The pipe geometry was found to have important effect on percentage breakage as illustrated in Fig. 5.7. Maximum breakage was found at the 90° bend geometry as might be expected. In the straight horizontal pipe, the lowest levels of attrition were found, i.e. most of the fragmentation took place in the bend. The percentage breakage was recorded after one; five; ten and twenty passes through the experimental rigs. Percentage breakage increased with applied air pressure drop for all geometries (Fig. 5.7). For example, aggregates conveyed when subject to corresponding 4 bar pressure demonstrated percentage attrition rates of 3.4%, 4.9% and 6.0% for the straight pipe, 45° bend and 90° bend respectively when subjected to

one pass through the rig. When pressure drop is increased from 2 bar to 4 bar through the 90° flow geometry, the percentage breakage ranged from 11.6% to 16.6 % after twenty passes. In addition, an increase in the number of passes results in an increase in the level of attrition. At 2 bar applied pressure, the percentage breakage increased from 3.6% to 7.2% between the 1st and 20th pass in the straight pipe rig. Higher breakage was found for the 90° bend as expected, as particle flow is forced to change the direction. This is because particles experience extensive impact loads mainly at the bends during pneumatic conveying as a result of the change in flow direction (Kalman, 1999).

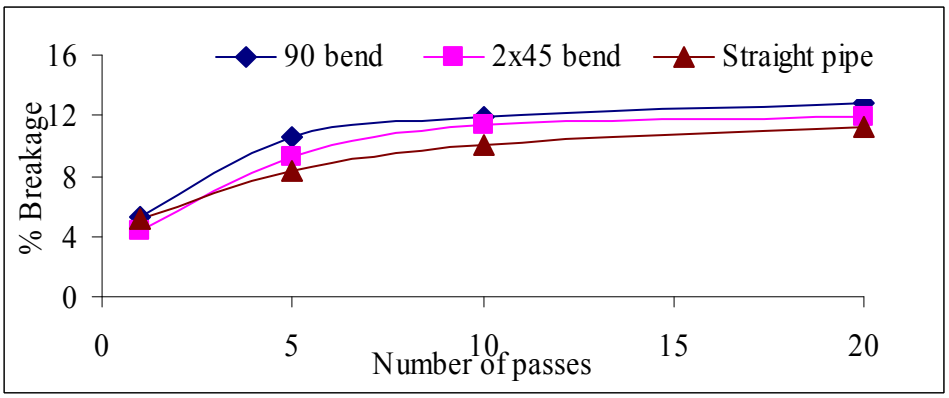
It is also clear from Fig. 5.7 that the rate of attrition is a function of the number of passes through the pipeline. The percentage breakage did not increase at the same rate between ten and twenty passes compared with earlier interaction, though there nevertheless appears to be some breakage with each pass. Maximum breakage was found during early passes, this is probably due to the preferential breakage of the weaker particles at the start of the attrition process. During pneumatic transportation, particles are exposed to repeated impacts which can result in progressive weakening of the product as a result of macro-void or crack propagation (Tavares and King, 2002). In terms of exposure time some, researchers (Han et al., 2003; Heffernan et al., 2005; Kalman, 1999; Kalman, 2001; Salman et al., 2002; Zumaeta et al., 2005) have concluded that the attrition rate decreases with time and tends towards some constant rate after a certain period of time. Once the weak particles have shattered an almost constant rate process, mainly abrasion, occurs in which the particles are smoothed and rounded.



(a) Applied air pressure 2 bar



(b) Applied air pressure 3 bar



(c) Applied air pressure 4 bar

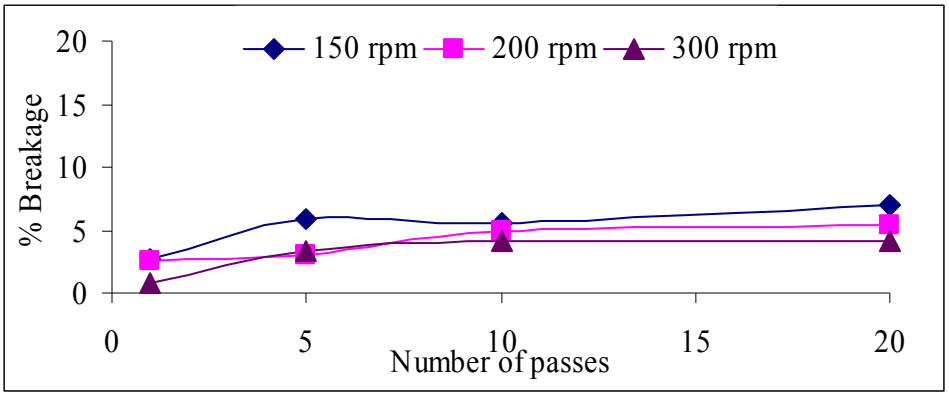
Figure 5.7: Percentage breakage of granola breakfast cereal manufactured at impeller speed 300 rpm, binder addition rate 0.33 g/sec and wet massing time 9 min as a function of different pipe geometries

3.3.4 Effect of manufacturing process parameters on granule breakage for granule produce by high shear granulation

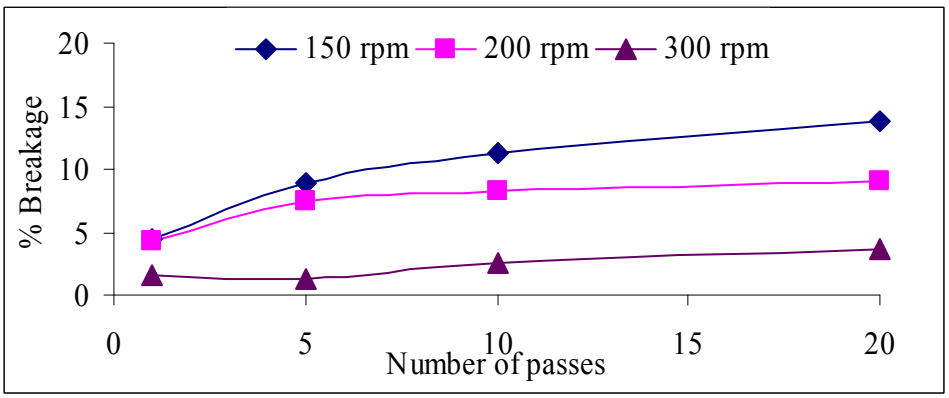
The effect of manufacturing process parameters on granule size of granola breakfast cereal formed at 150, 200 and 300 rpm is described in Chapter 3. Particles formed at higher impeller speeds are generally larger and harder than those formed at lower speeds. Particle strength, as demonstrated by levels of aggregate breakage during subsequent (conveying rig) processing are also likely to be a function of manufacturing process parameters, such as impeller speed. Conveying condition and aggregate strength have been shown to be linked as particle strength influences particle breakage and attrition in pneumatic conveying (Kalman, 1999).

The effect of impeller rotation speed on percentage breakage is shown in Fig. 5.8. Aggregates suffer the highest rate of breakage during initial passes, particularly when formed at low impeller speeds, and the rate quickly falls off after several passes. Moreover, granules produced at 300 rpm have the lowest levels of breakage while granules produced at 150 rpm shows highest breakage. After twenty conveying passes percentage breakage were 30.4%, 18.0% and 16.5% for granules produced at 150 rpm, 200 rpm and 300 rpm respectively when extreme conveying conditions applied. These results clearly express the importance of shear history during granule production on percentage breakage. This is because of the fact that granules become denser and stronger with increased applied shear force. Additionally, the breakage rates were almost negligible for granola produced at 300 rpm after initial passes. This may suggest the existence of a threshold applied shear rate during production for given subsequent processing configurations and applied pressures, which result

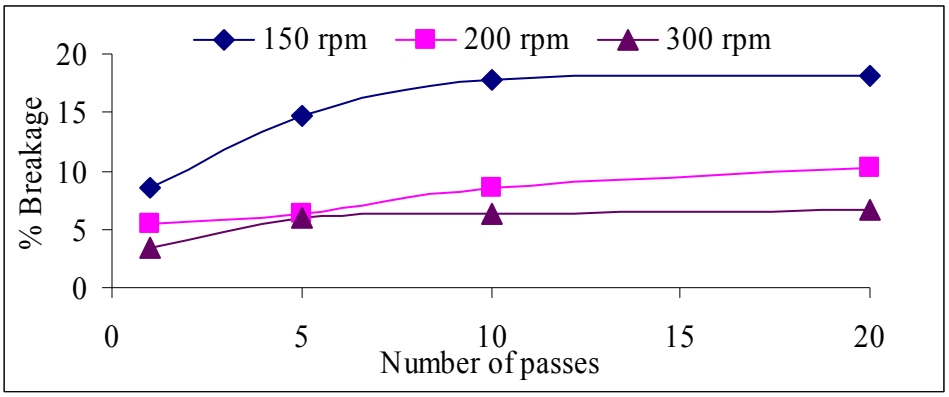
in strong particles with constantly low levels of breakage. This is in accordance with previous work carried out by Zumaeta et al.(2006).



(a) A straight pipeline



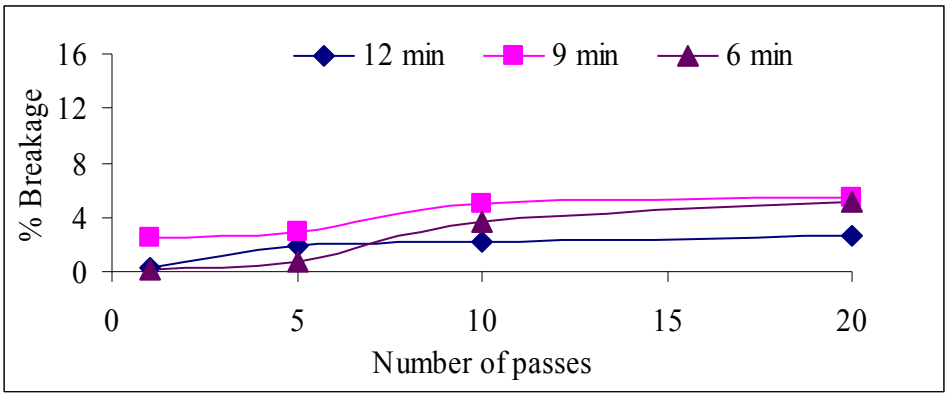
(b) Two 45° bend pipeline



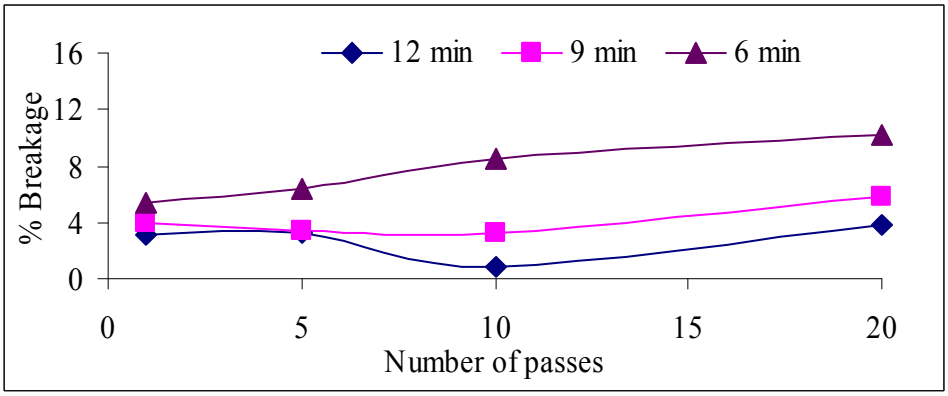
(c) A 90° bend pipeline

Figure 5.8: Percentage breakage for granules produced at various impeller speeds and flow geometries (granules produced at 0.22 g/s binder addition rate for 9 minutes of wet massing time)

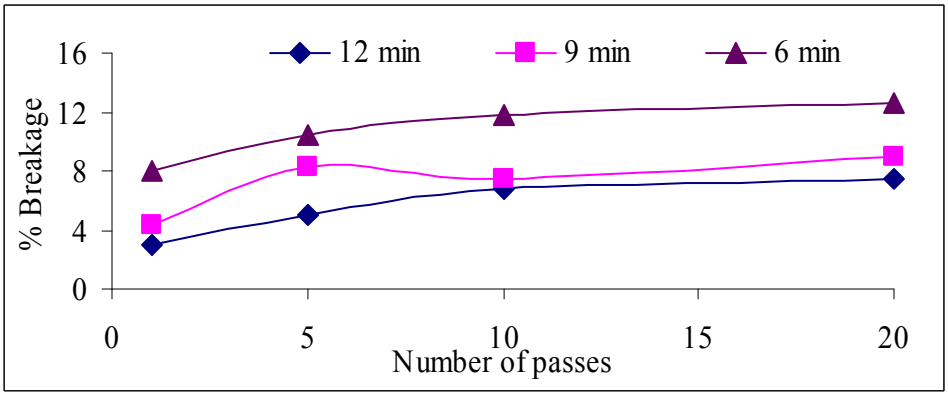
The effect of wet massing time on aggregate breakage is illustrated in Fig. 5.9. Granules formed at higher wet massing time appear to suffer the least from attrition, in general while short wet massing time (6 minute) result in increased rates of breakage. This is particularly the case where aggregates are subjected to the greater levels of impact associated with the rigs with (45° and 90°) bend. Generally for granules produced at higher wet massing times, the breakage might be expected to be smaller since hardness increases (Chapter 3). Higher wet massing times therefore produce stronger as well as larger granules with lower attrition rates compared with other aggregates.



(a) A straight pipeline



(b) Two 45° bend pipeline



(c) A 90° bend pipeline

Figure 5.9: Percentage breakage for granules produced at different wet massing times and various flow geometries (granules produced at 0.22 g/s binder addition rate and impeller speed of 200 rpm)

5.3.5 Particle breakage prediction: Simple breakage model for high shear granules

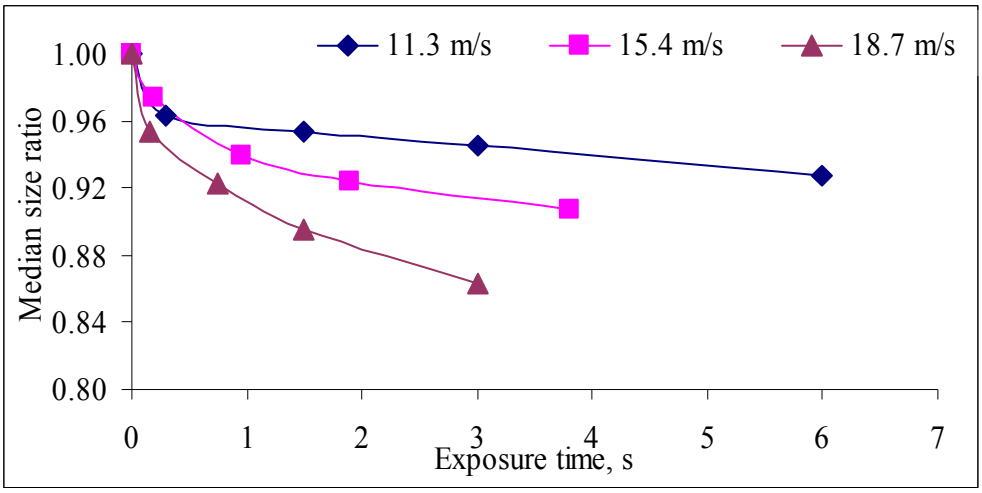
The effect of air velocity and exposure time on particle breakage was investigated for three different geometries (90° bend, two 45° bend and straight pipe). Median granule particle size was found to lie in the range 6.3 mm to 15.9 mm. In order to compare such a wide range of initial sizes, a normalised median size ratio was plotted against exposure time through the conveying pipe. Median size ratio was calculated as

$$B_r = \frac{d_T}{d_0} \qquad (5.2)$$

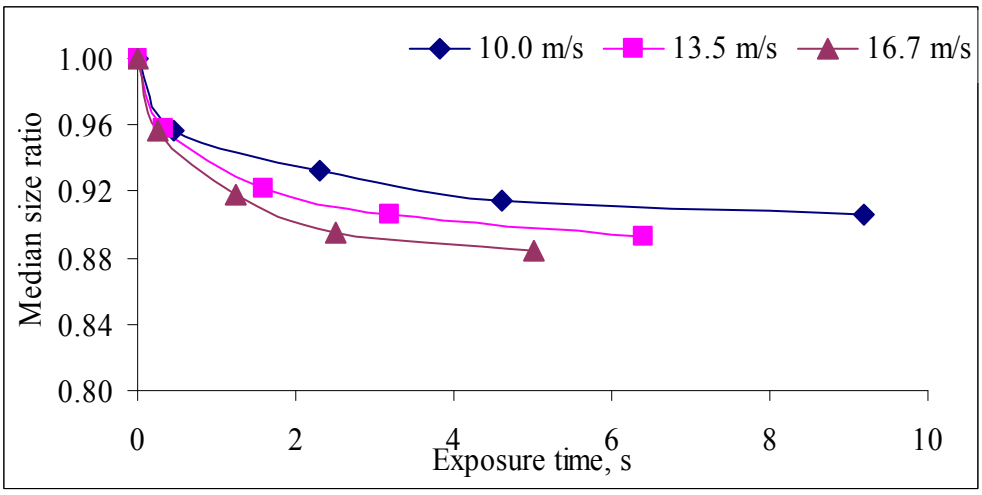
Fig. 5.10 illustrates the effect of exposure time on the median size ratio for the three applied pressures. The velocities shown in Fig. 5.10 relate to applied pressure of 2, 3 and 4 bar respectively. As one might expect, increased exposure time and increased air velocity resulted in greater granule breakage. The straight pipe rig also showed reduced percentage breakage relation to the rigs with bends, with the 90° bend pipe exhibiting the greatest proportional reduction in size. Moreover, in all cases, the average bulk of breakage occurred in the first few cycles with a clear reduction in the rate of breakage occurring as the number of cycles was increased. Zumaeta et al (2005) reported a study on particle breakage of whey protein precipitates conveyed through various geometries in a pneumatic rig and developed a power law model equation to represent the breakage frequency of the particles. A power-law model based on the above work is proposed here based on the key parameters of aggregate size, exposure time and velocity to describe the experimental data. This takes the form:

$$B_r = C_1(t + C_2)^n \qquad (5.3)$$

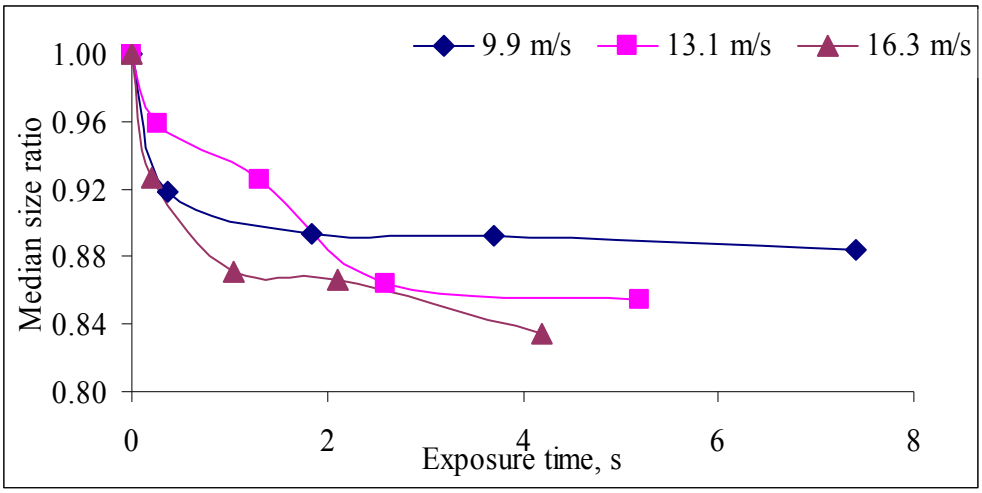
where B_r is median size ratio, t is exposure time in second (t > 0), n is -0.024; C_1 is a constant calculated as $C_1 = e^{-0.003v_a}$, where d_0 is the median granule size in mm, v_a is the air velocity in m/s and C_2 is another constant calculated as: $C_2 = d_o^{-0.969}$. The coefficient of determination (R^2) for the model was found to be 0.486.



(a) A straight pipeline



(b) Two 45° bend pipeline



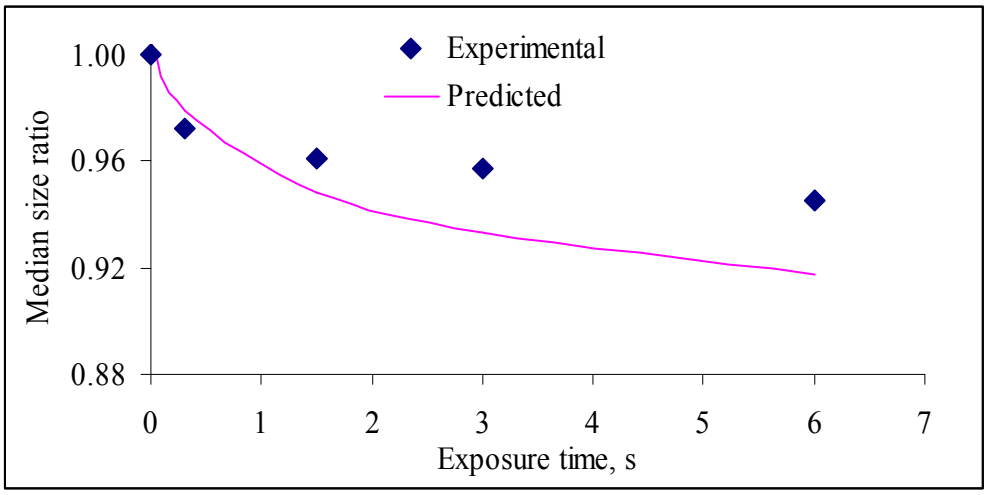
(c) A 90° bend pipeline

Figure 5.10: Effect of air velocity and exposure time on granule breakage

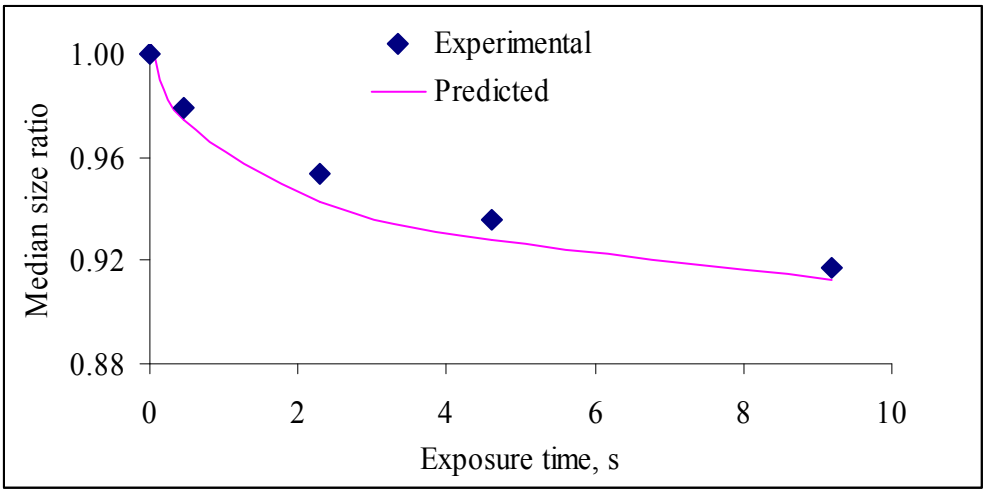
5.3.6 Model validation

The model results obtained from Eq. (5.3) were compared with the experimental results. The comparison of the model with the experimental results for the 90° bend pipeline, the two 45° bend pipeline and the straight pipeline for three applied pressures is shown in Figs 5. 11 – 5. 13. The model predicts the correct trends of the variations in median size ratio with time. Apart from the lowest applied pressure (2 bar), where there appears to be reasonably good agreement between the model and the experimental data, the model tends to underpredict the degree of breakage as evidenced by the experimental results. The model relates best to the two 45° bend i.e. in all cases lower where it offers a good correlation. A possible reason for the variation between the model and experimental data is that the model may not be able to either measure or predict the impact of various parameters such as hardness and impact forces due to large and natural variations in the complex system that present in the granola manufacturing process. Moreover there are other input factors which are not or can not be accounted for example, granules produced via the high shear

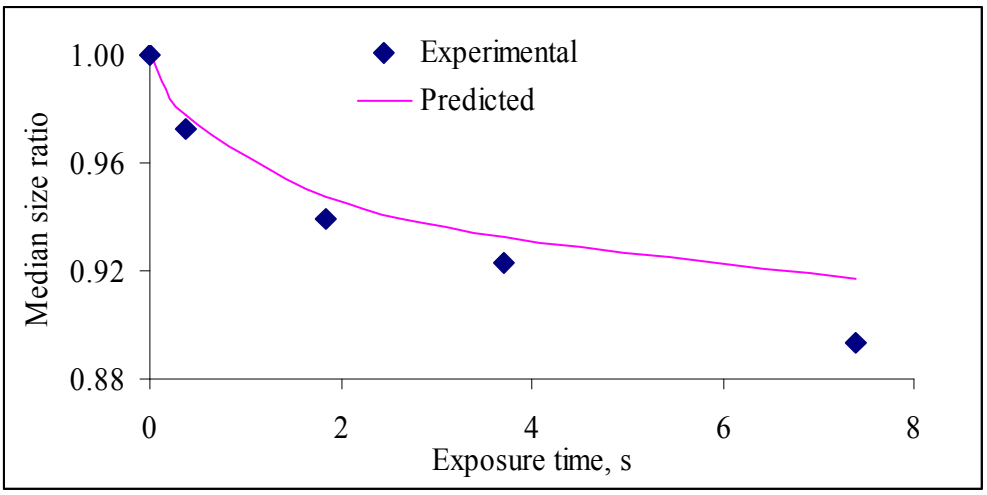
granulator are dense and the breakage mostly occurs via attrition which results in the formation of very small particles and fines. During pneumatic conveying, these smaller dusty particles are lost in the collecting cloth at the end of the rig. Granola samples were weighed before and after each conveying cycle, and it was observed that the average loss of fines is 1% (w/w) over twenty cycles. Therefore, this loss will have some effect on the experimentally measured average size sample which will be in fact higher than the measured value. While this effect is small and it should in fact lead to lower predicted experimental breakage rates, it nevertheless demonstrates that there are factors which are/can not be incorporated into the model.



(a) A straight pipeline

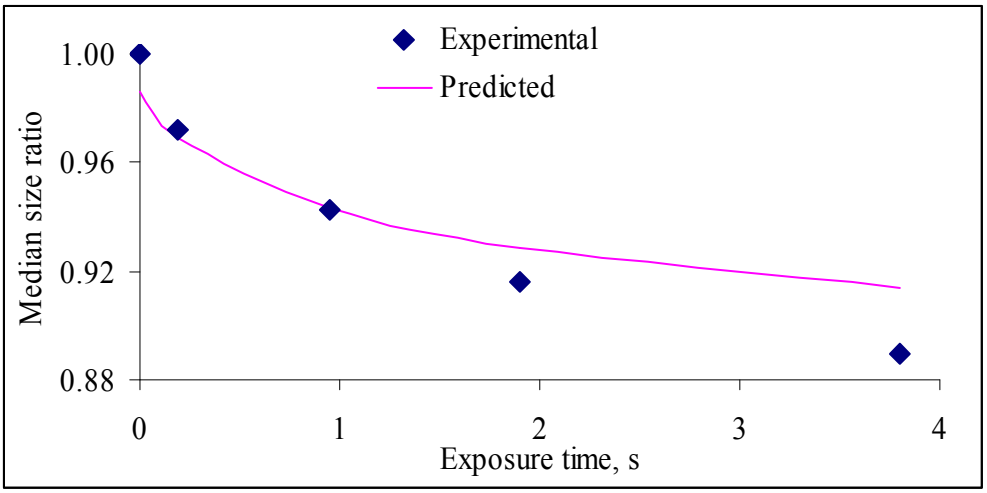


(b) Two 45° bend pipeline

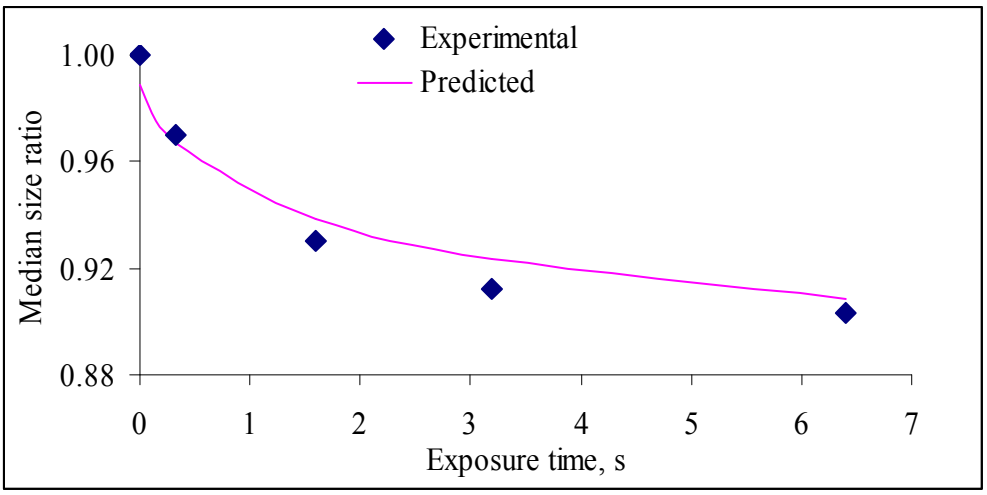


(c) A 90° bend pipeline

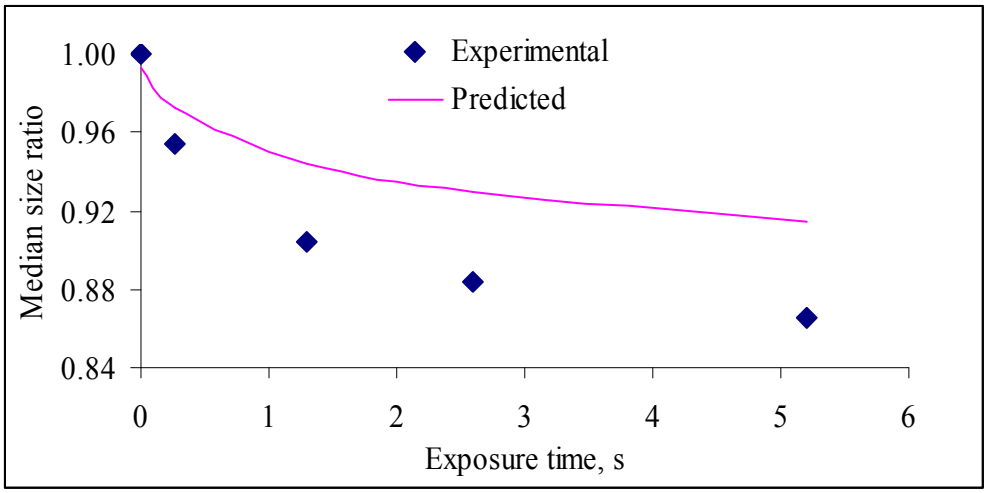
Figure 5.11: Median size ratio at different pipe geometries at applied air pressure of 2 bar (continuous line displays model results, points are experimental data)



(a) A straight pipeline

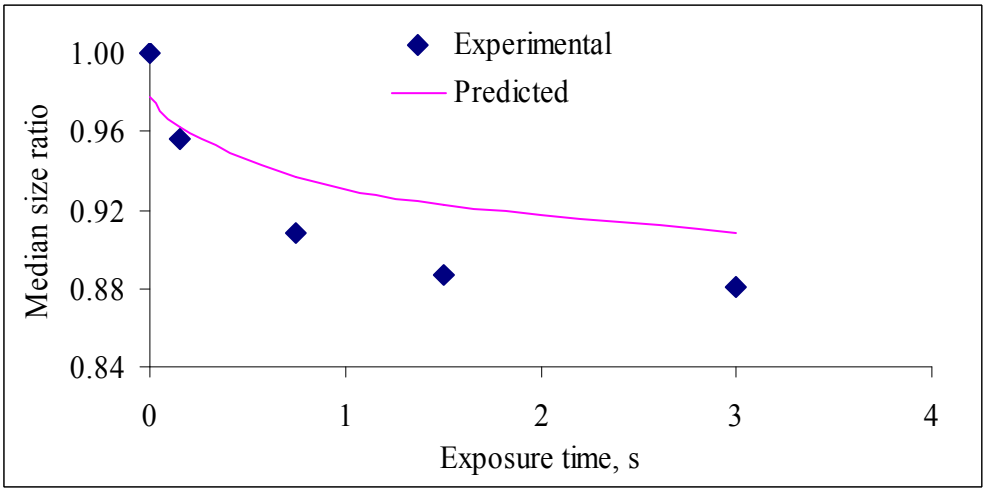


(b) Two 45° bend pipeline

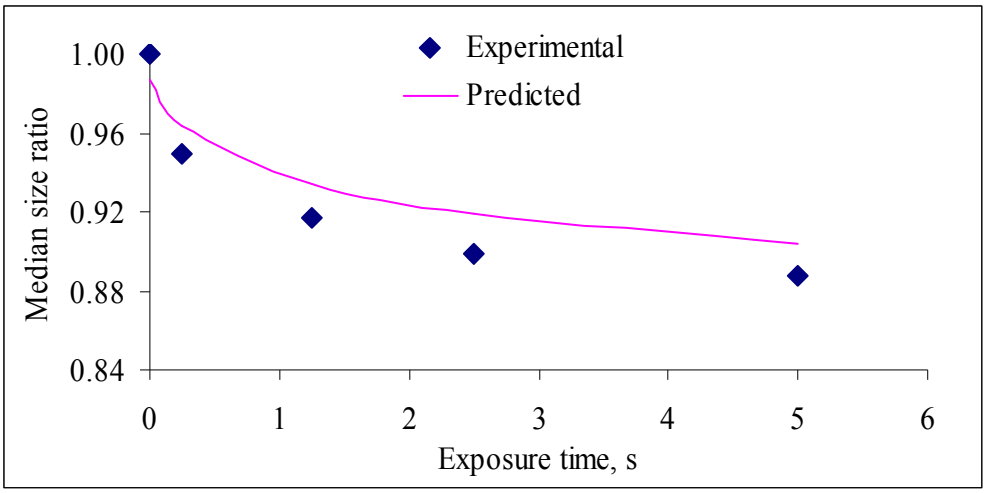


(c) A 90° bend pipeline

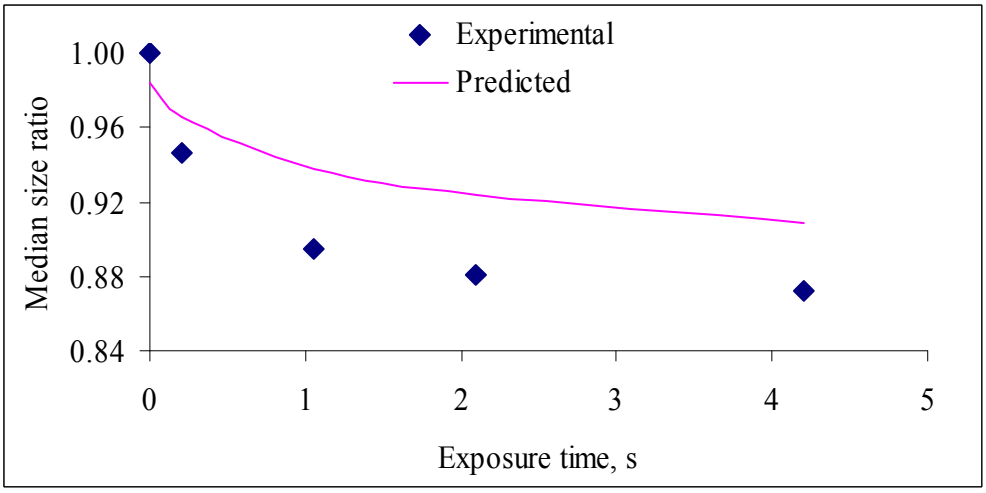
Figure 5.12: Median size ratio at different pipe geometries at applied air pressure of 3 bar (continuous line displays model results, points are experimental data)



(a) A straight pipeline



(b) Two 45° bend pipeline



(c)A 90° bend pipeline

Figure 5.13: Median size ratio at different pipe geometries at applied air pressure of 4 bar (continuous line displays model results, points are experimental data)

5.5.7 Breakage of granola produced in fluidised bed granulator

The breakage of granola breakfast cereal has been studied at three different geometries (a straight pipeline, a pipeline with two 45° bends and pipelines with one 90° bend) for granola manufactured by fluidised bed processing. Three different air pressures were applied to each configuration; 2 bar, 3 bar and 4 bar. During manufacture in the fluidised bed unit, the aggregated granola breakfast cereal was

produced subject to binder spray rates of 0.4 g/s, 0.8 g/s and 1.2 g/s and nozzle air pressure of 2 bar, 3 bar and 4 bar. The effects of flow geometry, applied air pressures and processing history on breakage will each be examined in turn. Typical granule size distribution after one, five, ten and twenty conveying passes is shown in Fig. 5.14.

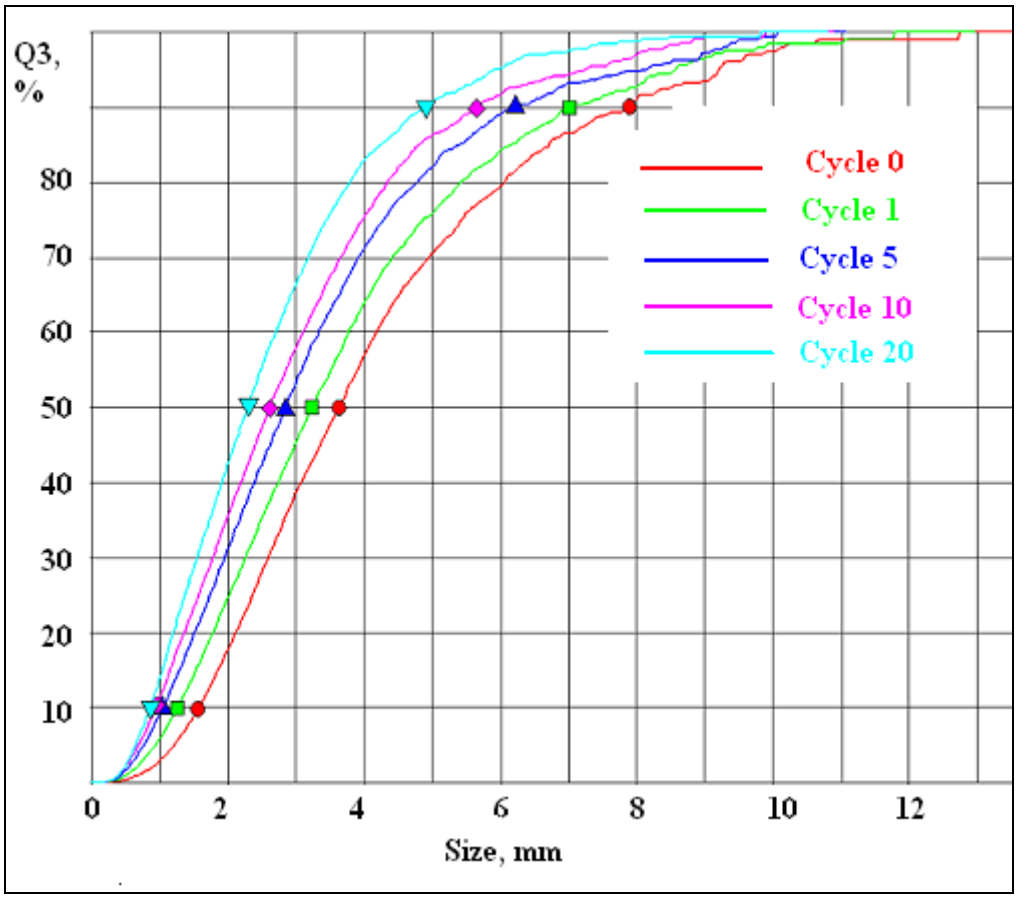
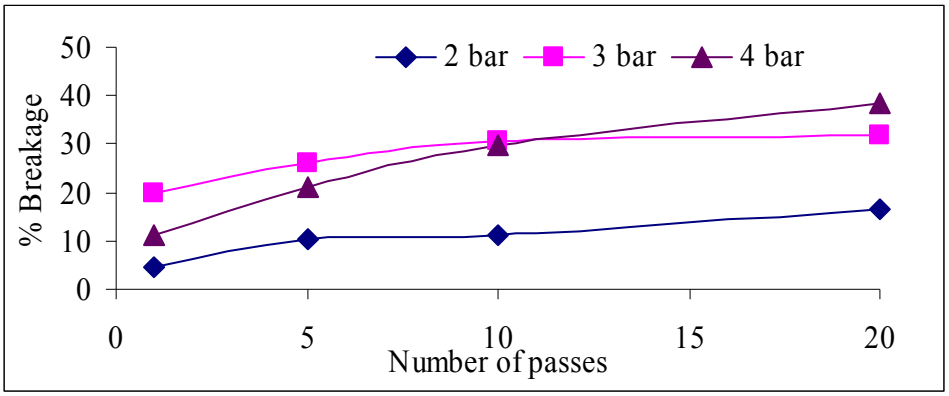


Figure 5.14: Typical granule size distribution for granola subject to various numbers of cycles through the 90° rig conveying passes (for granola produced in fluidised bed granulator)

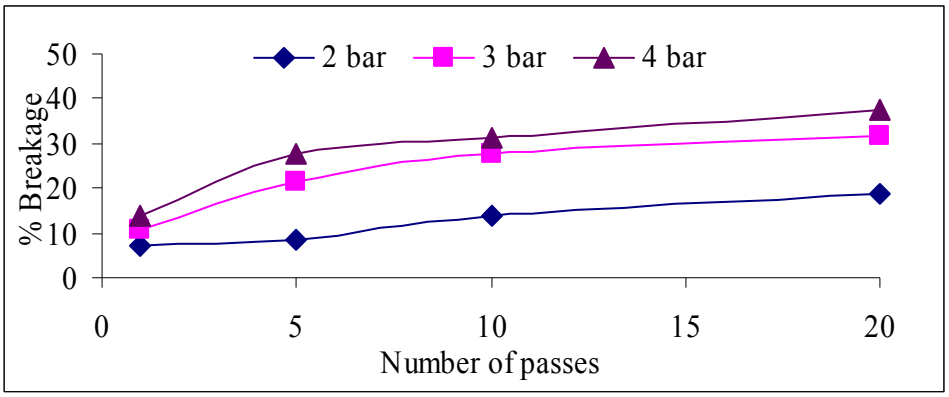
5.3.8 Effect of air flow velocity

In literature discussions on particle attrition, conveying velocity has been identified as the most important process variable (Mills, 1987; Taylor, 1998). The effect of air

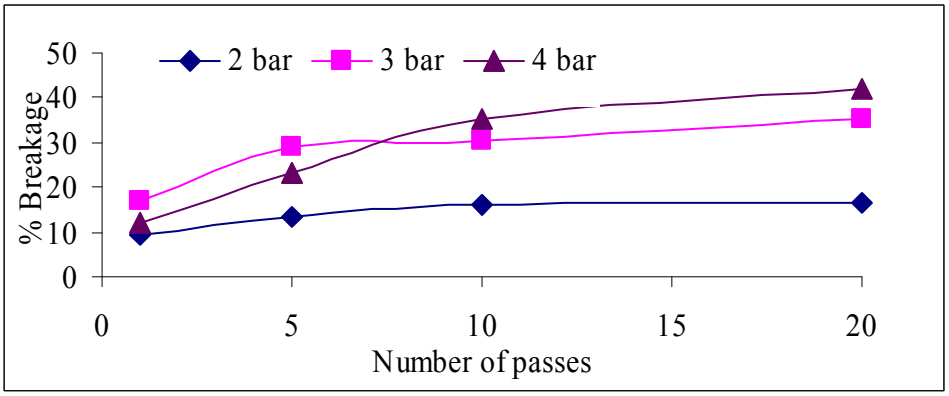
flow velocity (based on applied pressure) on percentage breakage of granola breakfast cereal is illustrated in Fig. 5.15. The granules are produced at nozzle air pressure 2 bar and binder spray rate 0.4 g/min. The percentage breakage increases from 16.6% to 42.0% when air pressure is increased from 2 bar to 4 bar for the 90° bend. A similar trend was found for other pipe geometries as breakage increases with conveying air pressure resulting in higher air flow velocity. As expected, minimum breakage was found for air velocity at 2 bar air pressure. This result agrees with Taylor (1998) who observed low attrition when the relatively low velocity used. Maximum breakage was found at early passes; this is probably due to the preferential breakage of the weaker particles at the start of the process as weak particles are broken during initial cycles leaving a stronger core which remains intact over the course of the twenty cycle regime. Overall the levels of breakage for aggregate formed in the fluidised bed unit are found higher than for the high shear granulator. This reflects the more porous, friable nature of the granules formed in the fluidised bed which have not been subjected to any impeller shear action.



(a) A straight pipeline



(b) Two 45° bend pipeline



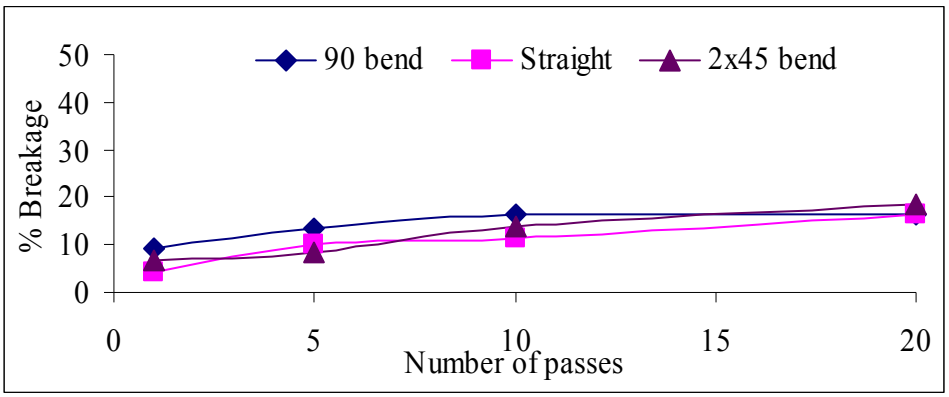
(c) A 90° bend pipeline

Figure 5.15: Percentage breakage for different applied air pressures at three flow geometries, for granules produced at 0.4 g/min binder spray rate and nozzle pressure 2 bar

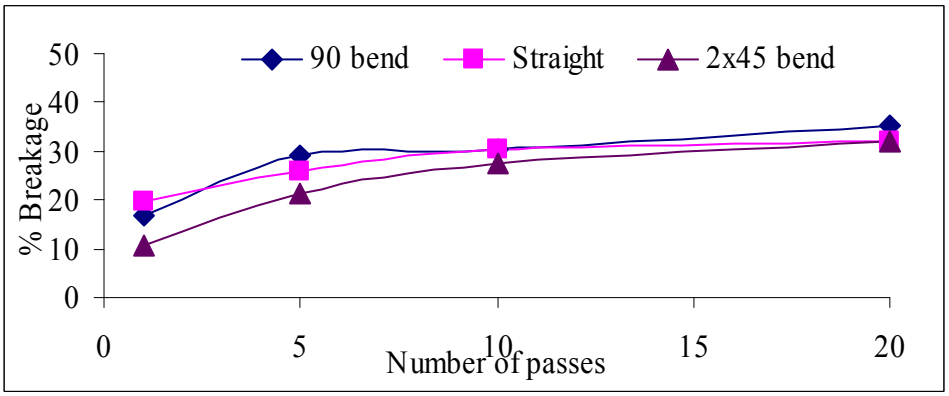
5.3.9 Granule breakage during pneumatic conveying at different pipe geometries

The effect of the shape of the pipe geometries on granule breakage produced in fluidised bed granulator was investigated by performing runs with the three selected geometries: a straight pipe, two 45° bends and a 90° bend. The effects of pipeline geometry on percentage breakage is illustrated in Fig. 5.16. The level of attrition was recorded after one; five; ten and twenty passes through the experimental test

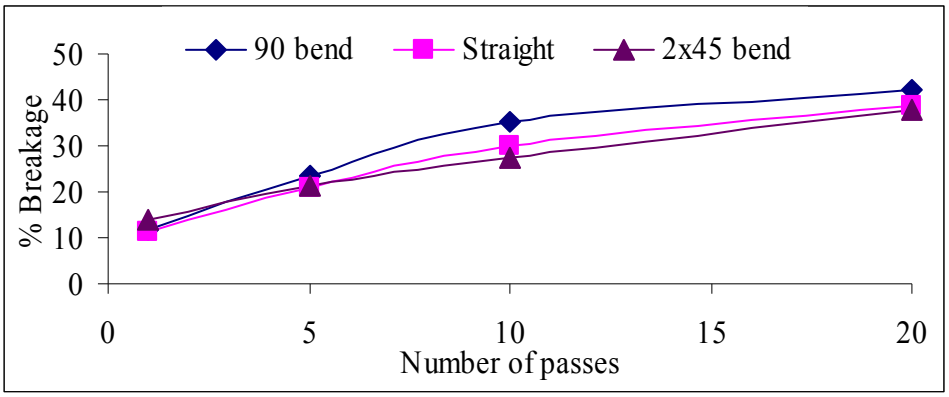
rigs. In general, an increased number of conveying passes leads to increased attrition levels. For air velocity associated with 2 bar applied pressure, the percentage attrition increased from 4.4% to 16.5% between the 1st and the 20th cycles for repeated tests in the straight pipe geometry. However the effect of pipeline of geometry did not did not appear to be as significant as operating conditions (applied pressure). For example, for the two 45° bend flow geometry, the percentage breakage after twenty passes ranged from 18.6% to 37.7% when applied air pressure increases from 2 bar to 4 bar (Fig. 5.15(b)). Maximum breakage was found at 90° bend geometry in all cases as might be expected.



(a) Applied air pressure of 2 bar



(b) Applied air pressure of 3 bar

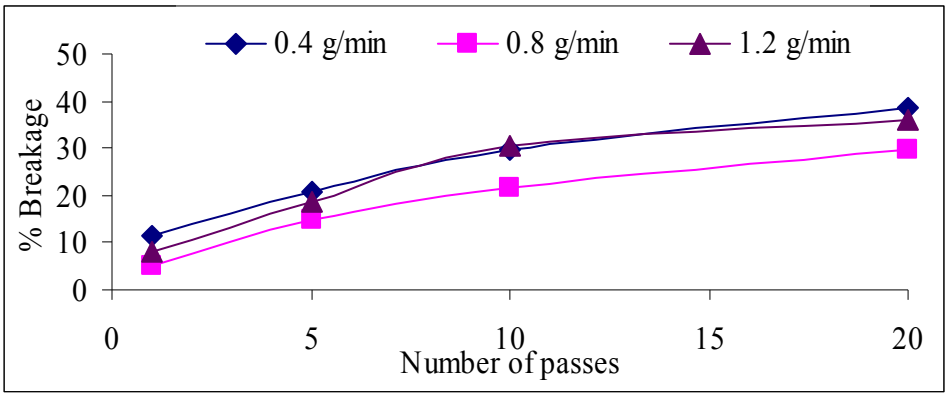


(c) Applied air pressure of 4 bar

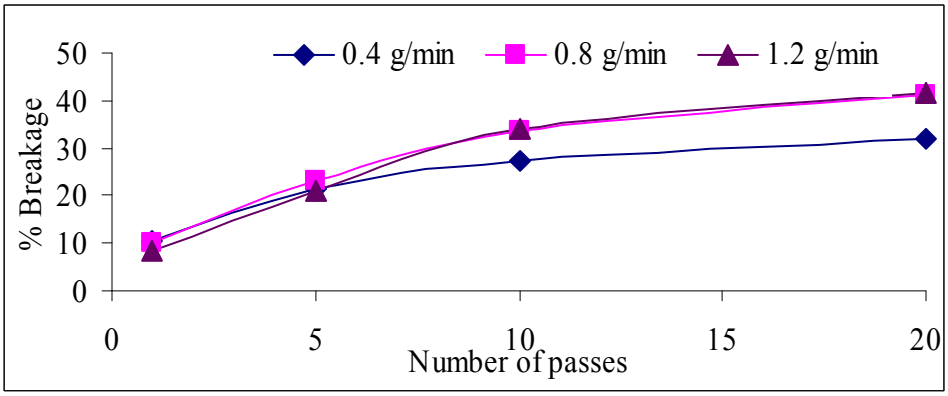
Figure 5.16: Percentage breakage rates for different flow geometries at three air pressures. Granules produced at 0.4 g/min binder spray rate and nozzle pressure 2 bar

5.3.10 Effect of binder spray rate on aggregate breakage during conveying

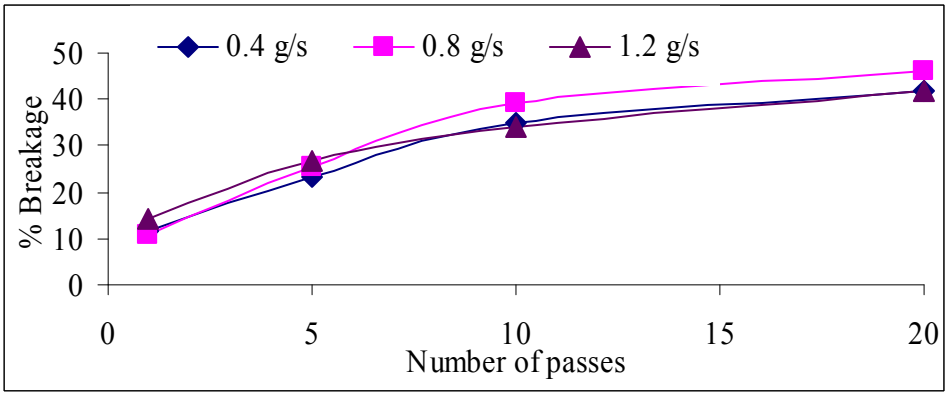
The effect of binder spray rate on the particle breakage at different pipe configurations is displayed in Fig. 5.17. Binder spray rate appears to have no appreciable effect on aggregate strength as indicated by subsequent breakage rate during conveying. This observation is further strengthened by the results displayed comparing the effect of binder addition rate at different nozzle pressures in the 90° bend pipeline (Fig. 5.18). Thus according to these results, it can be inferred that the binder addition rate does not play an important role in determining the granule strength or breakage during subsequent processing.



(a) A straight pipeline

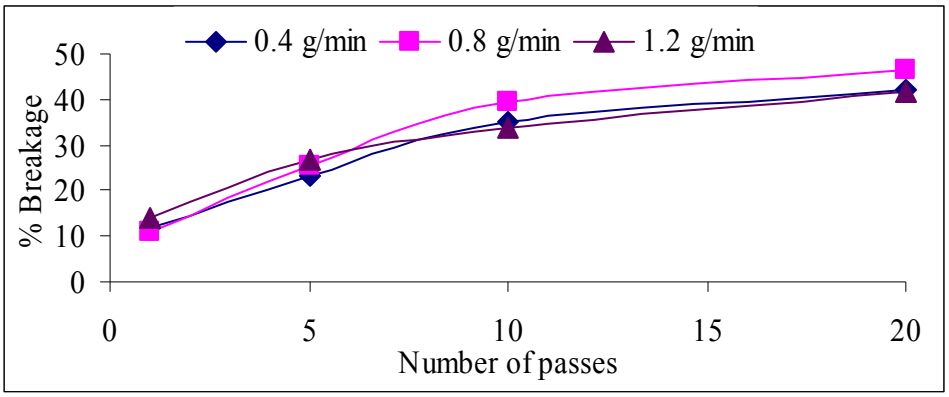


(b) Two 45° bend pipeline

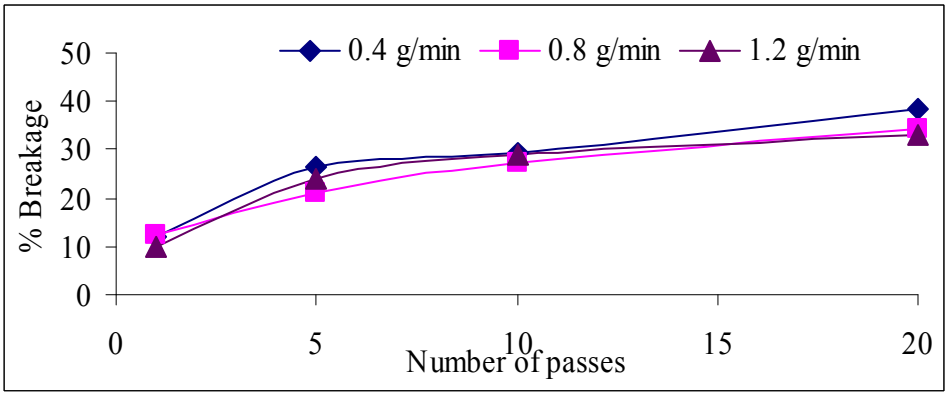


(c) A 90° bend pipeline

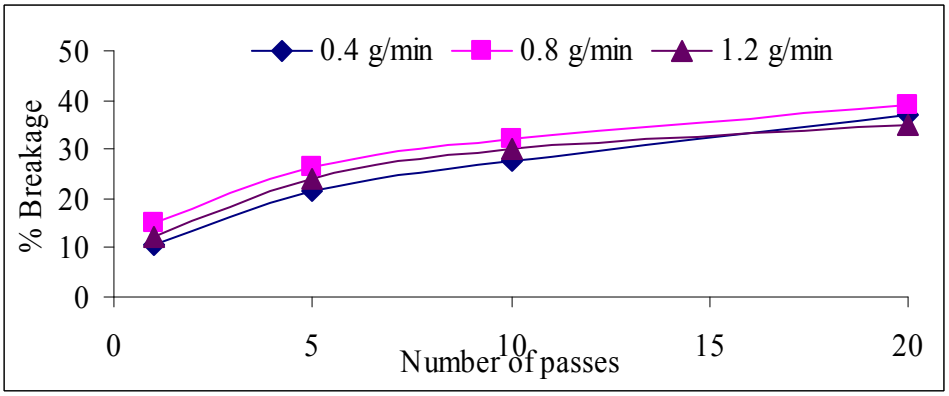
Figure 5.17: Percentage breakage for granules produced at different binder spray rates at various flow geometries, for granules produced at 2 bar nozzle pressure



(a) Nozzle air pressure 2 bar



(b) Nozzle air pressure 3 bar

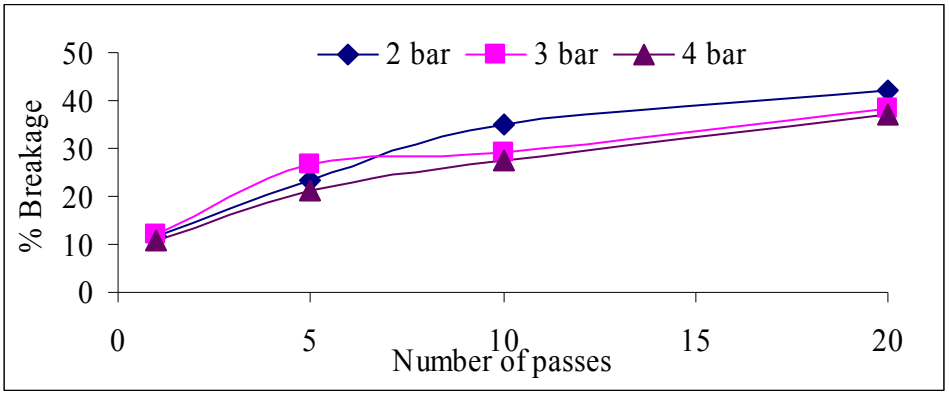


(c) Nozzle air pressure 4 bar

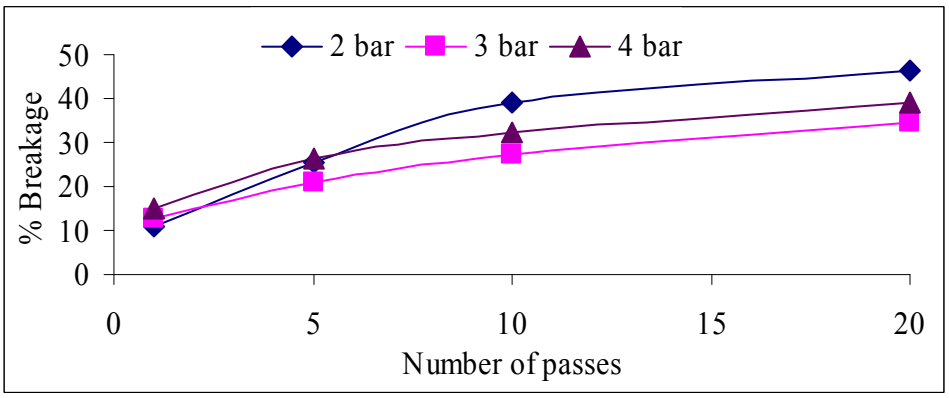
Figure 5.18: Percentage breakage for different binder spray rates at various nozzle pressures in 90° bend pipe at 4 bar conveying pressure

5.3.11 Effect of nozzle air pressure on aggregate breakage during conveying

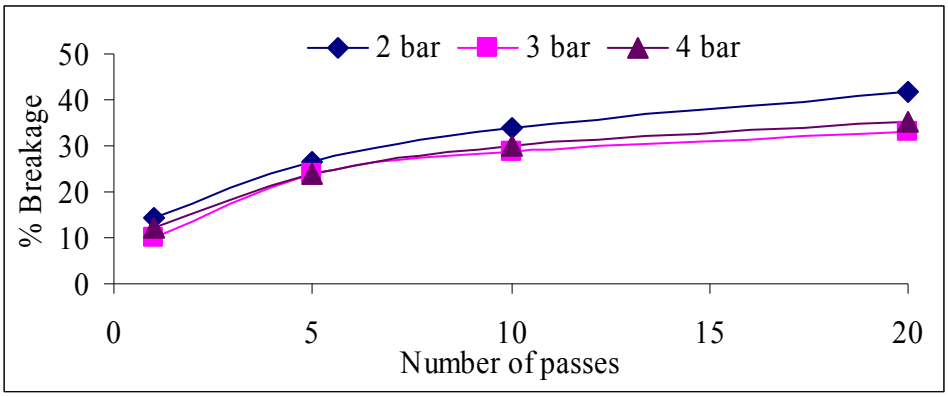
The effect of nozzle air pressure on the particle breakage at various binder addition rates in the 90° bend pipeline configuration is illustrated in Fig. 5.19. There appears to be little or no difference in breakage after initial passes for aggregates formed at all binder spray rates. However, after several passes, the breakage rate of granules formed at the lowest binder spray rate (2 bar) exhibit slightly higher levels of breakage than those produced at the other pressures. Perhaps this is due to a better distribution of binder at higher spray rate onto ingredients particles which in turn leads to slightly stronger granules better able to resist combined impacts.



(a) Binder spray rate 0.4 g/min



(b) Binder spray rate 0.8 g/min



(c) Binder spray rate 1.2 g/min

Figure 5.19: Percentage breakage rates for granules produced at different nozzle air pressures at various binder spray rates in 90° pipeline

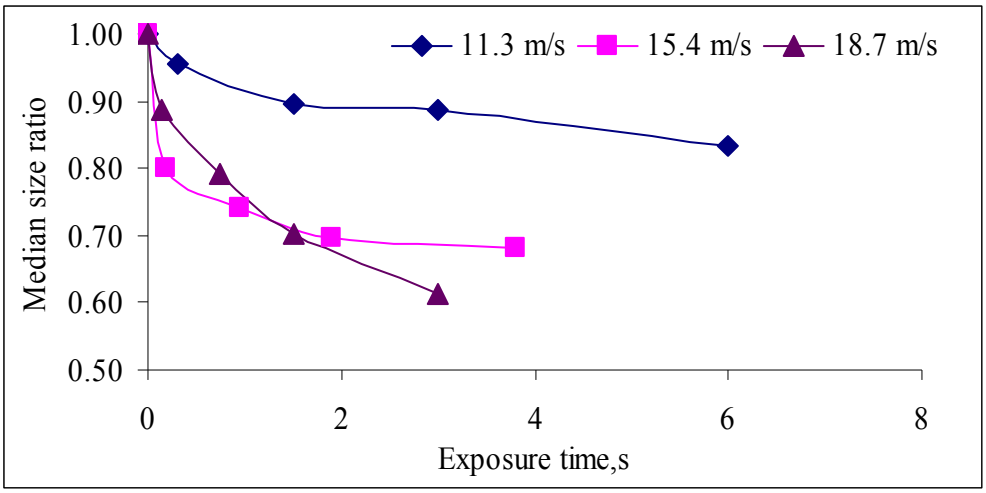
5.3.12 Particle breakage prediction: Simple breakage model for fluidised bed granules

The effect of air velocity and exposure time on particle breakage was investigated for three different geometries (straight pipe, two 45° bend and a 90° bend). Median granule particle size was measured for a number of these cycles. Median granule size was distributed in the range between 3.2 mm and 4.1 mm. The median size ratio was plotted against exposure time through the conveying pipe. Fig. 5.20 illustrates the effect of exposure time on the normalised median size ratio of the three applied pressures. The normalised size decreases significantly over initial passes through the conveying rig before falling significantly. Moreover breakage is highest for granola exposed to the highest pressure drop (4 bar) while granola exposed to the lowest pressure drop (2 bar) expresses for lower levels of attrition. This is probably due to the higher momentum at the higher velocity when some particles collide against the opposite pipe walls. As with granola produced by high shear granulation, a power-

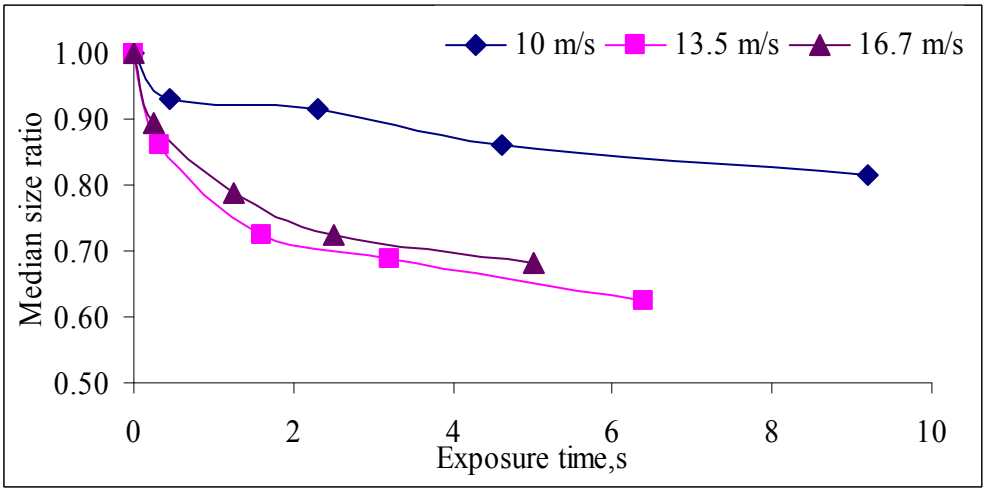
law model was developed to predict to the experimental data based on key process parameters.

$$B_r = C_1(t + C_2)^n \qquad (5.4)$$

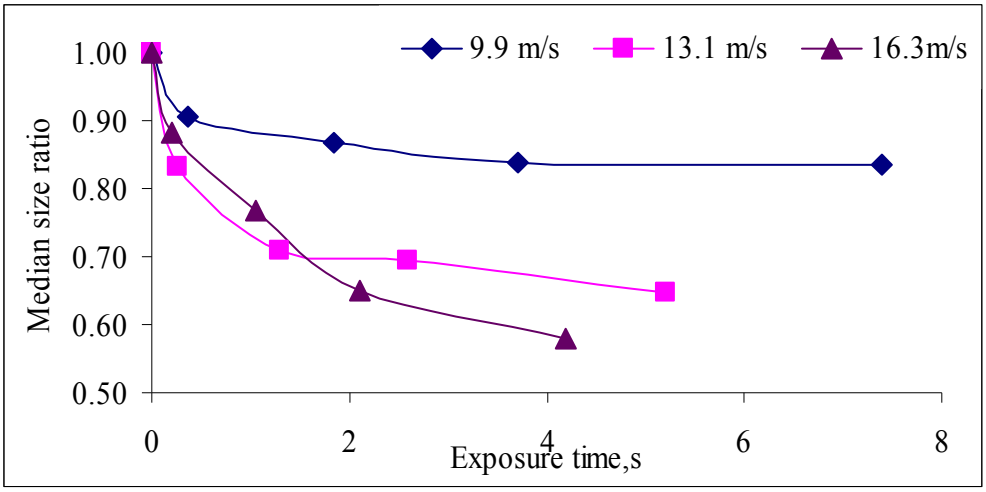
where B_r is breakage ratio, t is exposure time in second ($t > 0$), n was calculated as −0.149; C_1 is a constant calculated as $C_1 = v_a^{-0.076}$, where d_0 is the median granule size in mm, v_a is the air velocity in m/s, and C_2 is another constant calculated as: $C_2 = d_o^{-0.967}$. The coefficient of determination (R^2) for the model was found to be 0.781.



(a) A straight pipeline



(b) Two 45° bend pipeline



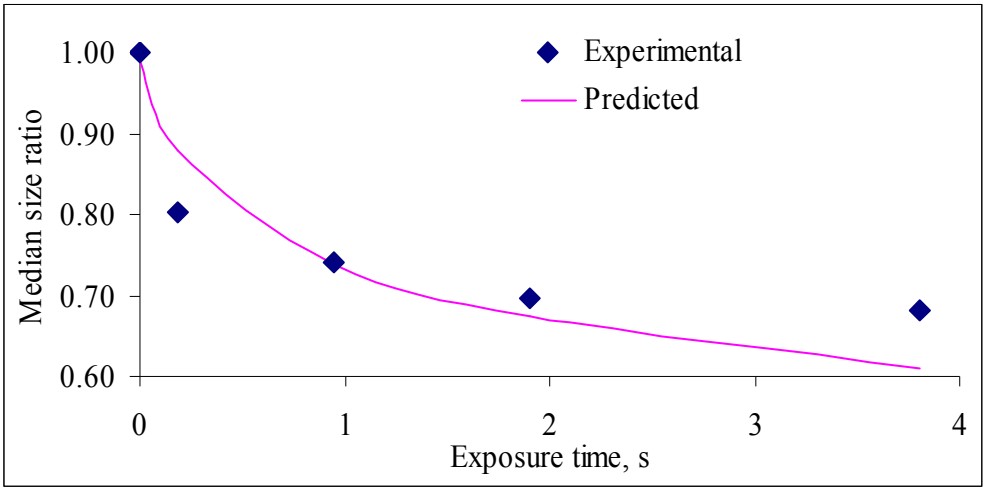
(c) A 90° bend pipeline

Figure 5.20: Effect of air velocity and exposure time of granule breakage

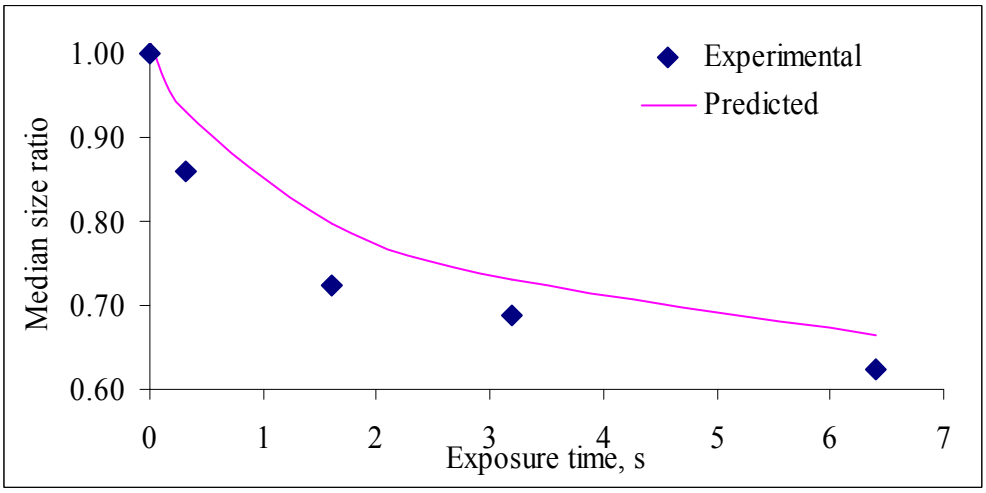
5.3.13 Model validation

As with the earlier model, the granule median diameter coefficient (d_o) is indicative of the granola manufacturing history. The results obtained from Eq. (5.4) are compared with experimental outputs for three different pipe configurations (Fig. 5.21). Overall it shows good agreement between the model and the experimental results. Although the model marginally underpredicts the level of breakage for the experimental results for the 90° bend and the two 45° bend pipelines, it provides a reasonable estimate with respect to the experimental results. On the other hand, the model slightly overpredicts breakage during a straight pipeline with high exposure times (Fig 5.21(a)). The largest disagreement between measured and predicted median size ratio was found towards the early conveying passes (upto 5) as higher granule breakage found at early passes during through the 90° bend and the 45° bend pipelines. The granules produced in fluidised bed granulator break a more easily during conveying than those produced by high shear granulation thus producing a

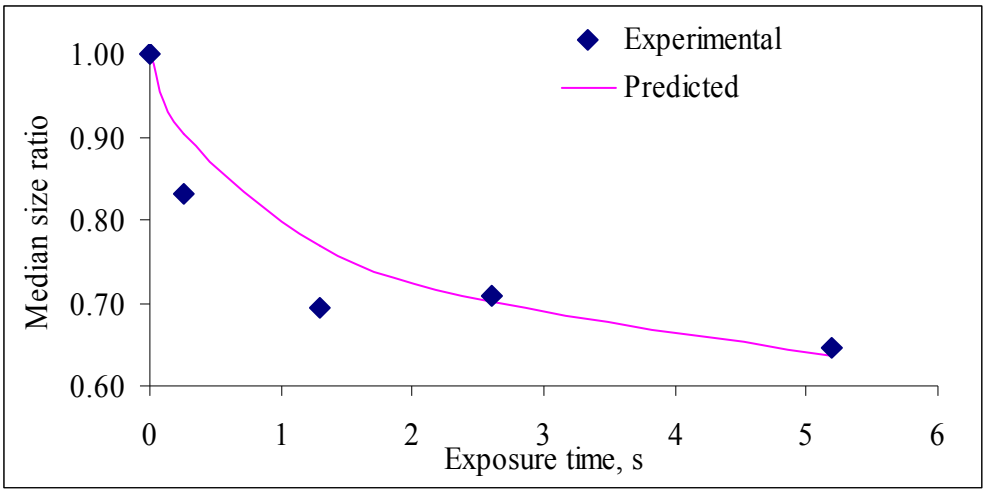
large degree of very small particles and fines. These smaller dusty particles are lost in the collecting cloth at the end of the rig. These losses tend to minimise the experimentally measured breakage so that it becomes lower than its actual value. Again, this could have a small impact on the accuracy of the actual breakage figures.



(a) A straight pipeline



(b) Two 45° bend pipeline



(c) A 90° bend pipeline

Figure 5.21: Granule median size ratio at various pipe geometries at applied pressure of 3 bar (continuous line displays model results, points are experimental data)

5.3.14 Model agreement against experimental data

The model can be used to predict the median size ratio within the range of conditions investigated. The developed model outputs were compared with the median size ratio of the observed experimental values. This is shown in Fig. 5.22. The model shows a reasonable fit with the experimental data, in particular given the complex nature of the natural product being produced, demonstrating R^2 value of 0.781.

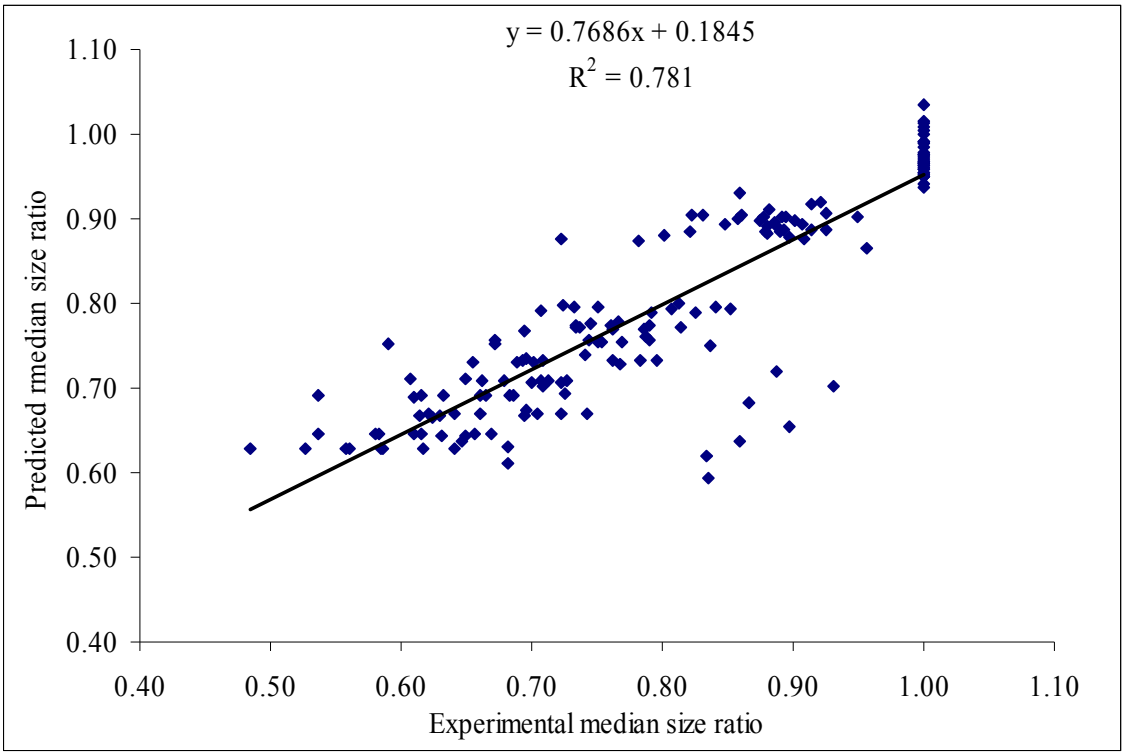


Figure 5.22: Experimental and predicted values of median size ratio

On a more general level however, it is clear that during pneumatic conveying the particles experience extensive impact loads at bends as a result of acceleration due to the change in direction. This is something which might be considered for any future development of the model, as something which may result in closer alignment between experimental and predicted (model) results. For example, in the case of the model employed, the straight pipe breakage rate shows large variation from the model as it is incapable of differentiating between the impact forces associated with the rigs with bends and for the straight pipe (0°).

5.4 Conclusions

In this chapter, the breakage of the granola during the pneumatic conveying has been examined. In the first section, the breakage of aggregate granola breakfast cereal manufactured in a high shear granulator and exposed to subsequent pneumatic

conveying was considered. It was observed that the percentage breakage of aggregates depends on the flow geometry, air flow rate and original particle size (itself a function meaning of impeller speed in the high shear granulator). Moreover, it was also shown that the aggregate manufacturing history can significantly affect the granule breakage. Applied impeller shear rate (agitation intensity) during high shear granulation is seen as particularly important in this regard and there appears to be an agitation threshold above which hard strong granules are formed which exhibit little breakage during subsequent processing. Wet massing time also has a significant effect on breakage. The highest breakage rate observed at the 90° bend pipeline geometry at 4 bar applied air pressure with granola produced at the lowest impeller speed of is 150 rpm for the shortest wet massing period of 6 minutes. On the other hand, aggregates produced at low agitation intensities tend to exhibit a higher degree of friability and conveying conditions assume significantly greater importance. A power-law model is derived to predict the median size ratio at various applied air velocities using a number of pipe configurations, taking into account shear histories by means of particle diameter.

In the second section the particle breakage of granola breakfast cereal in a fluidised bed granulator was taken into account. The effect of process parameters binder spray rate and nozzle pressure on the attrition with the number of passes through different flow geometries were examined. It was observed that the granule breakage of the particles depends on the flow geometry and air flow rate associated with pneumatic conveying. Furthermore, it was also shown that the aggregate production history does not appear to have a significant effect on the level of breakage, on the other hand no apparent trend was observed for various binder addition rates. One exception here might be that at lower nozzle pressure during binder spraying,

aggregates formed seem to be more prone to breakage during subsequent processing, particularly after extended exposure times. The highest breakage observed at 90° bend pipeline geometry at 4 bar applied air pressure. A power-law model was used to predict the median size ratio by using a number of pipe configurations.

The rate of breakage during pneumatic conveying decreased with number of passes in all cases. Granules produced in fluidised bed granulator shows significantly higher breakage than the granules formed in high shear granulator. The insights gained from this work can aid in developing a process for the production and conveying of granola to match consumer and manufacture requirements and for other similar products.

Chapter 6

General discussion and conclusions

In this study, optimisation of granola breakfast cereal manufacturing process by wet granulation and pneumatic conveying has been carried out. Granola is an aggregated food product used as a breakfast cereal and in cereal bars. Processing of granola involves mixing the dry ingredients (typically oats, nuts, etc.) followed by the addition of a binder which can contain honey, molasses and/or oil. The breakfast cereal market has been experiencing significant growth, and it is forecasted that in 2013 the global breakfast cereals market to have a value of $25,498 billion, an increase of 14.8% since 2008 (Anonymous, 2009). Aggregate food products can be produced via a number of manufacturing processes. In this work, the design and operation of two parallel wet granulation processes to produce aggregate granola products were incorporated:

a) a high shear mixing granulation stage followed by drying/toasting in an oven. b) a continuous fluidised bed followed by drying/toasting in an oven. High shear granulation is a more straightforward process compared with fluidised bed granulation. It is easy to handle fine particles and highly viscous liquid binder in a high shear granulator. In addition, there are less dusty particles associated with the final product produced in the high shear granulator than that produced in a fluidised bed granulator. However, high shear granulation is more amenable to producing granules which are denser and more spherical than fluidised bed granulation, particularly over extended processing times. Aggregates which display high sphericity and high density aren't preferred for commercial food products such as

breakfast cereals. Therefore, it is recommended that the total granulation time should be carefully selected to avoid producing overly dense granules. Moreover, it was observed that the dry mixing period, which takes place prior to the wet massing period, has a positive effect on the homogeneity of the final granola products. Therefore, it is essential to include a prior dry mixing period as part of the total granulation process despite the fact that this will increase the total energy consumption during the granulation. Three important process variables; impeller rotation speed, binder addition rate and wet massing were selected and their influences were investigated. It was shown that these parameters are key variables in determining the properties of produced granola breakfast cereal, in particular impeller speed. After that, binder addition rate and wet massing time shows significant impacts on granola physical properties. Increasing the impeller speed and wet massing time increases the median granule size while also presenting a positive correlation with density. The combination of the highest impeller speed and lowest binder addition rate resulted in granules with the highest levels of hardness and crispness.

In fluidised bed granulation, aggregation and drying processes occur in the same place; therefore this process is economically efficient and advantageous from a containment perspective. On the other hand, for the granola system studied, it is very difficult to find optimum operating parameters via this mode due to the range of ingredients, both fine and coarse, the inherent of natural variability that they posses and the variability in process parameters such as bed temperature and fluidising properties of the bed. Additionally, a mixture of honey and water was used as a binding agent which has high viscosity. This restricts spraying of binder liquid onto the particles effectively as it sticks to the wall of the fluidised bed chamber. Another

limitation of using fluidised bed granulation is that the fine particles (e.g. oat beta glucan) become trapped in the filter of the equipment hence requiring extra cleaning and preparation. Nozzle air pressure shows significant influence on granule size. The experimental results show that a decrease in nozzle air pressure leads to larger mean granule size. The combination of lowest nozzle air pressure and lowest binder spray rate results in granules with the highest levels of hardness and crispness.

The insights gained from this work can aid in developing processes for the production of granola or similar products to match with consumer and manufacture expectations and requirements. Commercial granola product was obtained and compared with the products produced in this study in terms of granule size and textural properties. The commercial granola samples show large variation in terms of size and textural properties across the brands investigated. Depending in the type of granola required (eg. to be sold alone or as accompaniment to yogurt) different manufacturing process and process parameters may be recommended. Large size granules can be obtained by high shear granulation processes. However lower hardness can be achieved by lowering impeller speed. From a consumer perspective, consumer preference tests associated with this work showed a preference for hardness levels below 100N. Commercial granola with yoghurt shows smaller size and hardness values than product sold alone. Lower size and hardness values can be achieved in a fluidised bed granulator which may be considered most suitable for these product type characteristics. Granola produced in a high shear granulator shows higher crispness values. The desired granola properties can be achieved by judiciously selecting appropriate process parameters.

The study also examined the particle breakage of granola during pneumatic conveying produced by both a high shear granulation and a fluidised bed granulation process. Products were pneumatically conveyed in a purpose built conveying rig designed to mimic product conveying and packaging. Three different conveying rig configurations were employed; a straight pipe, a rig consisting two 45° bends and one with 90° bend. It was observed that the least amount of breakage occurred in the straight pipe while most breakage occurred through a 90° bend pipe. In general, increasing the impact angle increases both the levels and rate of breakage. Particle breakage increases with applied pressure drop (and hence product velocity), and a 90° bend pipe results in more attrition for all conveying velocities relative to other pipe geometry. The impact load in the bend may exceed the threshold necessary to trigger the fracture mode in the particles, resulting in high breakage. Additionally for the granules produced in the high shear granulator, those produced at 300 rpm while being the largest also have the lowest levels of proportional breakage while smaller granules produced at 150 rpm have the highest breakage. Aggregates produced at low agitation intensities tend to exhibit a higher degree of friability and in these intensities conveying conditions assume significantly greater importance. This effect clearly shows the importance of shear history (during granule production) on breakage during subsequent processing. This is because of the fact that granules become denser and stronger with increased applied shear force. In terms of the fluidised bed granulation, there was no single operating parameter that was deemed to have a significant effect on breakage during subsequent conveying. However breakage was significantly more appreciable for all manufacturing operating conditions than for those produced by high shear granulation. In all cases, a simple

pipeline conveying system design (without 90° bends where possible) and with as low a carrying velocity as possible is recommended to reduce breakage.

A simple power law breakage model was developed and found to be suitable for predicting the breakage of granola breakfast cereal at various applied air velocities using a number of pipe configurations, taking into account shear histories. Further studies might look at the validation and refining of this model based on CFD simulations while other pipeline elements and constructs might also be considered.

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Appendix A

Commercial granola preparation and properties

This appendix (A.1) describes the production of a commercial granola product for comparative purposes with the lab scale process employed through this study. The process described is typical of granola production on a (small to medium) commercial scale and involves a process similar to the wet granulation process described in Chapter 3 and Chapter 4.

The remainder of the appendix details a number of quality related properties (size, hardness, crispness) for a range of commercially available granola and compares these with the range of values obtained for the granola produced by i) high shear processing and ii) fluidised bed granulation in this study (A.2) and lists the ingredients of each of the commercially available granola products tested (A.3).

A.1: Manufacturing process

All dry ingredients (Table A1) are weighed into a bowl. The ingredients are mixed well in a large mixing bowl to break down all the sugar lumps. Mixing proceeds for approximately 2 minutes. In a separate bowl the water, oil, vanilla extract and salt are whisked until they became cloudy in appearance. The wet ingredients (binder) are then poured into the dry ingredients in the mixing bowl while agitation continues, though the flow rate of binder is not controlled. The mixing time thereafter is not fixed; instead the operation is stopped and visually imparted for product appearance and texture. The mixture is then removed from the bowl and weighed out on trays (2.3 kg per tray) and spread evenly. The period for transferring from the tray to oven is also not fixed.

Table A1: Combination of ingredients for a commercial granola product

Ingredients by weight	**Weight**
Oat	11kg
Brown Sugar	2.7kg
Oat Bran	2.2kg
Sunflower Seeds	1.3kg
Almond Flakes	500g
Sesame Seeds	950g
Coconut	750g
Chopped walnuts	500g
Ingredients by volume	**Volume**
Cold water	1.5 /2 l
Sunflower Oil	2.25l
Vanila extract	40ml
Salt	60ml

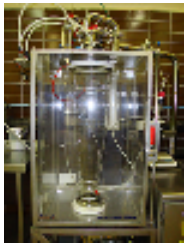
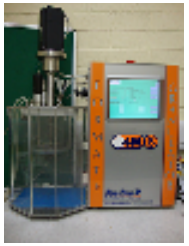
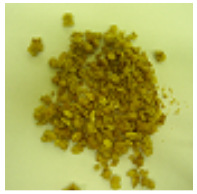
The trays are placed in oven at approximately 160°C for 45 minutes for cooking. The product temperature reaches approximately 135°C during this time. The product is then cooled down to 30–45°C depending on the season and ambient temperatures. When the product has reached 21°C or below it is placed it in the covered white storage bins. Toasted cereal is stored in these bins for a maximum of 24 hours prior to packaging into individual tubs or buckets by hand.

A.2: Commercial available granola properties

A number of commercially available granola products were obtained and measured for median size, hardness and crispness (Table A.2). These values are compared with corresponding values for the granola produced in this study. The granola sold in combination with yoghurt was found to be smaller, (visually) more porous and significantly lower hardness and crispness values than the granola which was sold as a stand alone product.

This type of product compares well with the product produced in the fluidised bed granulator and thus unit would be recommended for a product with these characteristics. The denser, stronger and larger aggregates produced in the high shear granulator would be more in line with the product quality characteristics of the other commercial products which were stand alone, perhaps reflecting the respective manufacturing processes of these products.

Table A2: Comparison of granule size and textural properties for a number of commercial granola with granola produced in this study.

Granola Properties	Marks & Spencer's Granola	Kelkin Granola	Stable Diet Granola	Glenisk Granola (with yogurt)	SuperValu Granola (with yogurt)	Fluidised bed graduation	High shear granulation
							
							
d_{10},mm	5.4	4.9	2.2	1.9	2.9	1.1 – 1.9	4.1 – 11.5
d_{50},mm	9.2	7.2	4.5	3.5	5.8	3.2 – 4.1	6.3 – 15.9
d_{90},mm	15.5	10.1	7.1	6.4	9.8	5.4 – 8.3	8.9 – 19.6
Hardness, N	37 ± 14	70 ± 17	69 ± 18	14 ± 3	19 ± 5	14 – 46	56 – 284
Crispness, dimensionless	183 ± 66	536 ± 110	572 ± 273	68 ± 10	100 ± 15	117 – 316	258 – 755

A.3: List of ingredients for a selection of commercially available granola

1. Marks & Spencer's Granola: Oat flakes (48%), blossom honey (19%), fruit juice infused blackcurrants (6%), fruit juice infused morello cherries (6%), fruit juice infused strawberries (6%), rapeseed oil, concentrated pomegranate juice (2%), pumpkin seed (1%), sesame seeds (1%), sunflower seeds (1%), desiccated Coconut, natural flavouring red berry, pomegranate

2. Kelkin Granola: Granola 80% (oat flakes, wheat flakes, unrefined cane sugar, water, rapeseed oil, coconut) papaya 3% (papaya, sugar, preservative: sulphur dioxide), pineapple 3% (pineapples, sugar, acidity regulator: citric acid: preservative: sulphur dioxide), sunflower seeds 3%, pumpkin seeds 2.5%, flaxseed 2.5%, flaked almond 2%, banana Chips 2% (bananas, coconut oil, sugar, banana flavoring), coconut chips 2%.

3. Stable Diet Granola: Jumbo oat flakes (total oat content 54%), brown cane sugar, sunflower oil, oat bran, sunflower seeds, sesame Seeds, coconut, flaked almonds, chopped walnuts, pure bourbon vanilla extract, sea salt.

4. Glenisk Granola with yogurt: Organic oat flakes (50%), organic honey (14%), organic invert sugar, organic cane sugar, palm oil, organic wheat flakes (7%), organic extruded rice (rice, maize, wheat, sugar), water, organic desiccated coconut , organic skimmed milk powder, organic chopped hazelnuts (1%), sea salt.

5. SuperValu Granola with yogurt: Oats (49%), glucose-fructose syrup, brown sugar, vegetable palm oil, wheat extruded rice (rice flour, wheat malt, sucrose, wheat gluten, dextrose, salt), desiccated coconut, salt.