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How does particle size influence caking in lactose powder?

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ABSTRACT

Particle size distribution (PSD) is known to influence product properties such as flowability and compressibility. When producing crystalline lactose, different steps can affect the PSD of the final powder. The aim of this study was to investigate the influence of PSD on caking and the mechanisms involved. Smaller particles showed higher moisture sorption and a greater caking tendency, measured by dynamic vapor sorption and ring shear testing, respectively. Therefore, moisture sorption isotherms appeared as a valuable tool to predict the effect of PSD on humidity caking, as confirmed by the results of ring shear testing. Controlling the amount of fines, characterized by a higher content of impurities, a larger specific surface area and a broader span of the PSD, was found critical to limit caking. More precisely, both the total surface area and the span of the PSD require close attention as they can significantly influence humidity and mechanical caking.

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1. Introduction

Among the whey-derived ingredients, lactose powder production is currently increasing significantly, with a production of nearly 1.26 million tonnes in 2013 and 1.5 million tonnes expected for 2017 (Affertsholt and Fenger, 2013). The numerous applications include e.g. dairy products, confectionery, baked goods and drugs (Lifran et al., 2000). Lactose powder has been produced since the beginning of the 20th century with the yield in focus (Paterson, 2009). The various applications in the food and pharmaceutical industries have led to different levels of quality requirements (Holsinger, 1988). However, these requirements have become stronger over the past decade with the increasing demand for infant formulae. Lactose is indeed the main ingredient in infant formulae as it provides an important source of carbohydrates to match the composition of human milk. Nowadays, quality is an essential differentiation factor in the highly competitive dairy market. One of the product parameters which lactose producers still strive to control to ensure constant quality is the particle size distribution (PSD) of the powder.

Particle size distribution is a very important characteristic as it governs several product properties (Ortega-Rivas, 2008) such as density, compressibility (Barbosa Canovas et al., 1987), flowability (Juliano and Barbosa-Cánovas, 2010), segregation (Barbosa-Cánovas et al., 1985), rehydration (Gaiani et al., 2007), etc. However, obtaining the right PSD is often a challenge. The crystallization step is crucial for this purpose but by no means straightforward given the complex physicochemical processes involved (Schuck, 2011). Downstream, the PSD can be modified unintentionally (e.g. by particle attrition) or intentionally (e.g. by milling).

Caking, i.e. the unwanted agglomeration of powder particles observed as lumps of various sizes and hardness, is another product property which can be influenced by the PSD of the powder (Rogé and Mathlouthi, 2000). Caking has been an issue in numerous powder handling factories for many years and still leads to significant economic loss. The main caking mechanisms in food powders have recently been reviewed by Carpin et al. (2016). Briefly, the first mechanism, i.e. amorphous caking, involves the creation of viscous flow of amorphous material between particles as a result of an increase in temperature beyond the glass transition temperature (T_g). Moisture can plasticize the material and decrease T_g . Humidity caking is the other mechanism in which moisture plays a critical role. When the relative humidity (RH) of the air surrounding a crystalline material increases, water molecules are indeed adsorbed onto the particle surface and can form liquid bridges between particles. If the RH exceeds the deliquescence relative humidity

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(DRH) of the material, a phase transition from solid to liquid state takes place, leading to a dramatic increase in the amount of moisture adsorbed. The third mechanism, i.e. mechanical caking, is rather an aggravating factor and can dramatically intensify the two above mentioned mechanisms. Inter-particle interactions are indeed increased when particles come closer to each other, for example as a result of compression.

If the PSD is modified, for example due to changes in the crystallization process, how will the caking tendency of the final powder be impacted? This question frequently arises in the powder handling industries and the answer is not straightforward. Mathlouthi and Rogé (2003) investigated the effect of particle size on moisture sorption and caking of sucrose. The higher the amount of fine particles (<250 μm) added to a standard sugar sample, the more sorption was recorded. The particle cohesiveness was found to increase with a decreasing particle size. Fine particles had moderate flow as measured with a Jenike shear cell. This result was explained by their small size and increased hygroscopicity. In addition, Provent et al. (1993) reported that the main cause for caking of a particular pharmaceutical powder was the particle size. By mixing coarse and fine particles in different ratios, they showed that caking was affected by both the total surface area of the powder and the distribution of particle sizes. However, these studies were performed on powders other than lactose, and one could question if the results are still valid for this latter compound. For example, impurities are known to influence the caking behavior of a given powder (Carpin et al., 2017). It is therefore worth investigating the caking mechanisms specific to the lactose powder.

Listiohadi et al. (2005) investigated the effect of milling on the caking behavior of lactose. The hardness of the powder cakes was found to increase for a smaller particle size but moisture sorption was not affected by the particle size. More recently, Modugno et al. (2015) published a study on the influence of the PSD on lactose caking. They sieved the lactose powder to obtain different size classes which were subjected to caking tests. They reported that lactose crystals with a d_{50} smaller than 300 μm could cake easily if the water content was higher than 3%: they explained their result by a greater number of liquid bridges in the small size classes compared to the large size classes. However, this result cannot be applied to dried lactose powders for which the water content is usually below 0.5% (Hourigan et al., 2013). Moreover and as in most studies, the span of the size classes was not taken into account, which can greatly affect the packing density of the powder and thereby the number of liquid bridges. Therefore, even though articles have been published on the subject, it remains difficult to assess how the PSD influences caking of a given powder. The aim of the present study was therefore to investigate the influence of the PSD on caking of crystalline lactose and the mechanisms involved. The expected results should provide a clearer picture of what needs to be measured and controlled in order to limit caking.

2. Materials and methods

2.1. Production of lactose powders on a pilot scale

Decalcified and decolorized ultrafiltered (UF) whey permeate was obtained from Arla Foods Ingredients (Viby J, Denmark). The solids content was raised to 60% in a Centritherm CT2 evaporator (Flavourtech, Griffith, Australia). For lactose crystallization control, the concentrate was cooled in a tank from 79 °C to 11 °C in about 18 h. Then, a Lemitec MD80 laboratory decanter centrifuge (Lemitec GMBH, Berlin, Germany) was used to produce five lactose powders with different washing grades. The slurry was first run through the decanter once without water for a pre-wash (Wash 0). This pre-washed slurry was then mixed with water at different water/

lactose slurry w/w ratios: 1/3 (Wash 0.3), 1/2 (Wash 0.5), 1/1 (Wash 1) and 2/1 (Wash 2). The different washing grades of the prewashed slurry were run through the decanter once more, and then dried in an Anhydro SFD 47 spin flash dryer (SPX Flow Technology, Søborg, Denmark) with an inlet temperature of 105 ± 2 °C and an outlet temperature of 85 ± 3 °C. Finally, the powders were packaged in two layers of plastic bags and a Kraft paper bag before transportation to the analysis laboratory where they were sieved and poured into airtight plastic containers of various sizes to minimize the headspace. The powders were stored at 20 °C prior to analysis.

2.2. Sieving

The lactose powders of different washing grades were sieved through a series of standard testing sieves (80, 160, 250, 355 and 500 μm) stacked in a sieve shaker (Retsch sieve shaker AS 200, Retsch GmbH, Haan, Germany) to separate them in different size fractions. The sieving was performed in batches of 500 g of powder for 5 min at a 1-mm amplitude.

For the investigation of the effect of the span on caking (Section 3.5.), two different size fractions of Wash 0.3 powder were produced by sieving; one by sieving Wash 0.3 through 100 and 355 μm sieves and another one by sieving Wash 0.3 through 160 and 250 μm sieves.

2.3. Measurement of particle size

The PSD of the different size fractions was measured by laser light scattering using a Malvern Mastersizer 2000 equipped with a Scirocco 2000 dry dispersion unit (Malvern Instruments, Worcestershire, UK). The span of the distribution was calculated as

$$\text{Span} = [(d_{90} - d_{10})/d_{50}], \quad (1)$$

where the diameters d_{10} , d_{50} , and d_{90} represent the particle size at 10%, 50%, and 90% in a cumulative size distribution respectively. The specific surface area was calculated from the surface weighted mean d_{32} .

2.4. Chemical composition

Protein and ash content were determined according to the methods described by Schuck et al. (2012). Total nitrogen content determined by Kjeldahl with a 6.38 conversion factor will be designated as protein content. Given the filtration steps in the lactose process, it is, however, unlikely that proteins remain in the final powder. Therefore, impurities formally expressed as protein may more likely be smaller nitrogen containing components such as peptides and amino-acids. Analysis of ash content was carried out in triplicate and the protein content was determined in duplicate.

2.5. Particle morphology — Scanning Electron Microscopy (SEM)

The surface morphology of the lactose samples was examined using a Scanning Electron Microscope (SEM, JEOL JCM-6000 — NeoScope II, Tokyo, Japan) operating at 15 kV. Samples were mounted on an aluminum stub and coated with a thin layer of gold (JEOL JFC-1300 auto fine coater) prior to analysis. The photomicrographs were taken at $\times 400$ magnification.

2.6. Moisture sorption measurements

Sorption isotherms of powders were obtained with a Dynamic Vapor Sorption (DVS) Advantage (Surface Measurement Systems

Ltd., London, UK) equipped with a Cahn microbalance. The experiments were carried out in duplicate at a constant temperature (25 °C) using a nitrogen flow rate of 200 standard cm³ min⁻¹. Approximately 40 mg of powder was subjected to ramping of RH from 0% to 95% in 10% RH steps with water as solvent. Equilibrium was considered to be reached if the rate of change in mass was less than 0.0002%.min⁻¹.

2.7. Quantification of caking

Caking, also known as time consolidation, was measured with a ring shear tester (RST-XS, Schulze-Schüttgutmesstechnik, Wolfenbüttel, Germany). The measurement procedure has been described in detail by Schulze (2008). First, the yield locus of each powder was measured for a normal stress at preshear σ_{pre} corresponding to a consolidation stress σ_c of approx. 9.3 kPa. This value was chosen to simulate powder consolidation at the bottom of a flexible intermediate bulk container (FIBC), also called Big Bag. Three points of incipient flow were used to draw the yield locus. Once the initial yield locus was established, the powder was pre-conditioned using the same σ_{pre} as above and stored at 20 °C and 60% RH for four days. A temperature and humidity data logger was used to check the storage conditions. Loads of 2 kg (corresponding to a consolidation stress of 9.3 kPa) were applied to the samples for the duration of storage. After storage, the samples were sheared to obtain the time yield locus. From this, the unconfined yield strength σ_1 and the ratio of σ_c to σ_1 , called *ffc*, were identified. The larger the *ffc* was, the better the powder flowed. By definition, a sample with an *ffc* lower than 1 was considered to be caked (Jenike, 1964). For each powder, the *ffc* was measured in triplicate, with the exception of the investigation of the effect of the span on caking (Section 3.5.), where the *ffc* was measured in duplicate.

2.8. Solid-state Nuclear Magnetic Resonance (NMR)

¹³C NMR spectra were obtained using proton decoupling, magic angle spinning (MAS) and cross polarization (CP). The spectra were recorded on a Bruker Avance I WB 300 MHz (7T) instrument (Bruker, Billerica, USA) at ambient temperature according to the method described by Gustafsson et al. (1998), with the following parameters: spinning rate 5 kHz, contact time 2 ms, acquisition time 147 ms, sweep widths 2190 ppm and delay between pulses of 3 s. For each spectrum, about 150,000 transients were cumulated with 49 k data points. The spectra were referenced to trimethylsilane (TMS).

3. Results and discussion

3.1. Particle size distribution (PSD)

The division between the different size classes obtained by sieving the two extreme washing grades (Wash 0 and Wash 2) was noticeably different (Fig. 1). Particles were generally bigger in Wash 0, which can be explained by the presence of agglomerates, visible on the sieve in the fraction above 500 μm . These agglomerates may be structured by liquid bridges, favored by the presence of fines ($x < 80 \mu\text{m}$); the dissolution of the fines upon washing, as indicated by the low amount of remaining fines in Wash 2, led to a narrowed PSD with very few particles over 355 μm . In line with these observations, the general trend for all washing grades was that a decreasing washing grade led to bigger particles (data not shown). Thus, apart from crystallization (Schuck, 2011), washing is another important processing step that must be controlled in order to obtain a given PSD. Moreover, the particle size should not be neglected when investigating the effect of the washing grade on a

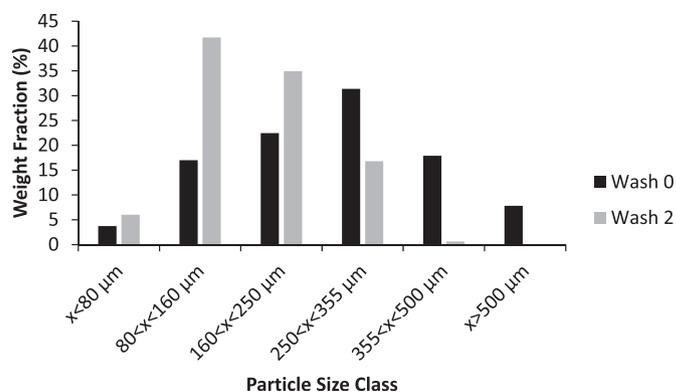


Fig. 1. Particle size distributions of Wash 0 and Wash 2 lactose powders sieved to separate 80, 160, 250, 355 and 500 μm fractions.

given powder parameter, such as for example caking.

3.2. Chemical composition

The different particle size fractions were analyzed separately for their impurity content. Impurities consisted in non-lactose components, mainly minerals and nitrogen containing components expressed as proteins, remaining in the powder after crystallization and washing. The fines ($x < 80 \mu\text{m}$) showed a higher content of proteins and ash than the bigger particles in Wash 0, Wash 1 and Wash 2 (Fig. 2). Wash 0 presented a minimum content of impurities in the size fraction $250 < x < 355 \mu\text{m}$; above this size fraction, increasing the particle size resulted in a higher content of impurities (Fig. 2). In Wash 1 and Wash 2, the general trend was a decrease in the impurity content with a larger particle size. Moreover, as expected, a smaller specific surface area was measured for larger particles (Fig. 2).

In general, and with the exception of the larger size fractions of Wash 0, the evolution of impurity content with the particle size followed the one of the specific surface area. This indicates that impurities are likely to be located mostly on the surface of the particles. In Wash 0, the agglomeration of impurity containing fines together or on a larger crystal may explain the greater amount of impurities in the size fractions $x > 355 \mu\text{m}$. As these agglomerates were most likely only present in the lowest washing grade Wash 0 (Fig. 1), they may explain the different evolution of impurity content with particle size for Wash 0 compared to the other washing grades (Fig. 2). The results indicate that the fine fraction should be given attention as it can greatly influence the level of impurities in the powder. Crystallization is an important processing step to control the amount of small crystals. However, small particles are often fragments of bigger crystals which are damaged during processing steps (Mathlouthi and Rogé, 2003) such as rotary drying in a spin flash dryer used in the present study. Considering the industrial process for lactose production, air transport between silos can for example be harsh to the particles which may be altered by attrition. In addition, milling is obviously a processing step which affects the particle size distribution of the final product (Hourigan et al., 2013).

3.3. Aspect of the fines

Microscopic observations of the fine particles can help identify whether these are whole but small crystals or fragments of bigger crystals. Scanning electron microscopes of Wash 0 and Wash 2 powders revealed differences in the agglomeration propensity of the fines (Fig. 3A and B). Wash 0 showed agglomerated particles

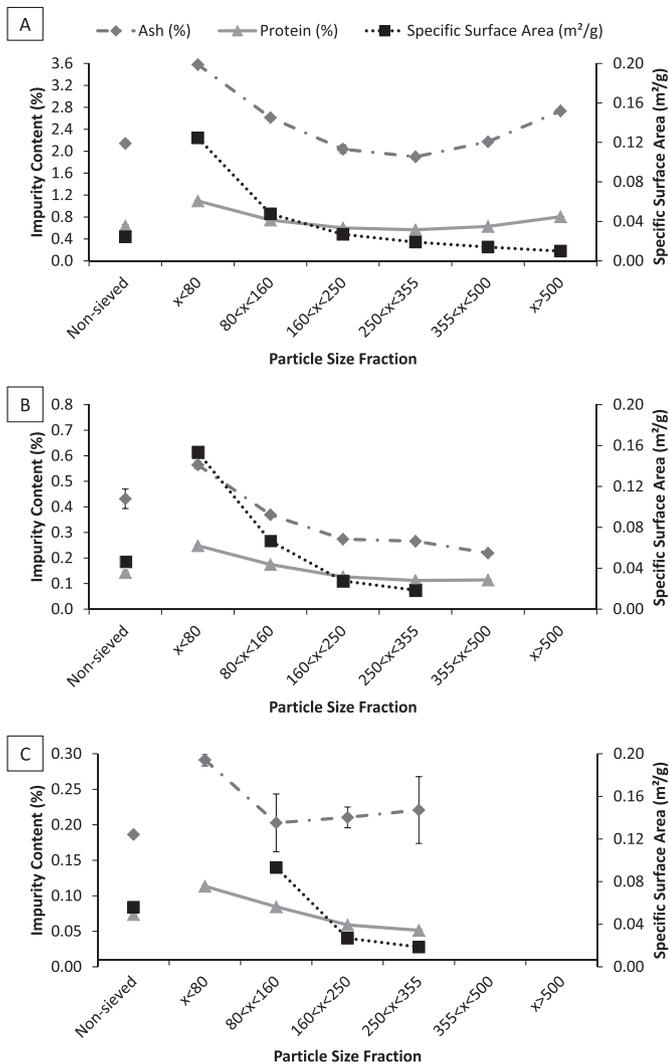


Fig. 2. Impurity content (either protein or ash content; left axis) and specific surface area (right axis) of lactose powders as a function of the particle size class: (A) Wash 0; (B) Wash 1; (C) Wash 2. Results are given as average \pm standard deviation (represented by the error bars). Number of repetitions = 4 for specific surface area, 3 for ash, and 2 for proteins.

(Fig. 3A) as expected from the particle size distribution (Fig. 1). In contrast, small particles in Wash 2 were a mix of well-formed crystals and agglomerates. Very small fragments were also present (Fig. 3B). For both powders, the agglomerated fragments were

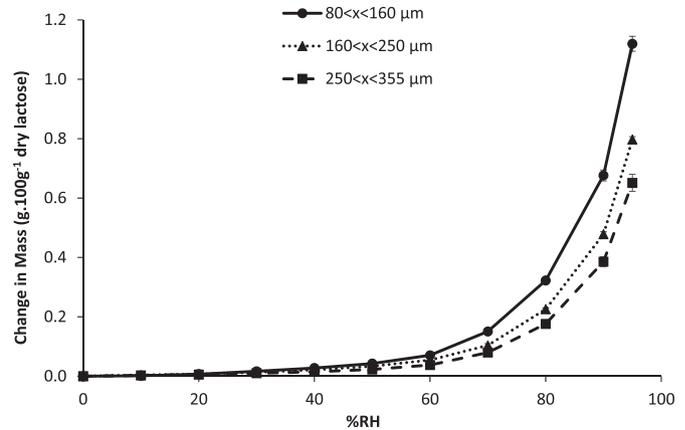


Fig. 4. Moisture sorption isotherms of lactose powder Wash 2 for three different particle size fractions: $80 < x < 160 \mu\text{m}$: circles; $160 < x < 250 \mu\text{m}$: triangles; $250 < x < 355 \mu\text{m}$: squares.

likely to originate from the surface of bigger crystals. Assuming that impurities were mainly concentrated on the crystal surface, these fragments thereby increased the overall impurity content of the fine fraction. A suitable technique which could distinguish between the composition of the surface and the inner part of a lactose crystal could confirm this hypothesis. For example, the surface composition of several milk powders, characterized by X-ray photoelectron spectroscopy (XPS), was found to differ from their bulk composition in lactose, proteins and fat (Murrieta-Pazos et al., 2012).

3.4. Moisture sorption

The difference in moisture sorption behavior depending on the size fraction was significant (Fig. 4). The final amount of water sorbed (at 95% RH) indeed increased for the smaller particles ($80 < x < 160 \mu\text{m}$) compared to the medium ($160 < x < 250 \mu\text{m}$) and bigger ($250 < x < 355 \mu\text{m}$) particles by 40% and 72%, respectively.

When the ambient RH increases, three mechanisms of water-solid interactions can be considered for the crystalline lactose-water system, i.e. adsorption of water molecules on crystal surface, capillary condensation and deliquescence (Mauer and Taylor, 2010). As moisture sorption occurs primarily on the surface, the larger specific surface area associated with smaller particles (Fig. 2) is consistent with a higher sorption behavior. Likewise, such an increase in moisture sorption was measured for smaller particles of glass beads (Stoklosa et al., 2012) and sucrose (Mathlouthi and Rogé, 2003) compared to bigger counterparts.

Moreover, capillary condensation occurs in pores between

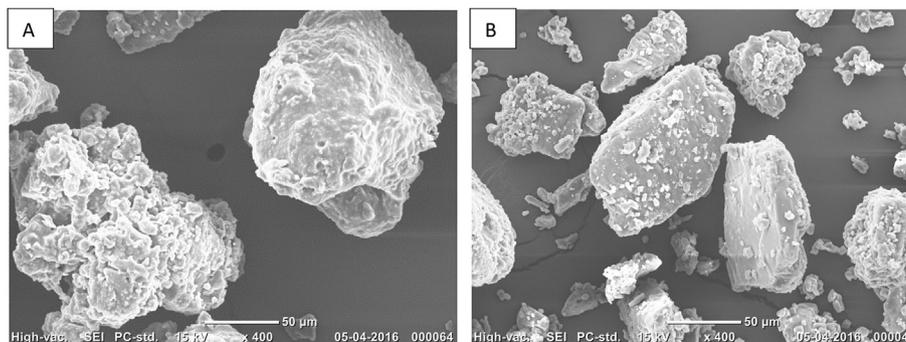


Fig. 3. SEM images showing small particles of different washing grade lactose powders: (A) Wash 0; (B) Wash 2.

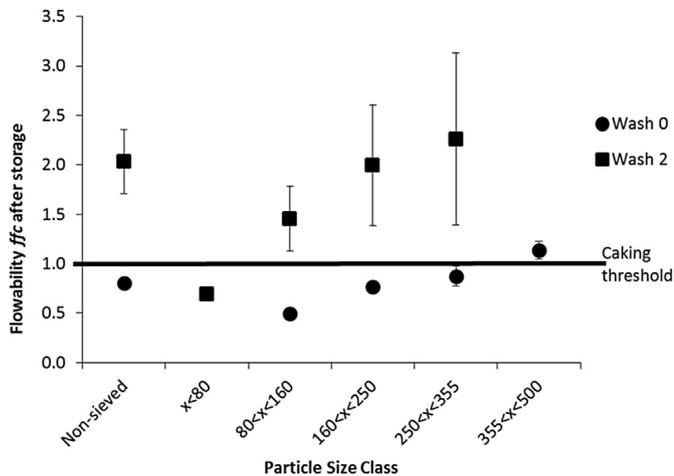


Fig. 5. Flowability ffc of lactose powders Wash 0 (circles) and Wash 2 (squares) as a function of particle size class after storage for four days at 60% RH and 20 °C and under a consolidation stress of 9.3 kPa.

adjacent particles. Using the Kelvin equation, it is possible to predict the critical capillary radius below which all capillaries will be full of water at a given water activity and temperature (Billings et al., 2006). The size distribution of the pores is expected to depend both on the average particle size as smaller particles give smaller pores, but also on the span of the particle size distribution as powder with a larger span can be compacted to a higher packing density and thus a structure with smaller pores (Bronlund and Paterson, 2004). The PSD of the different size fractions of Wash 2 powder showed a broader span value (Eq. (1)) for the fraction $80 < x < 160 \mu\text{m}$ compared to the fractions $160 < x < 250 \mu\text{m}$ and $250 < x < 355 \mu\text{m}$: 1.56, 0.88 and 0.73 respectively. This broader span certainly contributes to the higher moisture sorption behavior of the size class $80 < x < 160 \mu\text{m}$.

Finally, pure crystalline lactose is characterized by a very low hygroscopicity, with a deliquescence relative humidity (DRH) value of 95% (Salameh et al., 2006). However, for water-soluble crystalline solids it has been shown that impurities can increase moisture sorption below DRH and reduce the DRH value (Guerrieri et al., 2007; Tereshchenko, 2015). The higher content of impurities in the smaller particles (Fig. 2), especially if these impurities are located on the surface, is thus likely to influence the shape of the sorption isotherm.

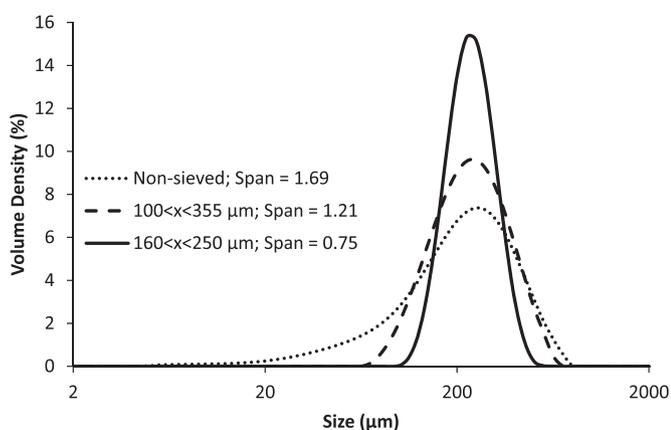


Fig. 6. Particle size distributions of different size fractions (non-sieved, $100 < x < 355 \mu\text{m}$ and $160 < x < 250 \mu\text{m}$) of lactose powder Wash 0.3.

In summary, the observation of increasing moisture sorption for the smaller size fraction can be explained by a larger surface area, a higher level of impurities and smaller capillaries due to a lower voidage fraction and due to a broader span of the size distribution.

3.5. Caking tendency

The caking tendency of the different size fractions of the least (Wash 0) and most washed (Wash 2) powders from the pilot trial was measured with a ring shear tester (Fig. 5). For both powders, it was clear that the smaller particles showed poorer flowability after storage (i.e. lower ffc value) and thus a higher caking tendency. It was even possible to exceed the caking threshold by selecting the right particle size fraction and thereby prevent the powder from caking under the chosen storage conditions. For a given powder, the non-sieved fraction had an intermediate caking tendency compared to the sieved fractions. The results for the caking tendency of the different size fractions of Wash 1 showed the same trend (data not shown).

The results for the caking measurements were consistent with the moisture sorption isotherms showing higher sorption for smaller particles. Humidity caking is therefore a critical mechanism to consider for caking in lactose powder under the selected conditions. Similarly, Paterson and Bronlund (2009) also concluded on a capillary condensation mechanism leading to significant liquid bridging in crystalline lactose powder.

Moreover, sugar dust from a sugar factory has been reported to contain amorphous material originating from the surface of sucrose crystals dried at high temperatures in a short time (Mathlouthi and Rogé, 2003). These fines were shown to adsorb significantly more moisture compared to microcrystals of the same size produced with a special crystallization process. In the present study, different size fractions of Wash 0 powder were analyzed by solid-state NMR but the amount of amorphous material was below detection limit (data not shown). Thus, amorphous material in the fines is probably present at too low levels to trigger amorphous caking, i.e. viscous flow of material to contact points. However, even in low levels, amorphous material can still promote moisture sorption and thus contribute to caking. Fragments of bigger particles surface among the fines, as revealed by SEM images (Fig. 3), should therefore be closely controlled as they can contain amorphous material.

The third mechanism reported in the literature for caking in food powders is mechanical caking (Carpin et al., 2016). This mechanism is highly influenced by the PSD of the powder through the number of possible contact points. As smaller particles are known to create more contact points than the bigger counterparts for the same volume of powder, mechanical caking takes part in the greater caking tendency of the smaller particles (Fig. 5). However, Bronlund and Paterson (2004) measured more moisture sorption and thereby a greater possibility of caking in unmilled lactose compared to the milled powder; they explained this difference by a wider PSD in unmilled lactose. The d_{50} , i.e. the mean particle size of the distribution, is therefore not the only parameter that should be taken into consideration when assessing the effect of a change in the PSD on caking.

In order to investigate the influence of the span, three different size fractions ($160 < x < 250 \mu\text{m}$, $100 < x < 355 \mu\text{m}$ and non-sieved) of the same lactose powder with very similar d_{50} values ($243 \pm 11 \mu\text{m}$) were subjected to caking measurements. The PSD of the three size fractions was measured to verify that they had different span values but similar d_{50} values (Fig. 6). A lactose powder with an intermediate washing grade (i.e. Wash 0.3) was used for these time consolidation experiments. This powder was chosen to allow moderate to strong caking to take place in the powder and thereby a differentiation of the samples while avoiding

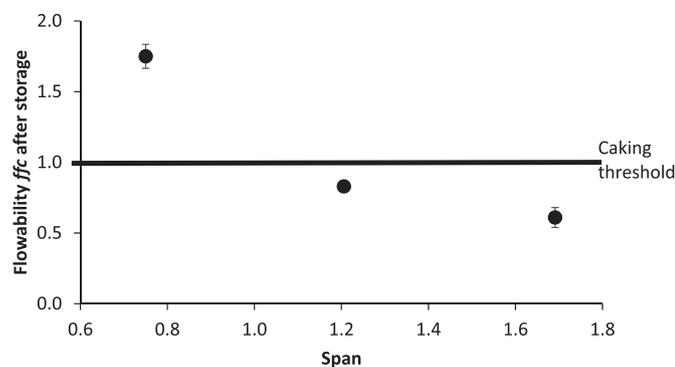


Fig. 7. Flowability *ffc* of different size fractions of lactose powder Wash 0.3 as a function of the span of the particle size distribution. The size fractions were $160 < x < 250 \mu\text{m}$, $100 < x < 355 \mu\text{m}$ and non-sieved, with the span being 0.75, 1.21 and 1.69 respectively. The measured *d* 50 were $249 \mu\text{m}$, $249 \mu\text{m}$ and $230 \mu\text{m}$ respectively. The *ffc* was measured after storage for four days at 60% RH and 20°C and under a consolidation stress of 9.3 kPa.

a strong caking of all samples.

Fig. 7 shows that for powders with similar *d* 50, a broader span of the size distribution clearly led to a stronger caking tendency, in agreement with the observations of Bronlund and Paterson (2004). The numerous contact points associated with a wider PSD allow more interactions between particles and thereby increase the possibility of mechanical caking and humidity caking by capillary condensation, as explained in Section 3.4.

The PSD of food powders can be difficult to control in industrial settings because of the many processing steps which can affect it. However, the results presented in this study clearly show the importance of controlling the PSD to be able to prevent caking in the final product. In order to assess the effect of changes in PSD on the caking tendency of the product, it is crucial to consider both the specific surface area of the powder and the span of the size distribution as they influence caking in different ways. Indeed, Provent et al. (1993) reported that mixing coarse and fine particles in a powder might lead to critical concentrations of fines for which the risk of caking was greatest. It might therefore be helpful to shift the size distribution slightly towards bigger particles in order to remove part of the fines. As shown in this study, the presence of more fines in a powder leads to a larger specific surface area and more impurities, thus increasing the amount of water adsorbed and decreasing the DRH value. Furthermore, a broader span promotes capillary condensation and mechanical caking. Given the importance of the humidity caking mechanism in a crystalline powder, the moisture sorption isotherm is definitely a valuable tool to help predicting the effect of the PSD on caking.

4. Conclusions

Smaller particles showed higher moisture sorption and a greater caking tendency compared to bigger counterparts. The larger specific surface area and higher content of impurities associated with the smaller particles increase the possibility of humidity caking. It is therefore essential to control the amount of fines, which was found to depend on the washing grade. More precisely, attention should be paid to both the span of the PSD and the specific surface area of the powder as these two parameters influence mechanical caking through the number of contact points and humidity caking through moisture sorption, capillary condensation and deliquescence.

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