Effects and Control of Humidity and Particle Mixing in Fluid-Bed Granulation

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The novel technique of spraying binder liquid in pulses of short duration on a bubbling fluidized bed was used to study the effect liquid distribution, mixing, and relative humidity has on granule growth. Two important mixing zones in the fluid-bed granulation process are identified. First, the wetting zone, where the spraying rate relative to the surface renewal rate determines the wetting of the granules and powder. The surface renewal has to be sufficient to prevent overwetting of the surface, which would result in defluidization of the bed. Second, the bulk zone, where the turnover rate of granules relative to the drying rate may determine the secondary growth rate of granules. The granule growth is influenced by the adsorption of moisture on the fluidized particles governed by the relative humidity of the interstitial gas, due to enhancement of interparticle forces and reduces the mixing intensity. Spraying with pulses has a great potential for controlling the granulation process.

Introduction

The fluid-bed agglomeration process is a "wet" agglomeration process, where binder liquid is sprayed upon a bed of fluidizing particles. The particles are wetted by the binder liquid and bound together by liquid bridges to form an agglomerate. During the process the wet agglomerate dries as liquid evaporates and binder solidifies at the contact points between the particles, giving the agglomerate its strength.

Fluid-bed agglomeration is widely used in industry for manufacturing a range of granular products. The granular products made should be well defined, of constant quality, and independent of their batch number. Unfortunately, these requirements cannot always be fulfilled. Sometimes the quality of the product can be improved by size classification, for instance, by sieving. The disadvantage of this is the large recycle flow, reprocessing of rejected product, and increase of size of the apparatus, which is costly. Therefore it would be most desirable to achieve a well-defined product without using classification methods, that is, "right first time." This can only be accomplished with a good control of the granulation process.

Control of the fluid-bed agglomeration process is difficult. Wetting, drying, and mixing of particles all take place simultaneously in the bed. The different variables affect each other and are therefore difficult to influence independently. Thus it is necessary to understand the important mechanisms involved and their relation to each other in order to be able to control or develop the granulation process effectively. Much research has been concentrating on methods to control the granulation during the process (see Gore, 1990; Nienow, 1993; Scheafer and Worts, 1978a,b; and Watano et al., 1996). Several authors used the bed humidity, moisture content, or the bed temperature as control parameter for the process (Scheafer and Worts, 1978a,b; Watano et al., 1996). These control parameters are all related to the liquid concentration in the fluid bed. The liquid concentration determines the adhesion between the particles and thus the agglomeration.

The binder liquid is distributed onto the fluid bed by means of a spraying nozzle. The nozzle determines the droplet-size distribution, the spray rate, and spray pattern upon the fluid-

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bed surface. The nozzle can be characterized independently of the agglomeration process itself. Generally, two-fluid nozzles with an extended liquid insert are used, because the droplet size can be varied independently from the liquid flow and the chance of clogging is reduced by the insert (Nienow, 1993). Some authors studied the effect of the liquid nozzle on the formation of granules (Schaafsma et al., 1998a; Scheafer and Worts, 1977, 1978a; Waldie, 1991). They found a relation between droplet size and granule size.

The distribution of liquid throughout the bed is determined by mixing of the wetted particles. The particle mixing in the fluid bed is difficult to characterize. The mixing intensity changes during the agglomeration process, due to the changing bed humidity and the changing size and density of the fluidized particles. Mixing influences the wetting of powder in the spray zone and the drying of granules in the bed. We suggest that the mixing intensity is an important factor, which determines the granule growth, and should not be underrated.

To gain a better understanding of the influence of liquid distribution and bed mixing on granule growth we applied a novel experimental technique by top spraying liquid in pulses of short duration onto the fluid bed to study the dynamic behavior of the agglomeration process.

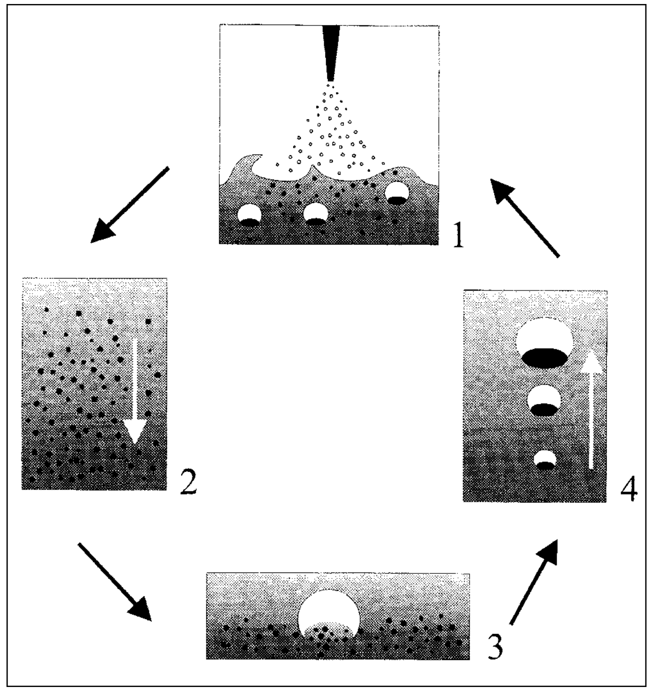

Figure 1. Particle transport in fluid bed granulation process.

(1) Forming of granules at bed surface; (2) segregation of granules; (3) bubble and wake forming near distributor plate; (4) transport of bubble and wake to bed surface.

Mixing, Liquid Distribution, and Drying in a Bubbling Fluid-Bed Agglomerator

The main contribution to particle transport in a bubbling fluidized bed is an upward transport of material in the wake of a bubble to the top of the bed with a consequent downwards motion in the bulk emulsion (Gibilaro and Rowe, 1974; Hoffmann et al., 1993; Nienow and Chiba, 1981; Rowe and Partridge, 1965). This principle is depicted in Figure 1 for granules formed at the bed surface.

The bubble volume determines the wake volume and therefore the particle transport. The void/wake ratio decreases with increasing bubble size. The total flow of gas in bubble voids is related to the minimum fluidization velocity (U_{mf}) with a simple relation referred to as the n-type two-phase theory (Grace and Clift, 1974; Toomey and Johnstone, 1952).

$$U_e = U_G - U_{mf}(1 + nf_b). \quad (1)$$

Here U_G is the fluidization velocity, U_{mf} is the minimum fluidization velocity, f_b is the volume fraction of the bed occupied by bubbles, n is a positive number, and U_e is the excess gas velocity. The part of the gas that is not necessary for fluidization is called the excess gas velocity (U_e), and is approximately equal to the total bubble void volume per second per unit cross-section area of the bed. In ideal two-phase flow theory n is zero; in practice n varies slightly with U_G, bed height, and particles used, and is therefore a difficult parameter when scaling up.

During the agglomeration process U_{mf} will increase as the granule diameter increases. As a result the excess gas velocity will decrease and so will the overall bed mixing.

Another aspect that influences the mixing is the humidity in the emulsion phase of the fluid bed. Hartholt (1996) found that, when the relative humidity (rH) of a fluid bed containing glass beads increases above a value of 60%, U_{mf} increases. This is due to the fact that the adsorption of water molecules on the particle surface increases at higher relative humidity. The adsorbed liquid layer can greatly enhance the interparticle forces and therefore the bed voidage at incipient fluidization by formation of liquid bridges between the particles. This starts to become significant after approximately two to three monolayers of water molecules (Fisher and Israelachvili, 1981). A decrease of the interparticle forces with increasing rH is also possible since the enhanced conductivity can decrease electrostatic forces. The influence of rH on the minimum fluidization velocity depends on the particle size, surface roughness, and material properties. The increase in U_{mf} results in decreased mixing. Eventually, the bed material becomes so cohesive that fluidization becomes impossible, resulting in channel formation and collapse of the fluid bed. Defluidization depends on the gas velocity as well: the gas velocity can be increased to overcome the larger interparticle forces. Considering the aspects mentioned earlier, it is necessary to control the relative humidity of the bed, since above a critical value it will influence the mixing behavior.

The axial distribution of the granules in the bed is also important for the process. Particle mixing and segregation in fluidized beds containing a mixture has been studied in several articles (e.g., Gibilaro and Rowe, 1974; Hoffmann and Romp, 1991; Naimer et al., 1982). We distinguish between two different zones in the fluid-bed granulator where different stages in the granulation process take place, when spraying top-down. The first zone is at the surface, the wetting zone, where the liquid concentration is high and powder is wetted by the binder liquid droplets. The second zone is the bulk-mixing of the agglomerates and primary powder in the bed, where the liquid concentration is lower and granules are dried.

In the wetting zone, a granule may be formed by one droplet or by several droplets, depending on the droplet size distribution and the residence time of a granule at the bed surface. The droplet size distribution, the spray density, and the spray rate determine, together with the surface renewal rate, the granule growth. In the most unfavorable situation, the spray surface will be overwetted, resulting in the formation of large lumps and defluidization of the bed.

Now considering the mixing in the bulk, a wet granule will segregate in the bed while drying and may reappear at the bed surface after a period of time, "turnover time." A granule can be dried or still partially wet when it appears at the spray surface, depending on the ratio between the turnover and drying times. This may be important for the agglomerate growth. If a granule is dry when reappearing at the spray surface, it will suck the binder liquid into its pores when it is rewetted. This uptake of liquid is a very fast process if the particle surface is well wettable by the liquid, as described by Schaafsma et al. (1998b) and results in less liquid available for binding other particles at the surface of the granule. If the granule is still wet when reappearing at the surface, less liquid is sucked into it when rewetted, resulting in more liquid available at the surface for further granulation. Smith and Nienow (1983) showed that the existence of intraparticle porosity slowed down or even prevented the granulation.

In order to study the effect of mixing and liquid distribution in the fluid-bed agglomeration process, we can spray liquid in pulses of short duration, for instance, 3-s spraying, with a lag time of 60 s, and repeat this during the whole process. The possible effects of this operation are shown in Table 1. By spraying for a short duration we achieve a characteristic time constant (the spray time relative to the lag or drying time), which allows us to study the effects on drying and mixing at the surface and in the bulk of the fluid bed.

As described in Table 1, pulse spray experiments can determine important time scales for mixing at the spray surface or in the bulk zone by varying the duration of the liquid spray time. The drying time should depend on the relative humidity, which should not exceed a certain level. If overwetting occurs, this will definitely influence the mixing in the bed.

Materials and Methods

A batch of 2.0 kg of α-lactose monohydrate 110 mesh powder was agglomerated using water containing 8% polyvinylpyrolidone [PVP, molecular weight (MW) ~ 24,500]. The U_{mf} of Lactose 110 mesh was measured as a function of the relative humidity as described by Hartholt (1996). Water adsorption on the surface of α-lactose monohydrate 110 mesh was measured by dynamic liquid vapor adsorption (DVS 1000, England). Nitrogen adsorption on the surface of α-lactose monohydrate 110 mesh was measured in a Quantasorp gas-adsorption apparatus (Quantachrome, U.S.A.). The specific surface area of lactose was determined with both water and nitrogen by the BET method.

Equipment

The experimental setup is shown in Figure 2. The product chamber is a conical cylinder made from stainless steel. The air distributor is a bronze sintered plate. A cyclone was used to clean the process air from lactose fines. It collected less than 2% of the total batch during a run.

The relative humidity and temperature were measured beneath the air distributor plate and above the fluid bed with an electronic humidity transmitter (Steinecker, Germany). A computer monitored the humidity and temperature on-line during the process. A two-fluid nozzle (Schlick 970-7-1, Germany) with an external liquid insert and pneumatic control was used for spraying droplets on the fluid bed. The spraying and lag time were controlled automatically by a computer, which directed the pneumatic system of the nozzle. The droplet volume distribution of the nozzle was measured by laser diffraction (Sympatec, Germany). The nozzle was placed 0.2 meter above the fluid-bed surface.

Operating conditions

The spray-pulse duration and lag time between spray pulses are listed in Table 2. After several pulses of spraying, the product was collected and classified by sieving. After sieving

Table 1. Expected Effect of Spray Pulse Duration on the Granule Size

Spray Rate	Fast Surface Renewal	Slow Surface Renewal
High spray rate	A higher droplet density at the spray surface may result in larger granules if sprayed with longer pulse durations.	Collapse of bed surface by overwetting and lump formation at spray zone.
Low spray rate	No effects of spray pulse duration on granule size, because granules are directly removed from the spray zone.	A long residence time in the spray zone may result in larger granules if sprayed with longer pulse durations.
Drying Rate	**Fast Bulk Mixing**	**Slow Bulk Mixing**
Fast drying	Granules are dry when rewetted at spray surface. No effects of spray pulse duration on granule size.	Granules are dry when rewetted at spray surface. No effects of spray pulse duration on granule size.
Slow drying	Granules may have different moisture contents because lag time between the spray pulses could be of the order of the drying time of a granule, resulting in smaller granules for longer lag times.	Granules may have different moisture contents because mixing time could be of the order of the spray pulse duration, resulting in larger granules for longer spray time durations.

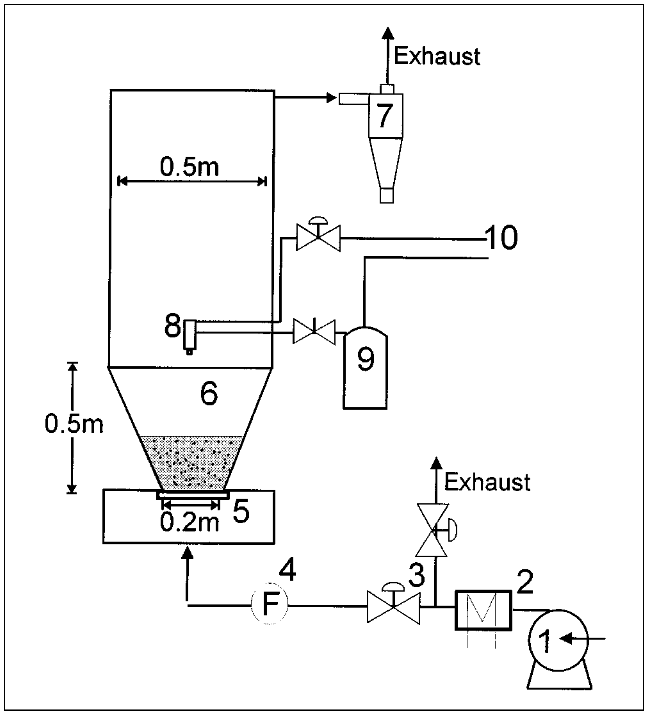

Figure 2. Experimental setup.

(1) Roots blower; (2) heater; (3) control valves; (4) mass flowmeter; (5) wind box; (6) product chamber; (7) cyclone; (8) two-fluid nozzle; (9) liquid pressure vessel; (10) air supply.

the whole product was replaced in the fluid bed and the procedure was repeated. The pulse-spraying experiments stopped when the fluid bed collapsed by overwetting of the particles.

Four fluidization velocities of 26, 32, 40 and 48 cm/s were chosen, all giving good mixing of the primary particles. A high fluidization velocity would have resulted in entrainment of particles, and too low a fluidization velocity would have resulted in a fast collapse of the fluid bed by insufficient mixing. The fluidization velocity is determined at the distributor plate, as the bed height is not constant during granulation and varies with the fluidization velocity as well. The spray rate was 42 g/min. We sprayed with two droplet sizes with an average volume diameter (d_{50}) of 99 μm (d_{10} 18 μm, d_{90} 255 μm) and of 16 μm (d_{10} 4 μm, d_{90} 65 μm). The inlet temperature was set to 40 or 60°C. The rH underneath the distributor plate varied from 6% to 15%, depending on the ambient conditions. The relative humidity in the fluid bed varied, depending on the spraying and lag time, during the process between 20% and 95%. An example of the change of the rH during the granulation process is shown in Figure 3 for a spray pulse of 15 and 30 s at a fluidization velocity of 48 cm/s.

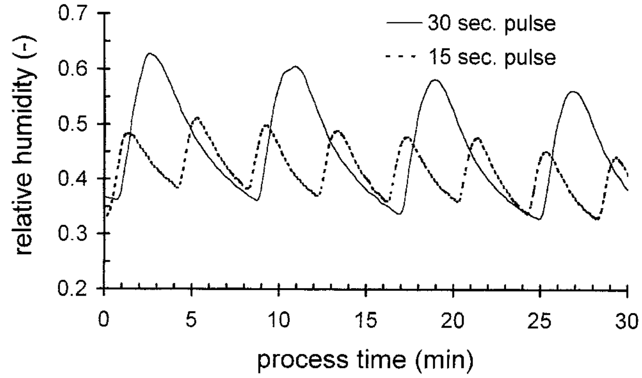

Figure 3. Example effects of spray pulse duration on relative humidity.

Table 2. Operating Conditions

Variation of Spray Time						
Inlet temp. 40°C	Spraying time (s)	3	5	10	15	30
	Lag time (s)	60	120	180	240	480
Variation of Drying Time						
Inlet temp. 60°C	Spraying time (s)	3	3	3	3	3
	Lag time (s)	9	15	30	45	60
Variation of Fluidization Velocities						
Inlet temp. 60°C	Spraying time (s)	3				
	Lag time (s)	45				

Results and Discussion

Influence of the relative humidity on fluidization

To examine the effect of the rH on the lactose powder, we performed a liquid adsorption and desorption experiment (Figure 4). The specific surface measured with the BET-equation using water as adsorbent was found to be 2,102 cm^2/g, assuming an effective area for a water molecule of 12.5 $Å^2$ (Kontny and Zografi, 1995). This can be compared with the surface calculated from N_2 adsorption, which was 1694 cm^2/g. The difference between these two areas can be explained. Tricehurst et al. (1996) found signs of dehydration of α-lactose monohydrate at its surface when it was dried by dry air. This observation was supported by Clydesdale et al. (1997) by modeling of α-lactose monohydrate crystal morphology, which indicated that water had a space-filling role rather than strong binding in the crystal structure. Water adsorption after drying would occupy these dehydrated areas,

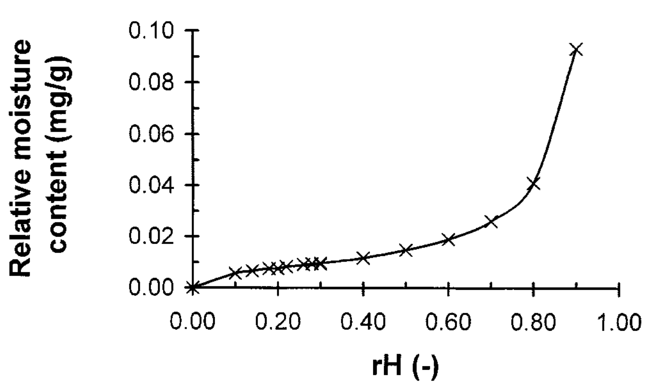

Figure 4. Adsorbed mass as function of rH.

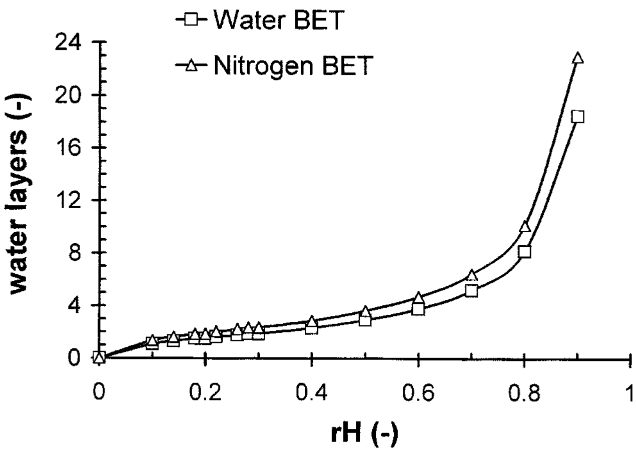

Figure 5. Number of liquid layers as a function of rH.
Calculated according to surface area measured with nitrogen BET and water BET.

whereas nitrogen is unable to reach those areas because of its larger size. Another explanation for the differences measured in surface area could be the absorption of water by protein pollution at the surface of lactose.

The number of liquid layers at the surface of a lactose particle can be calculated and are shown in Figure 5. We calculated the number of liquid layers for both areas determined by nitrogen and water adsorption. The true number of liquid layers should lie somewhere in between. The literature (Hartholt et al., 1995) shows that we can expect humidity to have an effect on the mixing behavior of the lactose, starting from about 60% rH. This can be seen to correspond to approximately four monolayers of water.

The U_{mf} of lactose is influenced by the relative humidity, as shown in Figure 6. The change of U_{mf} sets in above 45% rH, which corresponds to approximately two to three monolayers of water, which is the point at which liquid bridges can visually be detected according to Fisher and Israelachvili (1981). The influence of the humidity could be explained by the fact that at contact points between particles liquid can become mobile and form a capillary bridge, and thereby increase the minimum fluidization velocity. Above 75% rH we could not determine the minimum fluidization velocity due to severe channeling; this value corresponds to approximately to six to eight monolayers of water. The stronger binding between particles due to the liquid bridges can still be broken if a very high gas velocity is used to fluidize the particles. However, when the humidity in the emulsion phase is high, there is a risk that some regions in the fluid bed are insufficiently mixed and that nonhomogeneous mixing will result in strong deviations in the size distribution of granules by partial overwetting in some regions of the fluid bed.

Influence of pulse duration on granule growth

The mean size of lactose granules was measured during the pulse-spray experiments and is depicted in Figure 7. More than 90% of the lactose was agglomerated after spraying approximately 100-g binder liquid; some granules consisted of only two primary particles. All particles were mixed and no significant segregation was found.

The growth followed the same curve for all pulse durations and the bed collapsed during the spraying period at nearly the same mean granule size ($D_{50} \sim 390\ \mu$m). As the granules grew larger, U_{mf} increased and the mixing of granules at the spray surface decreased. In the end, the fast spray rate relative to the surface renewal caused overwetting of the bed surface, resulting in formation of large lumps and defluidization.

The mixing of the fluid bed can be roughly estimated. The air in the bubble phase roughly depends on U_e, according to the two-phase theory in its simplest form (Toomey and Johnstone, 1952). The wake fraction (f_w) was found to be one-third for large bubbles (Hoffmann et al., 1993; Nienow and Chiba, 1981; Smith and Nienow, 1983). The mixing can be determined and depends on the bed height, since U_g varies with the height of the conical-shaped bed. The bulk mixing intensity can be described as a turnover frequency, which is the wake volume transported to the bed surface in one second divided by the bed volume:

$$I_m = f_w U_e A \frac{\rho_p (1 - \epsilon_{em})}{M_{\text{bed}}} \tag{2}$$

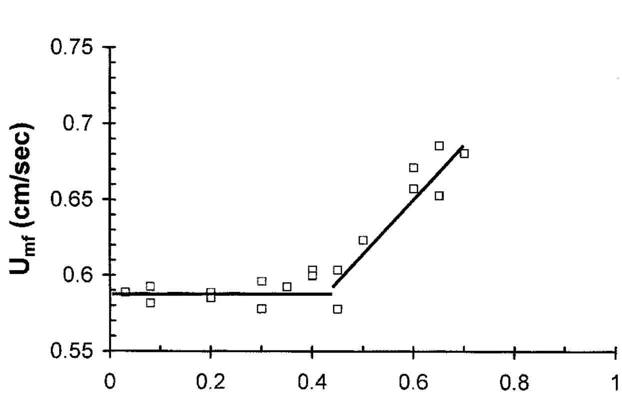

Figure 6. Minimum fluidization velocity as function of rH.

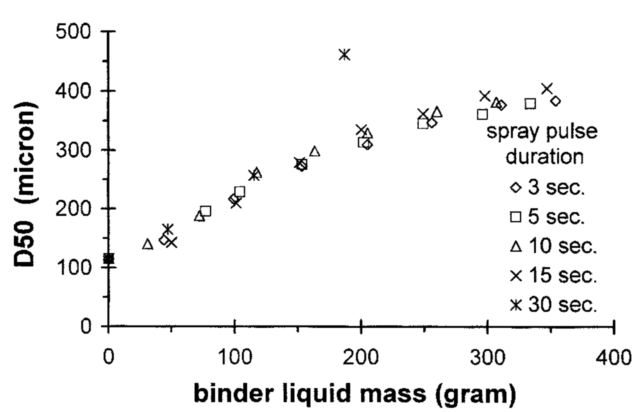

Figure 7. Growth of granules made by pulsed spraying, large droplet size.
U_G 32 cm/s, mean droplet size 99 micron, inlet air 40°C.

Table 3. Bed Mixing

d_{sv} (μm)	U_g (cm/s)	U_{mf} (cm/s)	Bed Height (cm)	U_e Bottom (cm/s)	U_e Top (cm/s)	I_m Bottom (Hz)	I_m Top (Hz)
Lactose 110 Mesh							
110	26	0.59	9	25.4	15.5	0.91	0.90
110	32	0.59	9	31.4	19.3	1.13	1.11
110	40	0.59	11	39.4	22.0	1.41	1.40
110	48	0.59	11	47.4	26.5	1.70	1.68

Here I_m is the mixing frequency (Hz), M_{bed} the bed load (kg), A the surface area of the bed at a given height (m^2), ρ_p the particle density (kg/m^3), and ϵ_{em} the porosity in the emulsion phase. Table 3 shows the wake transport and mixing frequency of the fluid bed for lactose 110 mesh at the distributor plate and the approximate bed height. The mixing frequency was found to be less than 0.9 Hz, depending on the fluidization velocity and bed height. The difference between the mixing frequency that is determined at the top of the bed and at the distributor plate (bottom) is small, because U_{mf} is very small in respect of U_g.

Since no influence of the spray durations was found, we conclude (Table 1) that surface renewal is a very fast process compared to our spray rate. The residence time of a newly formed granule is considerably less than 3 s, as we otherwise would see variations in the granule size due to different probabilities of wetting between the various spray durations (3 to 30 s). Regarding to the bulk mixing there is no effect of the granule size when changing pulse duration as well. Fast drying (Table 1) could explain this. However, after the liquid pulse was sprayed we measured an increased humidity above the bed for a long period of time (several minutes), so drying was slow compared to the spray time. That we find no effect of the spray-pulse duration may indicate that the chance of rewetting a granule during its time at the surface is small or that rewetting is just as effective as primary wetting. The chance of rewetting can be estimated by estimating the fraction of particles forming granules during one pulse period, if this fraction is small, so is the chance of rewetting. If every droplet results in a granule, then the number of sprayed droplets in a pulse period is the total number of wet particles. The number of wet particles divided by the total number of particles in the bed is the fraction of wet particles. The fraction of particles wetted during one pulse at the start of granulation varies from 0.5% to 5%, for 3- to 30-s spray-pulse duration, respectively, when spraying droplets with a D_{50} of 99 micron. This approach is a worst-case scenario, because not every granule will be expected to arrive in the small wetting zone at the top of the bed and not every granule will be formed by one droplet. Some granules will be formed by more than one droplet, depending on the spray rate and surface renewal (Schaafsma et al., 1998a), and thus coalescence of droplets in the wetting zone will result in an even lower fraction of wet granules in the fluid bed. Furthermore, as granules grow larger, mixing will decrease and fewer granules reenter the wetting zone.

The spray time of 30 s resulted in an early collapse of the bed (Figure 7, the single point above the growth curve). This could be due to the higher relative humidity, which for this long spray time exceeded 65% rH measured by the sensor above the bed. From the results in Figure 6 we might have expected bed collapse to occur at 75% instead of 65% rH. However, the relative humidity in the emulsion phase of the bed determines the spray limit and not the overall humidity measured above the bed. The difference between the two is due to the fact that the air bypassing the bed in bubbles contains less moisture than the air in the emulsion phase. The particle interaction determining the fluidization behavior of the bed is determined by the local relative humidity, which for most particles means within the emulsion phase.

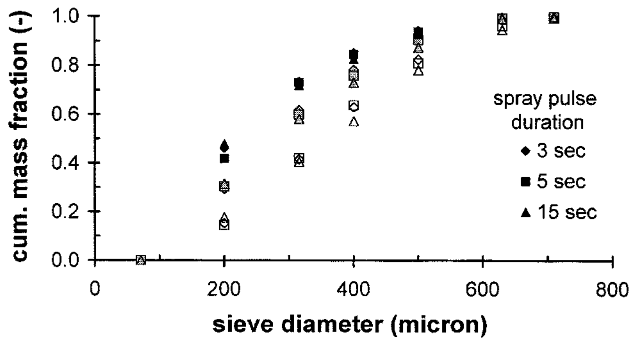

Figure 8. Size distribution of granules made by pulsed spraying, large droplet size.

U_G 32 cm/s, droplet size 99 micron, inlet air 40°C. Black symbols: 100 g binder liquid sprayed; gray symbols: 150 g of binder liquid sprayed; white symbols: 250 g of binder liquid sprayed.

As was the case for the granule mean size (Figure 7), the granule size distributions were unaffected by the spray-pulse duration. This is shown in Figure 8. The reproducibility of the experiments was good, as less than 4% deviation of one measured data point was found in repeat experiments.

Spraying with small droplets (mean diameter 16 μm) shows (Figure 9) the same characteristics as spraying with large droplets (mean diameter 99 μm) (Figure 7). The fluid bed collapsed at a granule mean mass size of about 330 μm, which was less than when spraying with the larger droplets (compare with Figure 7). Again, we see that the mixing has to be sufficient to avoid overwetting of the surface, It seems that

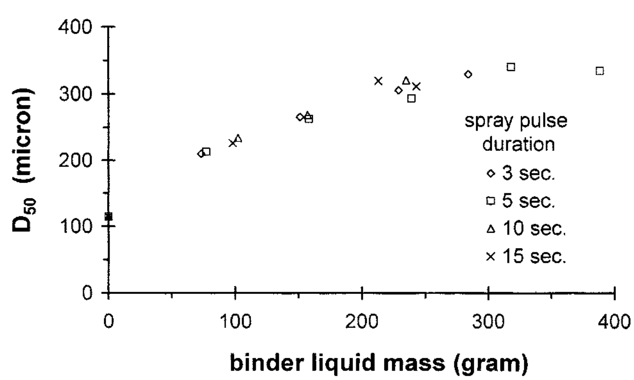

Figure 9. Growth of granules made by pulsed spraying, small droplet size.

U_G 32 cm/s, droplet size 16 micron, inlet air 40°C.

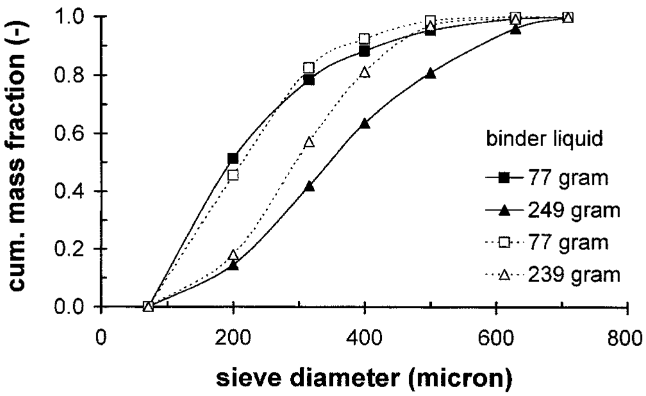

Figure 10. Variation of droplet size, influence on size distribution.

U_G 32 cm/s, inlet air 40°C, spray pulse duration 5 s. White symbols: droplet mean size 16 micron; black symbols: droplet mean size 99 micron.

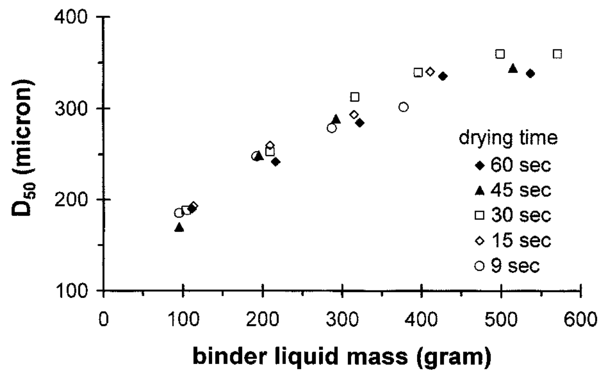

Figure 11. Growth of granules made by pulsed spraying, change of lag time.

U_G 32 cm/s, droplet size 16 micron, inlet air 60°C, spray pulse duration 3 s.

this mixing is very critical, which can be explained by the fact that as soon as larger lumps are formed at the surface, they decrease the mixing and cause further overwetting and defluidization. This was also suggested by Smith and Nienow (1983). Collapse of the bed occurs at a different total liquid mass when spraying with small droplets instead of large droplets. This is due to the fact that the distributions of droplets on the bed surface from the nozzle is different, and granule-size distribution and thus surface renewal is different (Figure 10).

The granules made by the smaller droplets had smaller spread in mass distribution than those made by larger droplets (Figure 10). The effectiveness in causing secondary growth due to rewetting an unsaturated granule is very low for the small droplets, because the granule absorbs a part of the droplet by capillary suction (Schaafsma et al., 1998b). This may also explain why the growth rate decreases during the process. Some authors such as Nienow and Rowe (1985), Smith and Nienow (1983) and Watano et al. (1995) suggest that the decrease of the growth rate is due to attrition by shear forces, which the granules encounter in the fluid bed. Breakage may play a role in more brittle granules, but we did not find any significant breakage of the granules during long fluidization experiments for this particular material.

By variation of the drying time between liquid spray pulses of a constant short duration, we are able to examine the influence of the relative humidity on the growth of granules. No influence of the humidity on the growth curve of the granules is seen, when the mean granule size (D_{50}) is plotted against the sprayed binder liquid mass in Figure 11. This could be explained by the drying rate being very slow compared to the mixing rate. The variation of the saturation between the granules made at different rH should then be small and of no consequence for the rewetting of the granules and the form of growth curve. In Figure 11 the last point in each series corresponds to bed collapse. This can be seen to occur earlier for the shorter drying times, which are also the conditions at which a higher rH was measured above the bed. This result is similar to the result found when applying a longer spray-pulse duration of 30 s (Figure 7), where a higher rH also resulted in an early collapse of the bed.

The final size of the granules at the moment that the bed collapses is smaller when a shorter lag (drying) period between the liquid spray pulse is applied (Figure 11). By comparing the U_{mf} of the granules with the rH at the time of bed collapse (Figure 12), a distinct relation is seen. Even at a very high rH of more than 90% the bed can still be fluidized for some time, which is probably due to the fact that it takes time to build up liquid at the surface of lactose by adsorption. This was also found by applying dynamic vapor adsorption experiments, where it took some time (up to an hour) to reach the equilibrium adsorption value. It seems from these results that the rH does not influence the granule size, or the growth mechanism of the granules directly, but only the point at which the bed collapses.

An increase in the fluidization velocity would, in accordance with our previous discussion, lead to a later collapse of the fluid bed, due to an increase of the excess gas velocity and thus mixing at the spray surface. This was found to be true, as shown in Figure 13, where the average granule size is plotted against the binder liquid mass for droplet mean diameters of 16 μm and 99 μm and fluidization velocities of 26, 32, 40 and 48 cm/s.

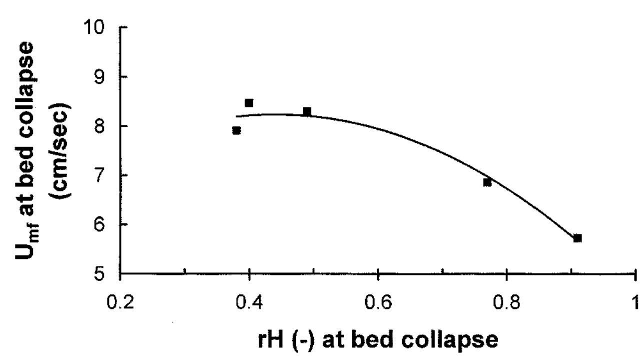

Figure 12. Relation between U_{mf} of final-sized granules and bed humidity, at the time of bed collapse.

U_G 32 cm/s, droplet size 16 micron, inlet air 60°C, spray pulse duration 3 s.

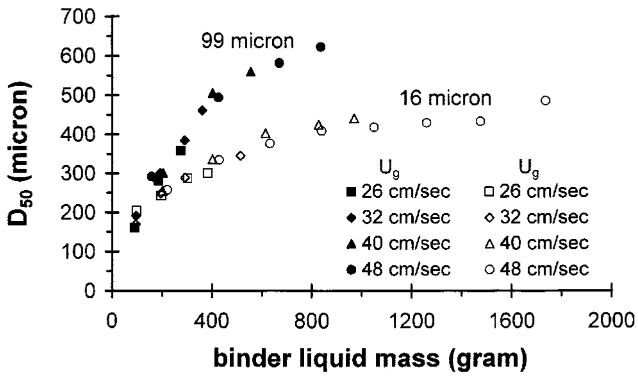

Figure 13. Influence of fluidization velocity on the granule growth.

Inlet air 60°C. White symbols: droplet mean size 16 microns; black symbols: droplet mean size 99 micron.

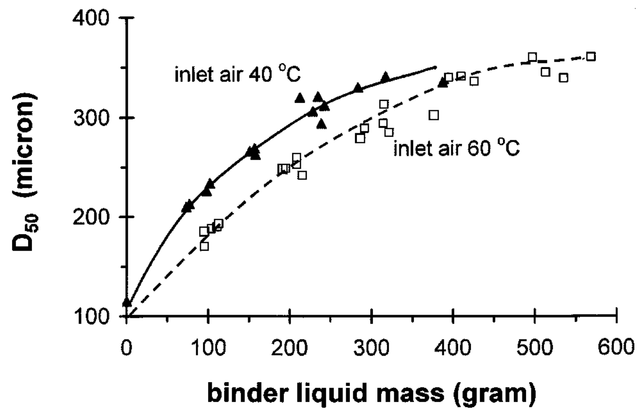

Figure 15. Influence of the inlet temperature.

U_G 32 cm/s, droplet size 16 microns. Black symbols: inlet air temperature 40°C, spray pulse 3–15 s, lag time 60 s; white symbols: inlet air temperature 60°C, spray pulse 3 s, lag time 60–30 s.

The increase in fluidization velocity can be seen not to influence the granule-size distribution when compared at the same amount of binder liquid sprayed. Thus a faster turnover of the fluid bed at a higher fluidization velocity does not influence the granule size. A higher fluidization velocity does enable us to spray more binder liquid before mixing in the spray zone becomes so bad that the fluid bed collapses. This is clearly shown in Figure 14, where the final granule size as the mean surface area diameter (D_{sv}) is plotted as a function of U_g for the droplet mean diameters of 16 μm and 99 μm. Here we plot the mean surface area diameter (D_{sv}) because this value is related to the fluidization velocity, rather than the average mass-based diameter.

The granule mean size (D_{sv}) at bed collapse shows a linear trend with the fluidization velocity (Figure 14). When we measured the U_{mf} of the granules at the point of collapse and calculated the excess gas velocity according to the ideal two-phase theory (Eq. 1) with $n = 0$, we found an excess gas velocity at the bottom (U_e) of approximately 24 cm/s for all the experiments in spite of the very different values of U_G. From this we can conclude that at the point of collapse we have the same excess gas velocity and thus the same rate of surface mixing. This surface mixing is the limiting factor at the applied spray rate before overwetting of the spray surface and collapse of the bed takes place.

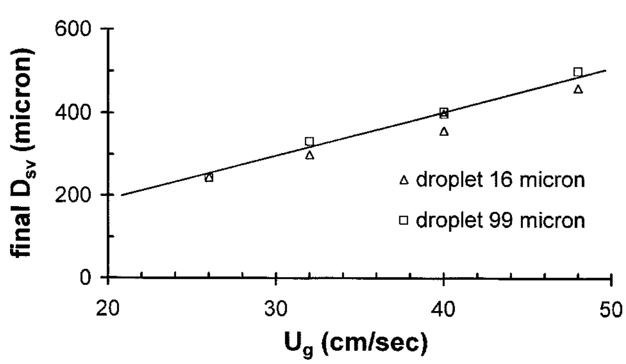

Figure 14. Granule mixing and bed collapse.

Final granule size (D_{sv}) at time of collapse as function of U_g, inlet air 60°C.

We found a distinct effect of the inlet temperature of the fluidizing air on the granule growth (Figure 15). At a lower temperature of 40°C, the granules grew to their final size faster than at 60°C. The final mean sizes of the granules at the point of the bed's collapse were approximately the same as for the two inlet temperatures. There are several possible explanations for this. Considering bulk mixing, we can conclude that (Table 1) the drying rate at a higher inlet temperature is enhanced relative to the mixing rate, which was not changed. Since the fraction of wet granules is small, the chance of rewetting of an already wet granule is small anyway, and so this is unlikely to affect granule growth. Another explanation could be that at the higher temperature the droplets dry faster in their flight from the nozzle to the bed surface, reducing the amount of effective wetting liquid (Nienow, 1993). Furthermore, primary growth could be reduced because the granules, when wetted, dry very fast, so that a large amount of binder liquid evaporates before they can bind other particles. According to Schaafsma et al. (1998b) this last effect is not likely to be an important factor, because this wetting is very fast. However, we can not exclude this argument yet. More research has to be done to make a distinction between the possibilities mentioned.

Concluding Remarks

The rH influences the granule growth if it exceeds a critical value above 50% at which the adsorbed liquid layers are sufficiently thick to cause significant interparticle cohesion. The rH changes the granule growth because it influences the mixing behavior in the fluid bed through this cohesion. Beneath this critical value no influence of rH on the final granule size was found.

The mixing at the bed surface and in the bulk of the bed is fast compared to the spray pulse duration. The residence time of a granule at the surface and the turnover time of the bed are less than 3 s, the minimal spraying time applied. The turnover time of particles at the bed surface should be short to prevent the bed surface from overwetting. As granules become larger during the granulation process, mixing reduces, and this can eventually result in overwetting of the surface and collapse of the bed. The proposed mechanisms of mixing behavior and their influence of granule size and growth can explain the results found experimentally.

The proposed effect of bed mixing on the growth (Table 1) does not seem to have any effect on the granules' growth because of the low fraction of wet granules in the fluid bed compared to the dry granules. The applied spray pulses did not effect the granule growth. The droplet-size distribution influences the growth rate and granule size. The fluidization velocity does not influence the granule growth rate, but influences only the end point of granulation. A higher inlet temperature of the fluidization air decreases the growth rate.

Control of the rH in the fluid bed is necessary, since above a critical level the rH of the emulsion phase has a large influence on the mixing, and the process is then difficult to control. A good way to control the humidity is by pulse spraying; as shown in this article, this results in a constant reproducible granule growth during the process, while the droplet size and spraying pattern are not influenced, since the spraying rate can be maintained. The spray pulse lag (drying) time depends on the drying capacity of the fluid bed and should preferably be short to ensure that the total process time remains short. Spraying with a pulse makes a given nozzle more versatile, because a larger range of spray rates can be used than under continuous spraying operation where higher spray rates will result in overwetting of the fluid bed. Spraying with a pulse is therefore believed to be superior to other ways of controlling the rH, such as changing the spray rate and thereby the droplet size and wetting at the bed surface, fluidization velocity, or the bed temperature, which are likely to change the granule-size distribution.

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