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Control of fluidized bed granulation

III. Effects of inlet air temperature and liquid flow rate on granule size and size distribution. Control of moisture content of granules in the drying phase

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Granulations were prepared in a fluidized bed granulator (Glatt, WSG 15) at different inlet air temperatures and liquid flow rates. Factors affecting drying rate were investigated, and the applicability of indirect methods for the control of fluidized bed drying are discussed.

Granule size was found to be inversely proportional to the difference between inlet air and wet bulb temperature in the granulation phase and directly proportional to the liquid flow rate. An increasing attrition in the drying phase was observed at increased liquid flow rate.

At varying experimental conditions a reproducible correlation was found between the moisture content of the granulation and the difference between product and wet bulb temperature.

As the formation of granules is induced by formation of liquid bridges between primary particles (10), moisture content is supposed to influence the size of the final granules. At any given time the moisture content of the granules depends on two factors, wetting and evaporation, which in turn are primarily controlled by liquid flow rate and inlet air temperature, respectively. *Ormós et al.* (11) have defined an equilibrium liquid flow rate as one at which liquid supply is balanced by evaporation, and a critical liquid flow rate as one above which fluidization is impossible due to cohesion in the bed. Ordinarily a liquid flow rate between these values is used.

Several authors (1, 3, 9, 12-14) have found that an increase in liquid flow rate results in a larger granule size, whereas *Thurn* (21) found no

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effect of this factor. Since the experiments were carried out at unchanged values of nozzle air flow rate, the air-to-liquid mass ratio was not kept constant, and the effect of liquid flow rate might therefore be due to a change in droplet size (16). *Ormós et al.* (11) used a constant value of mass ratio and found a slight decrease in granule size with increasing liquid flow rate. A simultaneous fall in droplet size might be expected (16), but the actual droplet sizes were not examined. To determine the influence of wetting on granule size it is necessary to keep droplet size constant at different liquid flow rates.

The fluidizing air is usually heated to 40-80° C in order to accelerate evaporation of liquid. Drying rate is described by the equation *(Ganderton* (4)):

$$\frac{dW}{dt} = \frac{h \cdot A}{\Delta H} \cdot \Delta T \qquad (1)$$

where $\frac{dW}{dt}$ is the mass transfer rate (drying rate), h is the heat transfer coefficient, A is the surface area, ΔH is the latent heat of evaporation, and ΔT is the difference between the temperatures of drying air and product surface. When the latter is saturated by water the product temperature is equal to the wet bulb temperature as shown earlier (15). Accordingly, *Scott et al.* (18) found drying rate to be constant and directly proportional to ΔT as long as the granulation surface was completely covered with water.

Granule size may be influenced in the granulation phase by inlet air temperature on account of a simultaneous effect on evaporation, which affects agglomeration by liquid bridges, as well as in the drying phase due to an influence on drying time, which affects the attrition (18, 21, 22). Several authors (3, 5, 7, 12, 14) have found that a rise in inlet temperature causes a decreased granule size, whereas *Thurn* (21) found no effect of this factor.

It is possible to control the moisture content of a fluidized granulation during the process by means of indirect measurements, because other parameters depend on granule humidity (2). These are temperature (3, 8, 17) and humidity (19, 21) of the outlet air and the product temperature (6). *Stahl* (20) controlled the temperature of the outlet air by varying the temperature of the inlet air in order to obtain equilibration of the product with the drying atmosphere and consequently a controlled moisture content.

The purpose of this work has been to examine the effects of liquid flow rate and inlet air temperature on granule size and size distribution in order to elucidate the influence of moisture content on granule form-

ation and growth. Further, the applicability of an indirect method of controlling the moisture content of the final granules has been investigated.

Experimental

Materials and formulation

Starting materials were 15 kg of mixtures (4:1 and 1:4) of fine-crystalline lactose and maize starch (15). Aqueous solutions (3,500 g) of gelatine (Ph. Nord. 63) (4 %), sodium carboxymethylcellulose (7L1, Hercules) (CMC) (3 %) and Kollidon® 90 (BASF) (4 %) were used as binder solutions and prepared as previously described (16).

Equipment and procedure

A fluidized bed spray granulator (Glatt, model WSG 15) was used. Nozzle, instrumentation and general procedure were the same as in the previous studies (15, 16).

Granule size and size distribution

Granule size distributions were characterized by d_{gw} and s_g (15). The granule size distribution at the end of the granulation phase was determined by taking a sample of about 400 g from the bed before drying. The sample was dried at room temperature in a tray drier, and sieve analyses were carried out in duplicate as previously described (15), whereas analyses of the final granules dried in the fluidized bed were performed in triplicate.

Moisture content

The moisture content during the drying phase was followed by taking samples of about 3 g from the bed with a sampling probe. Loss on drying was estimated by heating to constant weight in an oven at 105° C. Water content is expressed in percentage by weight of water of dry solids. The moisture contents of the starting materials were determined to be 12.0 % in maize starch and 5.4 % in lactose. The latter value includes 5.3 % of water of crystallization, which is also included in the mentioned results of moisture content of the granules.

Results and discussion

Temperature and humidity of inlet air

Preliminary investigations showed significant differences between series of experiments. An increase in inlet air humidity causing a larger granule size, the results might be accounted for by differences in water content of the fluidizing air, which varied between 3 and 10 g/kg of dry air. A rise in air humidity results in a higher wet bulb temperature and, consequently, in a lower ΔT-value (eq. 1). In order to keep ΔT constant and thus to obtain a constant drying rate, the temperature of inlet air was varied in the following experiments (Table 1).

Table 1. Correlation between humidity and temperature of inlet air at constant ΔT. The values in the Table are derived from a psychrometric chart.

Air humidity g/kg dry air	ΔT = 20° C		ΔT = 35° C	
	Air temp. ° C	Product temp. ° C	Air temp. ° C	Product temp. ° C
1	33	13	55	20
2	34	14	56	21
3	36	16	57	22
4	37	17	58	23
5	38	18	59	24
6	39	19	60	25
7	40	20	60	25
8	41	21	61	26
9	42	22	62	27
10	43	23	63	28
11	44	24	64	29
12	45	25	64	29
13	45	25	65	30
14	46	26	65	30

As previously mentioned it is necessary to distinguish between ΔT in the granulation phase (ΔT_{gran}) and ΔT in the drying phase (ΔT_{drying}), as ΔT_{gran} may primarily affect granule growth whereas ΔT_{drying} influences drying time and thus attrition. In the following experiments granule size was estimated on basis of tray dried samples collected at the end of the granulation phase in order to determine the effect of the variable in question, and in the final granulation dried in the fluidized bed in order to investigate the attrition.

The influence of ΔT_{gran} on granule size is shown in Fig. 1. By analysis of regression it was concluded that the correlation could be described by a straight line. As can be seen the increase in drying rate obtained at higher values of ΔT_{gran} results in a decrease of granule size. No significant influence on the granule size distribution (s_g) was found.

Table 2 shows the effect of ΔT on attrition (Δd_{gw}) expressed as the differences in granule size before and after drying. Increased values of ΔT_{gran} shorten the drying time due to a lower water content at the start of the drying phase. Higher values of ΔT_{drying} also reduce drying time. Drying time seems to have only a slight influence on attrition. However, the effect may depend on the mechanical resistance of the granules.

Since a high inlet air temperature counteracts granule growth in the granulation phase and reduces drying time and attrition in the drying

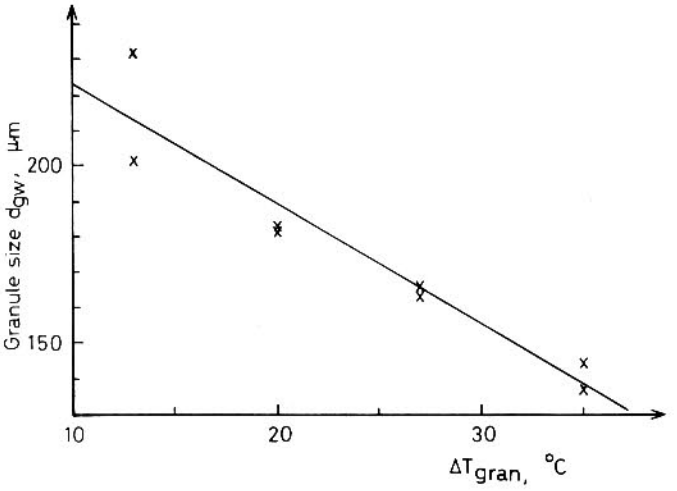

Figure 1. Correlation between ΔT_{gran} and granule size at the end of the granulation phase.
Starting materials: 80 % lactose + 20 % maize starch.
Binder solution: Gelatine 4 %, 40° C.
Liquid flow rate: 150 g/min. Nozzle air flow rate: 10 Nm^3/h.

phase, it is reasonable to use a higher ΔT-value in the drying phase. Therefore ΔT-values of 20 and 35° C during granulation and drying, respectively, were ordinarily used in the following experiments.

Liquid flow rate

Liquid flow rate was varied between 100 and 200 g/min, i.e. between the equilibrium and the critical values. The empirical droplet size equation derived in a previous work (16) was used to keep droplet size approxi-

Table 2. Influence of ΔT on attrition (Δd_{gw}, µm) in the drying phase at the experimental conditions described in Fig. 1. The values given in parantheses are the corresponding drying times.

ΔT_{drying}	ΔT_{gran}			
	13° C	20° C	27° C	35° C
20° C	32 µm (43 min)	24 µm (36 min)	27 µm (32 min)	12 µm (24 min)
35° C	24 µm (24 min)	15 µm (24 min)	18 µm (20 min)	24 µm (12 min)

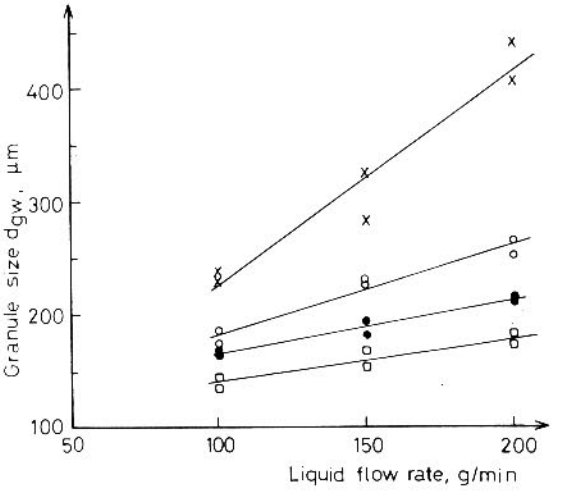

Figure 2. Correlation between liquid flow rate and granule size at the end of the granulation phase.
Starting materials: 80 % lactose + 20 % maize starch.
Binder solution temperature: 40° C. ΔT_{gran}: 20° C.
Binder solutions: ○: gelatine 4 %, d_{50} = 106 µm.
●: gelatine 4 %, d_{50} = 85 µm.
□: CMC 3 %, d_{50} = 135 µm.
×: Kollidon 90 4 %, d_{50} = 148 µm.

mately constant through variation of nozzle air flow rate. The results are shown in Fig. 2 and Table 3.

By analysis of regression it was found that the correlation between liquid flow rate and granule size can be described by a straight line, the slope of which depends on the binder solution used. The linear correlation is hardly valid at extremely low liquid flow rates where no granule formation occurs, or at flow rates near the critical value where growth may be uncontrollable. Increased liquid flow rate results in an increased number of liquid bridges and hence in a larger granule growth rate. Two droplet sizes of the gelatine solution were investigated, and the effects of droplet size and liquid flow rate on granule size were found by a two-factor analysis of variance to be significant at the 0.1 %-level. Interaction was found to be significant at the 5 %-level and was reflected by an increasing influence of liquid flow rate on granule size with increasing droplet size. Values of droplet size (d_{50}) were calculated from the equation previously derived (16).

A two-factor analysis of variance of the influence of liquid flow rate and droplet size on Δd_{gw} (Table 3) showed the effect of the former to be significant at the 1 %-level, whereas no effect of droplet size and no

Table 3. Influence of liquid flow rate and droplet size on attrition (Δd_{gw}) in the drying phase and on the geometric standard deviations (s_g) before and after drying.
Starting materials: 80 % lactose + 20 % maize starch.
Binder solution: Gelatine 4 %, 40° C.
ΔT_{gran}: 20° C, ΔT_{drying}: 35° C.

Liquid flow rate	Droplet size, d_{50}					
	106 µm			85 µm		
	Δd_{gw} µm	s_g		Δd_{gw} µm	s_g	
		before drying	after drying		before drying	after drying
100 g/min	8	1.88	2.05	13	1.77	1.98
	17	1.80	2.02	15	1.78	1.98
150 g/min	17	1.92	2.06	24	1.84	2.05
	21	1.86	2.04	21	1.80	2.00
200 g/min	45	1.87	2.12	50	1.82	2.15
	27	1.84	2.01	36	1.82	2.07

interaction were found. Increased attrition with increasing liquid flow rate has also been observed by *Ormós et al.* (11). A possible explanation is that agglomerates formed under these circumstances are looser due to a rise in growth rate and a shorter periode of mechanical stress in the granulation phase.

It can be seen from Table 3 that attrition in the drying phase causes a wider granule size distribution. The effect of droplet size on s_g was found by a two-factor analysis of variance to be significant at the 5 %-level whereas no effect of liquid flow rate was found. An increased droplet size results in a wider granule size distribution.

Drying rate

In order to elucidate factors affecting drying rate and time, experiments were carried out in duplicate under varying experimental conditions (Table 4). Droplet size was kept constant at different liquid flow rates by modifying the nozzle air flow rate. The quantity of water to be evaporated in the drying phase depends on evaporation during mixing and granulation. This was estimated as the difference between moisture content of the starting materials plus the quantity of water added and moisture content of the granules at the end of spraying. The maximum evaporation in granulation and drying phase was determined as the difference between the humidity of outlet and inlet air in the constant rate period during which product temperature is constant at the wet bulb temperature.

Table 4. Influence of mixing ratio of starting materials, liquid flow rate, nozzle air flow rate and ΔT_{drying} on evaporation of water. Binder solution: Gelatine 4 %, 40° C. ΔT_{gran}: 20° C.

Starting materials		Liquid flow rate g/min	Nozzle air flow rate Nm^3/h	ΔT_{drying} °C	Evaporation during mixing and granulation g water	Maximum evaporation in granulation phase g water/kg dry air	Maximum evaporation in drying phase g water/kg dry air
Lactose	Maize starch						
80 %	20 %	150	8.00	20	1,730 1,780	9 7	9 7
20 %	80 %	150	8.00	20	1,440 1,430	7 7	7 7
80 %	20 %	150	8.00	35	1,370 1,780	7 7	12 12
20 %	80 %	150	8.00	35	1,290 1,450	7 6	9 10
80 %	20 %	200	9.48	35	1,620 1,480	7 7	12 12
80 %	20 %	100	6.39	35	2,330 2,190	7 8	10 12
80 %	20 %	150	14.0	35	1,680 1,860	9 7	12 12

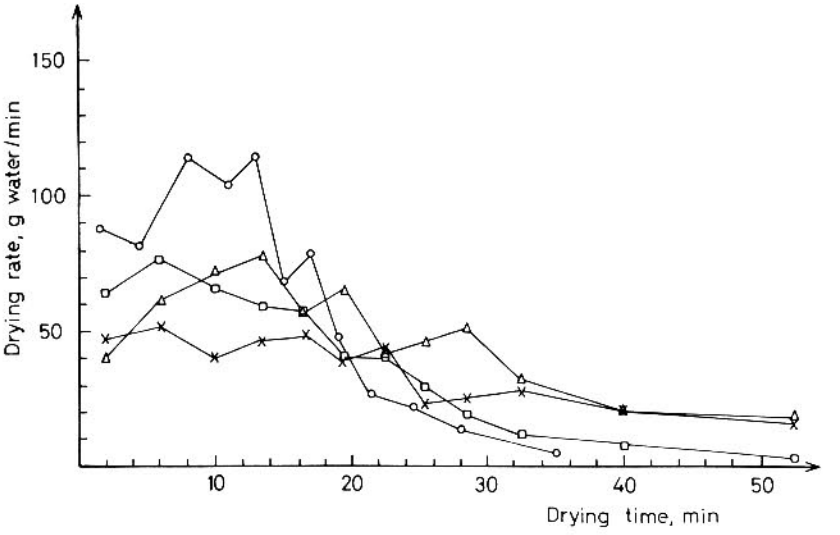

Figure 3. Correlation between drying time and drying rate.
Binder solution: Gelatine 4 %, 40° C.
Liquid flow rate: 150 g/min. Nozzle air flow rate: 8 Nm^3/h.
ΔT_{gran}: 20° C.
○: 80 % lactose + 20 % maize starch, ΔT_{drying}: 35° C.
□: 80 % lactose + 20 % maize starch, ΔT_{drying}: 20° C.
△: 20 % lactose + 80 % maize starch, ΔT_{drying}: 35° C.
×: 20 % lactose + 80 % maize starch, ΔT_{drying}: 20° C..

The time used for granulation being inversely proportional to liquid flow rate, evaporation during mixing and granulation is seen to be influenced primarily by liquid flow rate and secondarily by mixing ratio of starting materials. By analysis of variance the latter effect was found to be significant at the 5 %-level. Blends containing 80 % of maize starch show a slow evaporation. This could be explained by the fact that a great deal of water is bound by absorption in the starch.

In the granulation phase the maximum evaporation seems to be unaffected by the variables when ΔT is constant. In the drying phase two values of ΔT were used and, as was expected, an increase in ΔT results in a rise in evaporation. Due to absorption of water the surface of the granules containing 80 % of starch may not be completely wetted in the drying phase at the time when the inlet air temperature has reached the desired value, and maximum evaporation is therefore lower under these conditions.

Fig. 3 shows drying rate as a function of drying time at various values of ΔT and mixing ratios. Drying rates are calculated on basis of moisture

contents of samples collected as previously described, and the results are mean values of two estimations. The drying curves proceed ordinarily (4) through an initial constant rate period followed by a falling rate period with the break occurring at the critical moisture content. At ΔT-values of 35° C equilibrium is established during an initial heating period. The irregularities of the curves are caused by inevitable fluctuations in temperature and velocity of inlet air. *Scott et al.* (18) observed curves with similar shapes in the case of fluidized bed drying of various granulations.

As is to be expected from eq. (1) the highest drying rate was found at a ΔT-value of 35° C. When the surface of the granules is no longer saturated the rate falls and is governed by the diffusion of moisture from the interior. Water is mainly present as free water on the surface of lactose particles and therefore a sharper fall in drying rate is observed for the granulations containing 80 % of lactose compared with those containing 20 %. In the granulations with a high content of starch water is largely present in an absorbed form.

Similar drying curves were obtained from the remaining experiments in Table 4. As can be seen from eq. (1) drying rate depends on surface area and, therefore, on granule size. In accord with this expression *Zoglio et al.* (22) have found increasing drying rates with decreasing granule sizes. In the present experiments two granule sizes were obtained by variation of nozzle air flow rate, but no significant difference between their drying rates was observed.

Control of drying

In the introductory section various indirect methods of controlling the moisture content of a fluidized granulation were mentioned. Since the temperature of outlet air depends on loss of heat to the surroundings, and since the humidity of outlet air changes simultaneously with variations in humidity of the inlet air, measurement of the product temperature seems to be the most simple and accurate way of controlling the drying process in a fluidized bed. Therefore, the applicability of the method of *Harbert* (6) was investigated at the experimental conditions described in Table 4.

Product temperature remains at wet bulb temperature until the critical moisture content is reached and then rises due to a diminished drying rate as shown in Fig. 4. The slow release of water absorbed in the starch accounts for the influence of mixing ratio of the starting materials on the product temperature in the falling rate period in accordance with the effect on the drying rate (cf. Fig. 3). After drying for about 1 h product temperature approaches that of the inlet air in the case of 80 % of lactose, indicating that equilibrium moisture content is almost reached.

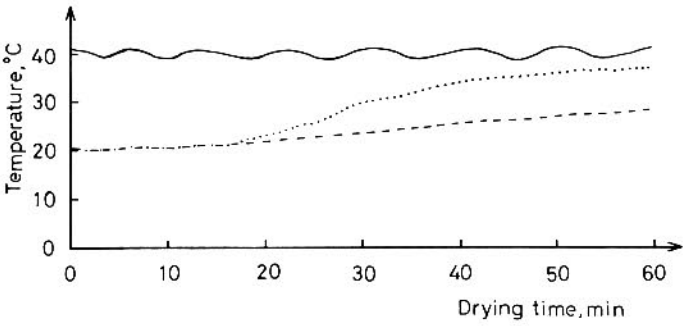

Figure 4. Correlation between drying time and product temperature.
Binder solution: Gelatine 4 %, 40° C.
Liquid flow rate: 150 g/min. Nozzle air flow rate: 8 Nm^3/h.
ΔT_{gran}: 20° C. ΔT_{drying}: 20° C.
———— Inlet air temperature.
..... Product temperature. 80 % lactose + 20 % maize starch.
– – – – – Product temperature. 20 % lactose + 80 % maize starch.

This does not occur at the high level of starch content. Drying is ordinarily stopped before equilibrium in order to attain a moisture content equal to that of equilibrium at room temperature.

The moisture content of the granules as a function of the difference between product temperature and wet bulb temperature at varying experimental conditions was investigated, and examples are shown in Fig. 5. Experiments were carried out in duplicate and the correlation is seen to be reproducible. Within the investigated ranges ΔT, liquid flow rate and granule size were not found to affect the relationship, this being solely dependent on the starting materials.

Control of drying is facilitated if water is present mainly as free water on the surface, since a slight variation in product temperature in such cases causes an inconsiderable change in water content due to a rapid rise in product temperature. In practice, therefore, variations in wet bulb temperature can ordinarily be ignored and product temperature alone can be used as a measure of water content. For large starch contents, however, accurate measurements of wet bulb and product temperatures are necessary to control moisture content.

Conclusions

Variations in inlet air humidity was found to affect granule size, but this influence can be eliminated by keeping the difference between temperatures of inlet air and wet bulb (ΔT) constant.

Granule size was inversely proportional to ΔT_{gran} and directly pro-

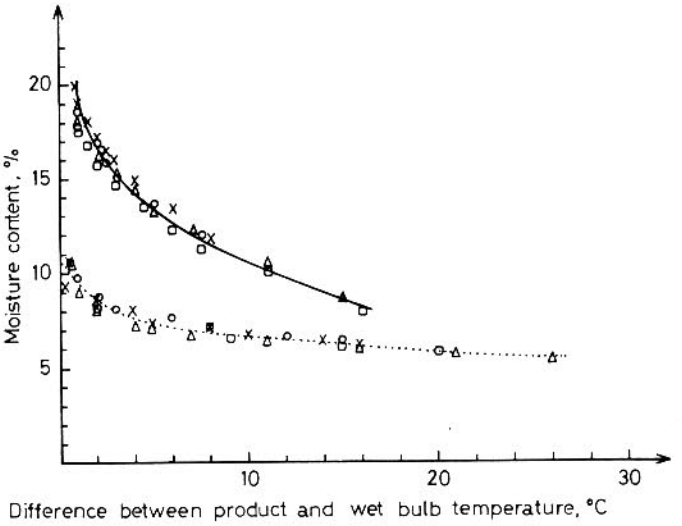

Figure 5. Moisture content of the granules as a function of the difference between product and wet bulb temperature.
Starting materials: 80 % lactose + 20 % maize starch (.....)
20 % lactose + 80 % maize starch (————)
Binder solution: Gelatine 4 %, 40° C. ΔT_{gran}: 20° C.
Liquid flow rate: 150 g/min. Nozzle air flow rate: 8 Nm^3/h.
○ ×: ΔT_{drying} = 20° C.
□ △: ΔT_{drying} = 35° C.

portional to liquid flow rate. In both cases an increase in granule size is caused by a rise in the quantity of water, and consequently in the number of liquid bridges, on the surface of the particles, indicating that granule size is directly proportional to the moisture content of the bed in the granulation phase.

The attrition in the drying phase was shown to depend primarily on the mechanical resistance of the granules and secondarily on drying time.

Product temperature was found suitable for the control of fluidized bed drying, since the correlation between water content of the granulation and the difference between product and wet bulb temperature is solely affected by the water-binding properties of the starting materials.

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