Electrostatic charging during a melt agglomeration process

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Abstract

Lactose monohydrate was melt agglomerated in an 8-l high shear mixer using stearic acid, polyethylene glycol (PEG) 3000, and a mixture of stearic acid and glycerol monostearate as meltable binders. Electrostatic charging during processing at relative air humidities of 35 and 75%, respectively, was estimated from the voltage of a monitoring probe inserted into the mixer. Stearic acid produced a high level of electrostatic charges, whereas PEG 3000 gave rise to a markedly lower level. Addition of glycerol monostearate to the stearic acid reduced the level of electrostatic charges. A correlation was found between the resistivity of the binder and the level of electrostatic charges in the material. With the stearic acid and the binder mixture, the level of electrostatic charges was higher at a low air humidity. The amount of adhesion to the bowl was found to depend on the level of electrostatic charges. © 1999 Elsevier Science B.V. All rights reserved.

1. Introduction

When two dissimilar materials make contact, charge transfer from one of the materials to the other occurs (Bailey, 1984; Pavey, 1998). On separation, materials that are poor conductors or isolated conductors will retain the transferred charge, i.e. the particles will become electrostatically charged (Jonassen, 1998; Pavey, 1998). The magnitude of the electrostatic charging of a given material is dependent on several parameters such as the surface area of contact, the size and shape of the particles, the contact pressure, surface im-

purities, the environmental conditions (Bailey, 1984), and the magnitude of frictional forces (Pavey, 1998).

Electrostatic charging of particles during processing of powders will normally be undesirable. Electrostatic charging has been found to cause an adhesion of the material to the walls of the processing equipment (Gamlen et al., 1982; Bailey, 1984; Guardiola et al., 1996; Wolny and Opalinski, 1996), an ignition hazard associated with electrostatic spark creation (Blythe and Reddish, 1979; Cartwright et al., 1982), and a reduction or a failure of lung deposition of aerosolized fine particle doses from dry powder inhalers (Byron et al., 1997; Mackin et al., 1997). However, electrostatic charging of powders has been utilized for

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powder coating (Grosvenor and Staniforth, 1996; Mazumder et al., 1997) and for a formation of ordered powder mixtures (Staniforth and Rees, 1982).

An abatement of static electricity might be achieved by a proper grounding of all parts of the equipment (Kassebaum and Kocken, 1997) or by inserting a grounded probe inside the processing equipment (Blythe and Reddish, 1979). Other ways of reducing the static electricity are application of a radioactive ionization device (Kassebaum and Kocken, 1997) or a superimposed electric field (Ebadat and Bailey, 1990). Further, static electricity may be reduced by maintaining a high relative air humidity (Cartwright et al., 1982; Guardiola et al., 1996), or the presence of antistatic agents (Kassebaum and Kocken, 1997) might reduce the static electricity.

Polyethylene glycols have been successfully applied as binders in melt pelletization since a high yield of pellets is obtained (Schæfer et al., 1992; Schæfer, 1996; Schæfer and Mathiesen, 1996b). In contrast, different hydrophobic binders were found less suitable for melt pelletization. Lack of pelletization capabilities, adhesion of the material to the bowl, and electrostatic charging of the materials were amongst the obstacles met (Schæfer et al., 1990; Thomsen et al., 1994; Peerboom and Delattre, 1995). A low hydroxyl number of the meltable binder was found to increase the amount of adhesion to the bowl. This was ascribed to a higher affinity of a hydrophobic binder towards the PTFE bowl lining used in those experiments (Peerboom and Delattre, 1995). A regular helix-like movement of the mass during processing was previously found to favor the formation of pellets (Schæfer et al., 1993b). With hydrophobic binders, distortion of the material movement due to material building up on the bowl might thus be partly responsible for the lack of pelletization capability.

An application of glycerol monostearate in a binder mixture with some meltable binders that were more hydrophobic was found to improve the technical performance of the process. A reduction in the electrostatic charging and in the adhesion of material to the bowl was achieved, and a regular movement of the material during processing was observed (Thomsen et al., 1994).

Melt agglomeration in high shear mixers provides good conditions for generation of static electricity. The numerous numbers of collisions and the high shear forces involved ensure the establishment of contact and frictional forces, which enhance the likelihood of generation of static electricity. A relation between static electricity and the tendency for material to build up on the bowl was indicated by the results obtained by Thomsen et al. (1994). The role of electrostatic charging in a melt agglomeration process is unclear, however, since a qualitative measure for the generation of static electricity has not been provided. A knowledge of electrostatic charge generation during processing is necessary to aid the explanation of the differences in the pelletization capabilities of different meltable binders.

The purpose of the present work was to investigate effects of binder type and air humidity on generation of static electricity by melt agglomeration in a high shear mixer.

2. Materials and methods

2.1. Materials

Lactose 450 mesh (α-lactose monohydrate, DMV, The Netherlands) was used as starting material. Stearic acid (BP 93 grade, Jahress Fabrikker, Norway), glycerol monostearate (Ph.Eur. grade, Grindsted A/S, Denmark), and polyethylene glycol (PEG) 3000 (Hoechst, Germany) were used as meltable binders. Stearic acid and PEG 3000 were used as flakes whilst glycerol monostearate was used as a powder.

The size distribution by volume of the lactose was determined by a Malvern 2601Lc laser diffraction particle sizer (Malvern Instruments, UK). The median particle diameter and the span were 22 μm and 2.3. The span is the difference between the diameters at the 90 and the 10 percentage points relative to the median diameter.

The BET multipoint surface area of the lactose, determined by a Gemini 2375 Surface Area Analyzer (Micromeritics, USA), was 0.76 m^2/g.

The true densities of the materials were determined by an AccuPyc 1330 gas displacement pyc-

nometer (Micromeritics, USA) using helium purge. The density of the lactose was 1.549 g/cm³.

The water content on a wet-weight basis, i.e. the water content corresponding to the loss on drying, was estimated by volumetric titration as previously described (Schæfer and Mathiesen, 1996a). The water content was found to be 5.0% for the lactose.

The melting ranges of the meltable binders were estimated by a Perkin Elmer DSC 7 differential scanning calorimeter as previously described (Schæfer and Mathiesen, 1996a).

The viscosities of the molten binders were estimated at 56°C by a rotation viscosimeter, Rotovisco RV 12 (Haake, Germany). A MV sensor system and a MV II K rotor were applied with PEG 3000, whilst a NV sensor system was applied with stearic acid and the binder mixture of stearic acid and glycerol monostearate. The viscosity values used are the results obtained at 256 rpm.

The electrical resistance of a sample of the meltable binders was measured at 52°C. The binders were melted and poured into a measuring chamber. The measuring chamber was constructed from polypropylene with parallel walls constructed of brass and polypropylene, respectively. The dimensions of the parallel brass electrodes were $39.5 \times 50.2 \times 15.0$ mm (H × L × W), and the separation between them was 50.0 mm. The chamber was placed in an oven and thermally equilibrated, and the resistance was measured by an electrometer (Keithley 610B, Keithley Instruments, Inc., USA) at a field strength of approximately 2400 V/m. During the measurements, the relative air humidity in the room outside the oven was approximately 40% at a temperature of 20–23°C. The bulk resistivity, ρ, was calculated from the bulk resistance, R, and the internal dimensions of the chamber according to Eq. (1).

$$\rho = R \cdot \frac{A}{d} \tag{1}$$

where A is the area of the brass wall, and d is the separation between the brass walls.

2.2. Equipment

The 8-l laboratory scale high shear mixer (Pellmix PL 1/8, Niro A/S, Denmark), described in a previous paper (Schæfer et al., 1993a), was employed in the experiments. The temperature of the heating jacket was set to 35°C in all the experiments.

A hygrometer (Testo 600, Testo, Germany) was used to measure the relative air humidity in the room.

2.3. Mixing procedure

The load of the mixer was 1 kg of lactose. The concentration of the meltable binders was kept constant at 23% m/m of the amount of lactose.

In all the experiments, an impeller speed of 1300 rpm was applied until the product temperature reached 58°C after which the impeller speed was lowered to 400 rpm, and the massing was continued for 4.5 min.

The procedure for cooling of the agglomerates was the same as described in a previous paper (Schæfer, 1996). The adhesion of material to the bowl was estimated as the difference between the amount of material fed to the bowl and the amount of material that could be freely emptied after processing.

A relative air humidity of either approximately 35 or 75% at a temperature of 18-20°C was maintained in the room. The higher level of air humidity was achieved by evaporation of water. To ensure a similar absolute humidity in the room and within the mixer, the lid of the mixer was kept open 30 min prior to the performance of each experiment.

2.4. Static electric measurements

A lid of polymethylmethacrylate was constructed for the Pellmix to be applied during the electrostatic measurements. A monitoring probe was inserted through a centred hole in the lid and mounted to the lid by means of a PTFE stopper. The probe was made of a thin stainless steel rod with a diameter of 3 mm and a length of 215 mm. At 115 mm from the lid, the probe was bent in an

angle of 120°. This positioned the probe tip approximately 25 mm above the impeller and 65 mm apart from the inner surface of the bowl (Fig. 1).

The probe was connected to one terminal of an electrometer (Keithley 610B, Keithley instruments, Inc., USA) via a 10^{-10} F capacitor with a leakage resistance of $1 \cdot 10^{14}$ Ω. The other terminal of the electrometer was connected to earth. During all the experiments, the electrometer was operated in the charge measuring mode at the $30 \cdot 10^{-7}$ C range and in the center zero reading. Charges could thus be directly read from $-15 \cdot 10^{-7}$ C to $+15 \cdot 10^{-7}$ C without changing the scale of the electrometer.

Since the external capacitor of 10^{-10} F is in series with the input capacitor of the electrometer, 10^{-7} F at this range, the charges read on the electrometer are also the charges located on the probe. Consequently, the charges on the probe could have been measured without the external capacitor. However, without the external capacitor, the probe would have represented a point of virtually zero potential, which is probably far from the situation during operation without measurements. It would have been preferable to locate the probe at a site of zero potential, but it was considered essential to perform the measurements in the interior of the chamber, where most of the activity takes place. With the relatively small external capacitance, the probe distorts the field conditions only to a minor degree, and we can calculate the voltage at the site of the probe by the charge measured over the capacitance of the external capacitor applying Eq. (2) (modified from Jonassen, 1998):

$$V = \left(\frac{(C_m + C_e)}{C_e} \right) \cdot \left(\frac{q}{C_m} \right) \qquad (2)$$

where q is the measured charge; C_m is the electrometer capacitance (10^{-7} F); and C_e is the capacitance of the external capacitor (10^{-10} F).

If the electrical field at the tip of the probe exceeds the breakdown field strength, ions of both polarities will be formed by an electric discharge and move in the field. With sharp edges of the probe, the discharge is likely to be a corona discharge (Jonassen, 1998). Neutralization of the charges on the material by oppositely charged air ions thus reduces charge accumulation on the material. The probe charges deduced from the measurements are probably a combination of charges induced on the probe from the field from the charges in the material and to some extent charges transferred by air ions caused by corona discharges. With the set-up used for these experiments, there was no possibility of distinguishing between induced charges and charges directly transferred to the probe by air ions. The charges and voltages measured are thus taken as the best available measure of the electrostatic state in the material during the experiments described.

2.5. Granule characterization

2.5.1. Size distribution

The granule size distribution was estimated by a sieve analysis of a sample of about 100 g drawn by scooping from the cooled fraction finer than 4 mm, and the geometric mean diameter (d_{gw}) and the geometric standard deviation (s_g) were calculated. A series of 12 ASTM standard sieves in the range of 180–2000 μm was vibrated for 10 min by a Fritsch analysette 3 vibrator (Fritsch, Germany).

Size fractions including the three sieve fractions closest to the mean diameter were prepared by vibrating for 5 min, and these size fractions were applied for the measurements in Sections 2.5.2 and 2.5.3.
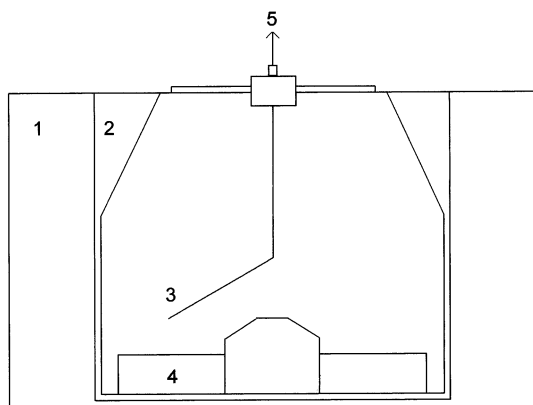

Fig. 1. Schematic drawing of the position of the probe in the high shear mixer. (1) heating jacket, (2) PTFE lining, (3) measuring probe, (4) impeller, and (5) connection to the electrometer.

Table 1
Physical properties of the meltable binders

Meltable binder	True density (g/cm^3)	Water content (%)	Melting range (°C)	Peak temperature (°C)
Stearic acid	0.990	0.0	46–56	55
Glycerol monostearate	1.033	0.5	47–63	58
PEG 3000	1.225	0.6	49–57	55

2.5.2. Intragranular porosity

The intragranular porosity was estimated by a mercury immersion method as previously described (Schæfer et al., 1992; Schæfer and Mathiesen, 1996a).

2.5.3. Water content

The water content on a wet-weight basis of the granules was estimated by volumetric titration as previously described (Schæfer and Mathiesen, 1996b).

2.6. Experimental design

A series of 20 experiments was carried out using either stearic acid, PEG 3000, or a mixture of 20 parts of stearic acid and three parts of glycerol monostearate as meltable binder. For each binder, the relative air humidity was adjusted to provide 35 or 75%.

With stearic acid and the binder mixture, experiments were performed with and without the measurement of static electricity being conducted. With PEG 3000, experiments were only performed where measurement of static electricity was conducted. All experiments were carried out in duplicate.

3. Results and discussion

3.1. Binder characteristics

The physical properties of the meltable binders are shown in Table 1. The melting ranges and peak temperatures of stearic acid and PEG 3000 are found to be similar, whilst glycerol monostearate has a slightly wider melting range and a slightly higher peak temperature.

At 56°C, the viscosity of PEG 3000 was 316 mPa·s, whilst the viscosities of stearic acid and the binder mixture of stearic acid and glycerol monostearate were 12 and 14 mPa·s, respectively.

The bulk resistivity for each of the three binders and for the mixture of stearic acid and glycerol monostearate is shown in Table 2. It appears that the resistivity of stearic acid becomes considerably reduced when glycerol monostearate is admixed.

Handling of insulating materials is likely to increase the risk of static electricity generation, because such materials often gain charge easily and retain charge because of their high resistivities (Bailey, 1984). Thus, a higher resistivity of the meltable binder indicates a higher risk of electrostatic charging.

3.2. Electrostatic charging during the process

Fig. 2 shows an example of the relationship between the impeller speed, the power consumption of the impeller motor, the product temperature, and the monitored changes in the static voltage of the probe during processing.

Initially, an impeller speed of 1300 rpm is applied. During processing, the product temperature increases steadily because of heat caused by friction. When the product temperature reaches the

Table 2
Bulk resistivity of meltable binders at 52°C

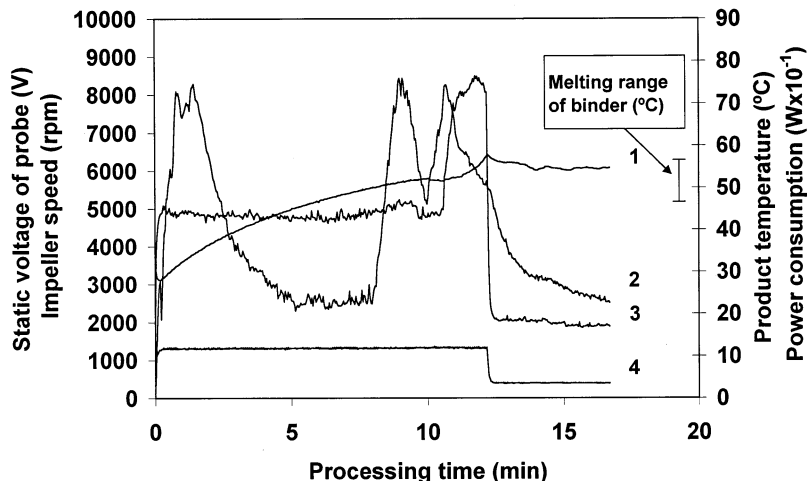| Meltable binder	Bulk resistivity (Ωm)
Stearic acid	5.9×10^{10}
Glycerol monostearate	3.2×10^{6}
Stearic acid/glycerol monostearate (20:3)	2.2×10^{9}
PEG 3000	2.1×10^{4}

Fig. 2. The relationship between: (1) product temperature; (2) the static voltage of the probe; (3) power consumption; and (4) impeller speed during processing. Meltable binder: stearic acid; relative air humidity: 35%.

melting range of the stearic acid, a distribution of the molten stearic acid upon the lactose particles is initiated. At approximately 53°C, corresponding to a processing time of about 11 min, sufficient binder is distributed to initiate agglomerate growth by coalescence as reflected by the sharp rise in the power consumption. At a product temperature of 58°C, i.e. a temperature slightly above the melting range of the binder, corresponding to a processing time of approximately 12 min, the impeller speed is lowered to 400 rpm in order to maintain the product temperature within the melting range of the binder (Section 3.4). Consequently, an abrupt fall in power consumption and a slight fall in product temperature are observed. During the remainder of the process, the product temperature and the power consumption are almost constant.

During the process, charges are continuously separated, but as the level of charges increases, a neutralization process sets in until an equilibrium state has been reached. This state is presented by the voltage of the probe. The record of the static voltage shows initially a high level which gradually levels off within the first few minutes before reaching the melting range of the binder. Later in the process, two more peaks are observed within the melting range of the binder. When agglomerate growth by coalescence commences after 11

min of processing, the static voltage decreases. An abrupt fall in the static voltage is seen when the impeller speed is lowered from 1300 to 400 rpm after a processing time of 12 min.

At collisions between particles covered with the molten stearic acid, a higher area of contact is established due to the deformability of the molten binder. A higher contact area have previously been found to increase charge transfer (Coste and Pechery, 1981). The transfer of charges may further be eased at a higher temperature (Vick, 1953). The ability to retain the transferred charges is promoted by a high resistivity of the material (Bailey, 1984). The high resistivity of the stearic acid (Table 2) combined with an increased area of contact might explain why the static voltage increases shortly after the product temperature has reached the melting range of the stearic acid.

Carter et al. (1992) reported that the specific charge, i.e. surface charge to mass ratio, of larger particles was lower than that of smaller particles due to the lower surface area available for charge transfer. The decrease in the static voltage seen as the agglomerate growth commences may be attributed to the formation of larger agglomerates which reduces the surface area available for charge transfer.

A lower impeller speed reduces the velocity of the agglomerates and thus the contact pressure at

collisions between agglomerates. A lower specific charge as a result of a lower particle velocity has been reported by Carter et al. (1992). This could be attributed to a lower contact pressure which reduces the deformation and thus the area available for charge transfer (Coste and Pechery, 1981). This might explain the abrupt fall in the static voltage seen when the impeller speed is lowered.

3.3. Effects of air humidity and binder type

The effect of the relative air humidity in the room on the level of the static voltage of the probe obtained with stearic acid at 35 and 75% relative air humidity, respectively, is shown in Fig. 3. Due to the temperature dependency of the relative air humidity, the relative air humidity at the elevated temperature within the mixing bowl is lower than in the room and changes during processing. At the start of the process, the corresponding values of the low and the high relative air humidity within the mixing bowl were calculated to be approximately 14 and 29%, respectively, and at the end of the process, approximately 4 and 9%, respectively. The air humidity within the bowl was unaffected by water evolution or consumption during the process since the water content of the final agglomerates was found to be identical to that of the starting materials.
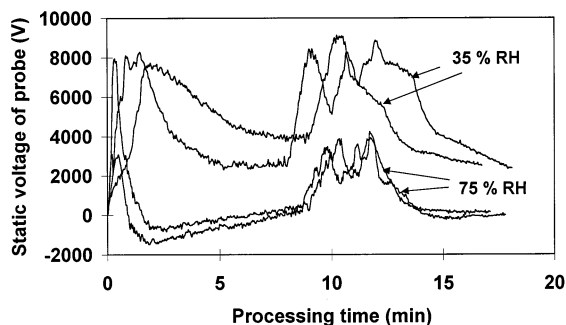

Fig. 3. Effects of the relative air humidity (RH) on the static voltage achieved during processing. Results of two repeated experiments at each humidity level are shown. Meltable binder: stearic acid.

Initially, variations in the level of static voltage and in the duration of the initial peak are seen between repeated experiments. However, the initial peak is maintained for a shorter time at the high air humidity, and a positive charge is obtained irrespective of the air humidity. For repeated experiments, a better reproducibility of the magnitude of the charges is obtained from the onset of the second peak. However, random variations in the dry mixing time give rise to a relative displacement of the curves, but the onset of the second peak in the voltage of the probe was always obtained at the same product temperature.

Generally, a considerable reduction in the level of static voltage is seen at the high air humidity. On prolonged massing, a charge reversal, i.e. reversal of charge polarity, is seen at the high humidity level. A high air humidity has previously been reported to reduce the tendency of electrostatic charging (Cartwright et al., 1982; Guardiola et al., 1996). The effect of a higher air humidity is ascribed to a creation of a thicker moisture layer on the particle surfaces and, in some cases, to the absorption of water vapor by the materials. The moisture layer provides a conducting path for the charges to leak away (Nguyen and Nieh, 1989; Schönert et al., 1996; Jonassen, 1998) and might decrease the resistivity of a material (Grosvenor and Staniforth, 1996; Wolny and Opalinski, 1996). Due to the high resistivity of stearic acid, the charge leakage becomes improved by the presence of a conducting moisture layer. For some materials, a charge reversal has been reported (Cartwright et al., 1982; Bailey, 1984). This charge reversal was found to be more likely at high air humidity and with high mechanical stresses.

The effect of the relative air humidity on the level of static voltage of the probe obtained with the binder mixture of stearic acid and glycerol monostearate is shown in Fig. 4. The admixture of glycerol monostearate to stearic acid did not affect the onset temperature of the two final peaks in the static voltage of the probe. Similar to the experiments with stearic acid, charge reversal and reduced levels of static voltage are achieved at the high air humidity. However, irrespective of the air humidity level, the admixture of glycerol monostearate to the stearic acid reduces the level of static voltage.
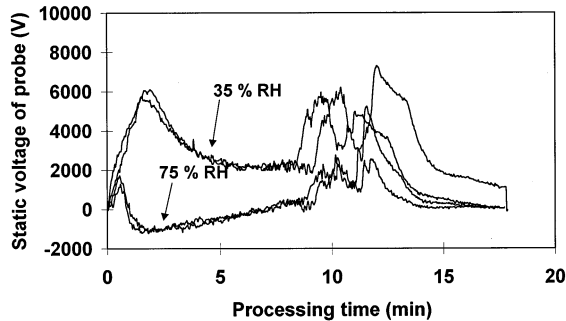
Fig. 4. Effects of the relative air humidity (RH) on the static voltage achieved during processing. Results of two repeated experiments at each humidity level are shown. Meltable binder: stearic acid/glycerol monostearate (20:3).

With PEG 3000, low levels of static voltage that are unaffected by the level of humidity are achieved (Fig. 5). Contrary to the other binders, charge reversal is seen with PEG 3000 irrespective of the relative air humidity. PEG 3000 has a low resistivity and will thus leak charge more readily than the other binders investigated. Therefore, charge leakage cannot be expected to become improved significantly by the presence of a conducting moisture layer.

By comparing Figs. 3–5, the level of static voltage achieved in the present experiments seems to be related to the resistivity of the meltable binder (Table 2). A high resistivity of the meltable binder increases the ability of the material to retain charge and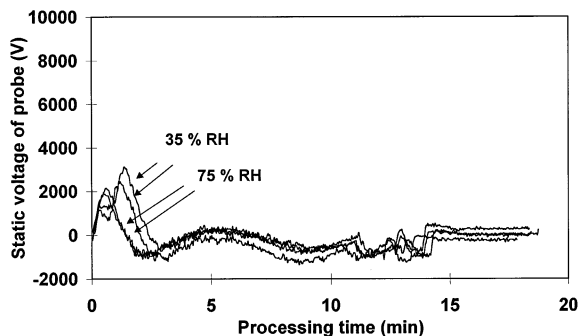, thereby, increases the accumu-lation of charge on the material. However, the actual magnitude of the charge will depend on an equilibrium between the charge generation and charge dissipation (Pavey, 1998). The viscosity of the meltable binder will influence the generation of charge by affecting the deformability of the binder and the agglomerates. A low binder viscosity increases the deformability (Schæfer and Mathiesen, 1996b) and, therefore, increases the available area for charge transfer upon contact (Carter et al., 1992). The viscosities of the molten binders and the binder mixture indicate that generation of charge is more likely with stearic acid and the binder mixture, because a larger contact area is established on collisions between particles. Additionally, the viscosity of the meltable binder affects the agglomerate growth rate and thus the size of the particles. Agglomerate growth will reduce the surface area available for frictional forces. This effect is complex because the initial growth rate has been found to be favored by a low binder viscosity, whilst the subsequent growth was favored by a high viscosity (Schæfer and Mathiesen, 1996b). With stearic acid and the binder mixture, the initial growth rate is expected to be higher, and the increased size of the agglom-erates might counteract the effects of the higher deformability. Since the stearic acid and the binder mixture have a similar viscosity and cause agglomerates of a similar size (Table 3), it is most likely that the lower static voltage achieved by the binder mixture can be attributed to the lower resistivity.

Binder characteristics such as the viscosity that affect the generation of charge and binder charac-teristics such as the resistivity that affect the dissi-pation of charge should, therefore, be taken into considerations for a more accurate prediction to be made of the effects of the meltable binder on the level of static voltage achieved during melt pelletization.

3.4. Adhesion

Electrostatically charged particles may adhere strongly to either neutral or oppositely charged surfaces (Bailey, 1984; Pavey, 1998). Adhesion can be caused, however, by a number of mecha-

Fig. 5. Effects of the relative air humidity (RH) on the static voltage achieved during processing. Results of two repeated experiments at each humidity level are shown. Meltable binder: PEG 3000.

Table 3
Effects of the type of binder and the relative air humidity (RH) on the mean granule size (d_{gw}) and size distribution (s_g)

	Meltable binder					
	Stearic acid		Stearic acid/glycerol monostearate (20:3)		PEG 3000	
	35% RH	75% RH	35% RH	75% RH	35% RH	75%RH
d_{gw} (μm)	1602; 1436	1122; 1357	1290; 1414	1222; 1430		
d_{gw} (μm)[a]	1204; 1169	1187; 1297	1374; 1235	1295; 1374	378; 377	360; 383
s_g	1.43; 1.49	1.53; 1.47	1.51; 1.45	1.49; 1.52		
s_g[a]	1.54; 1.60	1.50; 1.54	1.50; 1.56	1.61; 1.55	1.86; 1.84	2.00; 1.90

[a] Measurement of static electricity during the process.

nisms such as electrostatic forces, van der Waals' forces, and liquid bridges. In melt agglomeration, each of these mechanisms are likely to affect the amount of adhesion to the bowl. The predominant mechanism will depend on the nature of the materials and on the processing conditions.

Fig. 6(a) shows that a considerably higher amount of adhesion is obtained for stearic acid compared to PEG 3000 and the binder mixture of stearic acid and glycerol monostearate. At each humidity level, a higher level of static voltage was found for stearic acid. With stearic acid at 75% relative air humidity, however, a lower level of static voltage was obtained compared to the binder mixture at 35% relative air humidity (Figs. 3 and 4). The differences in the amount of adhesion to the bowl obtained for the different binder types, therefore, cannot be explained solely by differences in the level of static voltage.

In an experiment where the measurement of static electricity is conducted, the monitoring probe might induce corona discharges (Kassebaum and Kocken, 1997). In order to investigate the effects of the monitoring probe on the amount of adhesion, experiments were also performed in which the measurement of static electricity was not conducted. In those experiments, the monitoring probe was dismounted. In previous melt agglomeration experiments with lactose monohydrate and PEG 3000 in the same mixer without a probe (Schæfer et al., 1993a), the amount of adhesion was found to be similar to the 5% obtained in the present study with the probe present. With PEG 3000, it is therefore

assumed that the amount of adhesion is unaffected by the presence of the monitoring probe. Consequently, only stearic acid and the binder mixture were applied as the binders in the experiments in which the probe was dismounted.
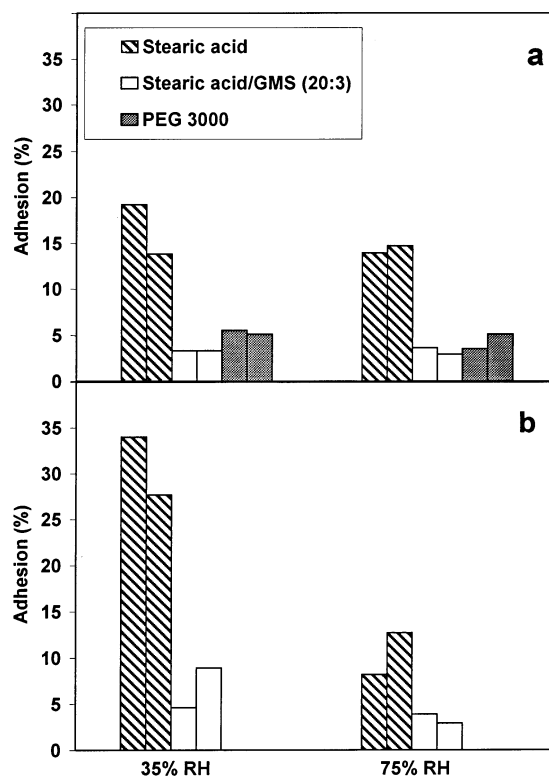

Fig. 6. Effects of the relative air humidity (RH) on the adhesion of material to the bowl lining. Results of two repeated experiments at each humidity level are shown. (a) Probe present; and (b) probe dismounted.

A considerably higher amount of adhesion to the bowl is obtained with stearic acid in experiments without the monitoring probe (Fig. 6(b)) compared to experiments with the monitoring probe. However, this effect is seen only at the low air humidity. With the binder mixture, the absence of the monitoring probe did not clearly alter the amount of adhesion to the bowl. Charge accumulation can be expected to increase in experiments where the monitoring probe is absent, because charge neutralization due to corona discharge is hindered. Therefore, the electrostatic forces are assumed to become higher and more important in relation to the adhesion.

The adhesion caused by electrostatic forces is counteracted by gravitational forces and by a detachment of particles adhering to the wall by the moving material. A certain magnitude of the electrostatic forces, therefore, will be necessary to cause adhesion. In experiments with the binder mixture and PEG 3000, the level of static charges on the material is assumed to be below this threshold level independent of the air humidity and the presence of the monitoring probe. Consequently, the amount of adhesion seen is low and is caused by other mechanisms than electrostatic forces. With stearic acid, the threshold level is exceeded, and the amount of adhesion, therefore, depends on the actual level of static voltage and thus on the air humidity as well as on the presence of the monitoring probe.

It is seen from Table 3 that the mean granule size and the size distribution of the agglomerates are not clearly affected neither by the air humidity nor by the measurement of static electricity during the process. Further, there was no indication of differences in the intragranular porosity of agglomerates prepared from the same binder type. This indicates that the agglomerate growth is unaffected by the differences in the level of static voltage achieved in the present experiments.

PEG 3000 results in a markedly lower mean granule size and in a wider size distribution than stearic acid and the binder mixture, because the amount of meltable binder was kept constant without any attempts to optimize the binder concentration for each binder.

In the present experiments, the amount of adhesion was sufficiently low to enable melt pelletization with stearic acid in contrast to the findings of other authors (Schæfer et al., 1990; Thomsen et al., 1994). Preliminary experiments indicated that less adhesion to the bowl was obtained, and melt pelletization was enabled with a low impeller speed at a product temperature within or just above the melting range of the stearic acid. Less adhesion was probably obtained in the present experiments, because a low temperature within the bowl increases the relative humidity within the bowl. This will reduce the level of static voltage and thus the amount of adhesion to the bowl. Accordingly, the higher product temperature in the experiments of Thomsen et al. (1994) is assumed to have caused a higher level of static voltage giving rise to so much adhesion that a pelletization with stearic acid was hindered.

4. Conclusions

The resistivity of a meltable binder seems to be related to the hydrophilicity of the binder since a low resistivity was found with the hydrophilic PEG 3000, whilst a higher resistivity was found for the more hydrophobic binders glycerol monostearate and stearic acid. In a melt granulation process in a high shear mixer, a high level of static voltage was obtained when a meltable binder with a high resistivity was applied.

Charged particles might adhere strongly to opposite charged and neutral surfaces. Therefore, adhesion of material was found to increase when high levels of static voltage were obtained during processing. This explains why the PEGs generally have been found to cause less adhesion than the hydrophobic meltable binders. A marked adhesion of material to the bowl might result in an unacceptably low yield of the process and might hinder the helix-like movement of the mass that is a prerequisite of pelletization.

The relative air humidity within the bowl will be markedly lower in a melt agglomeration process compared to aqueous wet granulation, because the temperature is higher and no water is added. Adhesion problems related to static elec-

tricity, therefore, will be more pronounced in melt agglomeration processes. In order to reduce adhesion caused by static charges, a meltable binder with a low resistivity should be applied if possible. However, a binder with a low resistivity might not be suitable in the formulation of a prolonged release product if binders of a low resistivity are generally hydrophilic. If a meltable binder with a high resistivity has to be used, the electrostatic charging of the particles has to be reduced. This can be done by inserting a grounded probe within the mixing bowl, or the process can be performed at a high relative air humidity.

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