Chemical Engineering Journal 83 (2001) 1–5

www.elsevier.com/locate/cej

Experimental investigation of droplet deposition on a single particle

Rames C. Panda [a,*], Jesko Zank [b], Holger Martin [b]

[a] *Chemical Engineering Division, CLRI, Adyar, Chennai 600020, India*
[b] *Instituet fuer Thermische Verfahrens Technik, Universitaet Karlsruhe (TH), D-76128 Karlsruhe, Germany*

Received 4 January 1999; received in revised form 24 January 2000; accepted 26 January 2000

Abstract

Spray coating or granulation is used to produce coarse granular solid particles by spraying solutions or suspensions in the form of fine droplets on fluidized particles followed by drying in a stream of fluidizing air. The quality and thickness of granulation or coating on the particle should be uniform. A part of the liquid-droplets from the spray-nozzle collide with the particle, of which, a part bounces back and the remaining part adheres on the surface of the particle giving layered-growth. In this work, the granulation process is carried out on a single spherical particle. Experiments are conducted to study the influence of process parameters, drying conditions, impact velocities and physical properties of sprayed solutions on the kinetics of granulation and on the morphology of the end product. A change in the drying condition changes the viscosity and surface tension of the sprayed solution. With increase in droplet velocity, the growth rate decreases. The results are useful to model the droplet deposition behaviour in fluidized bed granulation. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Coating; Growth mechanism; Fluidized bed; Spray granulation

1. Introduction

Coarse granular solid particles are produced from raw materials like solution, suspension or molten liquid in a fluid bed spray granulator. The particle growth takes place simultaneously under spraying and drying. After achieving a desired size, the product is taken out of the bed. The size of end-product varies from 0.3 to 10 mm.

Fig. 1 explains the principle behind the fluid spray granulator. The liquid to be coated is atomized and is sprayed as fine droplets on fluidized particles. Some of the droplets after colliding with particle, adhere and get deposited on the surface of the particle giving rise to layered-growth. The fluidizing hot air is used to dry the particle. The particle moves randomly between humidifying and drying zones in the bed with simultaneous material and energy transfer. In a continuous process, a part of the granulated product follows the classifier directly whereas the other part comes through cyclone separator to the classifier directly according to their sizes.

As described by Uhlemann [1], the particle acts as a nucleus for granulation. Loeffler [2] explained dust-deposition in different filters and the centrifugal process. Schubert [3] reported capillary mechanism in the drying of solid particles. Kleinbach and Riede [4] optimized the process parameters for the layered-growth on solid materials. Link [5] investigated the granulation mechanism on a single particle. But the mechanism of droplet deposition followed by coating and granulation is not yet fully understood. In this work, solutions of different solids are sprayed as fine droplets on the surface of a single fluidized particle which in turn develops a layered-growth (through subsequent drying). This process is used to study the physical mechanisms behind granulation. This will help to understand the behaviour of process parameters in real spray-granulation process in fluidized bed with many particles.

2. Experimental

2.1. Scope

Aqueous solutions of sodium chloride (20%), calcium chloride (20%), lactose (16%) and maltodextrine (20%) are sprayed on a single floating spherical aluminium particle (diameter = 1.3 mm) to investigate the droplet deposition behaviour in spray-granulation process. The influence of droplet velocities and fluidizing air temperature on growth kinetics of the particle is studied. A pinch of polymer, namely, polyvinyl pyrolidon (PVP) or carboxymethylcellulose (CMC) is mixed with aqueous lactose solution to study the viscous effect on growth kinetics. The morphology of the surface of granulated particles are examined under

---

[*] Corresponding author.

PII: S1385-8947(00)00164-9

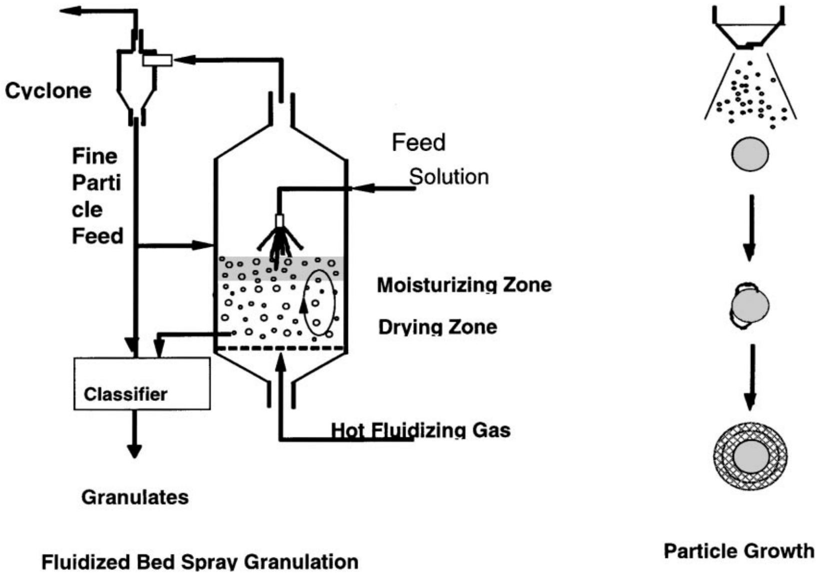

Fig. 1. Schematic representation of fluidized bed spray-granulation process.

scanning electron microscope (SEM). The densities and viscosities of these solutions under present study are measured with the help of densitimeter and falling-ball-viscosimeter, respectively.

2.2. Experimental set-up

The experimental set-up is shown in Fig. 2. In a glass cylinder of 150 mm diameter, a nozzle carries the drying air stream V_{kugel} upwards, whose temperature through the controlled heating H_{kugel} can be adjusted. This installed controlled-thermo element is enclosed and protected from liquid-droplets by Teflon tape.

The sample-nucleus, made of aluminium and of about 1.3 mm diameter is suspended in the stream flow coming out of the nozzle. Though collision of droplets on the nucleus gives perturbation in its equilibrium position, it is kept floating in the stream. The solution is transported from a storage tank (cell) through a micro-dosing pump to an ultrasonic atomizer suspended from the top of the granulation chamber and is atomized. The originated droplets are accelerated by projecting in the carrier air stream. The carrier air stream

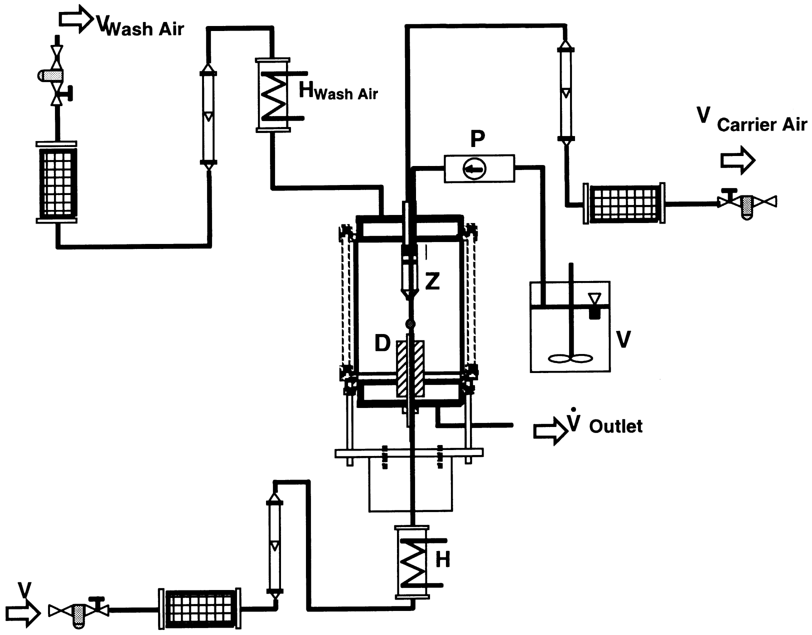

Fig. 2. Experimental set-up to study spray granulation.

is used to accelerate the coagulated/agglomerated droplets (of about 35 μm). The droplets which do not collide with the nucleus, are removed and carried away by a hot stream of air. The temperature of all inlet and exit air streams are measured and the signals are connected to a recorder.

2.3. Experimental procedure

At first, the solution to be granulated is prepared by weighing and mixing the compositions correctly. The operating parameters are tuned. The fluidizing air stream is so tuned that the floating particle is stable. The distance of the particle from nozzle is maintained at about 5 mm. The washing stream of air (to remove dust from chamber) is adjusted to 200 l/h and the spray stream of air (carrying the droplets) is tuned to its desired value. About 4×10^{-6} l of liquid is atomized and sprayed (in the chamber) in each time interval through a timely pulsed (once in every 5 s) micro-dosing pump. With spraying and drying, the fluidized particle develops a layer of coating on its surface. At the end of 200th spray, the dosing pump is stopped and the coated particle is dried in the same stream of air for 3–5 min. The coated particle is then taken out of the compartment and the amount of coating is gravimetrically determined. The particle is again brought back into the stream of air coming through the nozzle. Spraying is done again and is repeated at the end of 400th and 600th spray. The amount of growth is estimated and is plotted against the number of sprays. The result shows that there is enough deposition confirming sufficient coating of material on the surface in 600 sprays. The linearity of the curves (plot of coated weight against number of spray; not shown here) also reveals that building up of layer is not remarkably influenced by the physical properties of the material. Hence the growth rate is defined as the amount of material deposited (dried) per stroke of pump or sprayed liquid (BSP). Each experiment was repeated at least three times. Regression analysis was carried out after calculating the mean value, slope and standard deviations.

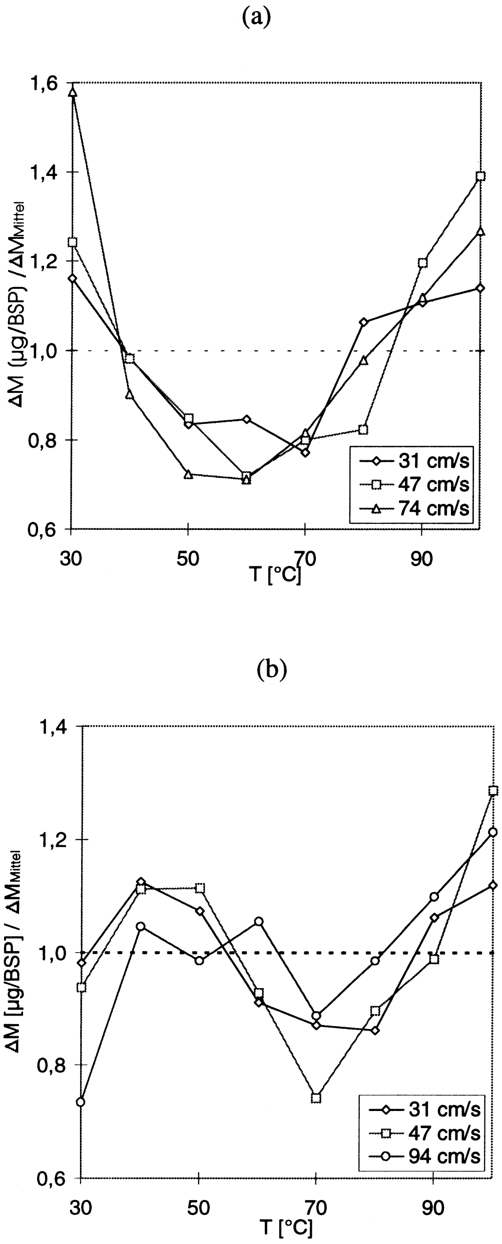

Fig. 3. Influence of fluidizing air temperature on particle growth for different droplet velocities with: (a) 20% aqueous sodium chloride; (b) 16% aqueous lactose solution.

3. Results and discussion

3.1. Influence of droplet velocity and particle temperature

The influence of process variables on droplet deposition is studied by varying droplet velocities or fluidizing air temperature. The droplets are supposed to have a unique velocity (actually there exists a velocity distribution). Though the growth rate does not vary abruptly with droplet velocity, significant change in growth rate is observed with change in fluidizing temperature.

Fig. 3a and b show the specific growth rate (which is equal to the ratio of growth of particle (coating) per spray to its mean value) under varying fluidizing temperature conditions with spray of sodium chloride and lactose solutions, respectively. Under different droplet velocities (31, 47 and 74 cm/s) the experiments are conducted. In Fig. 3a, it may be clearly seen that, in the case of sodium chloride, the specific growth rate shows a positive deviation from its mean value in lower and higher temperatures signifying more uncertainty in achieving expected granule size. (The minimum growth was found at about 40°C and lies in the range 2–4.2; the maximum growth was found at 80°C and lies in the range 3–5.3.) The graph also gives an idea of probable product sizes at different fluidizing temperatures and with different droplet velocities. With higher droplet velocities and lower fluidizing temperature, the size of the granule becomes larger than expected. From Fig. 3b, it is evident that coating with aqueous lactose solution at lower temperature and with higher droplet velocity produces lower size of particle than the expected mean. (The minimum growth was found at about 70°C and lies in the range 4–2.8; the maximum growth was found at 50°C and lies in the range 5.8–3.) It is observed that uncertainties in getting a desired size of product are more in the case of sodium chloride than in lactose. At higher temperatures, the deposition is maximum and it is minimum at about 60–70°C. It is also observed that with increase in fluidization temperature, the dust production due to overspray also increases inside the fluidization chamber.

The results with lactose are similar to Link [5]. The surface tension of lactose solution on the surface of particle is a function of temperature and shows optimal behaviour at around 50°C. At higher temperature the surface properties increase and an amorphous lactose is observed. But no such optimal behaviour is observed in the case of sodium chloride solution. At lower temperature, the growth rate is low due to higher wettability and with increase in temperature of fluidizing air, the droplet deposition increases. Fig. 4 shows the effect of droplet velocities on the mean values of growth rates per spray. It can be observed that the droplet deposition decreases with an increase in droplet velocity. Only a few of the liquid-droplets collide the fluidizing particle. A part of the colliding droplets reflect back from particle surface and the others adheres on surface. The droplets that bounce back from particle surface increase with increase in velocity of droplets and hence lesser deposition. These results are helpful to understand the adhesion characteristics of materials in droplet deposition. The effect of temperature for a droplet velocity (74 cm/s) is also studied for aqueous solutions of 20% maltodextrin and 20% calcium chloride. Although the effect is less in case of maltodextrin as compared to the case of lactose, the growth rate changes in a similar way. Due to hygroscopicity of calcium chloride, the investigation was carried out from 60°C onwards. The growth rate is observed to be relatively higher at 80°C (similar to sodium chloride).

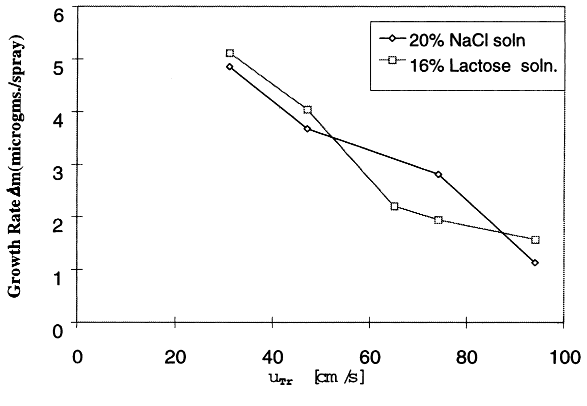

Fig. 4. Influence of droplet velocity on particle growth.

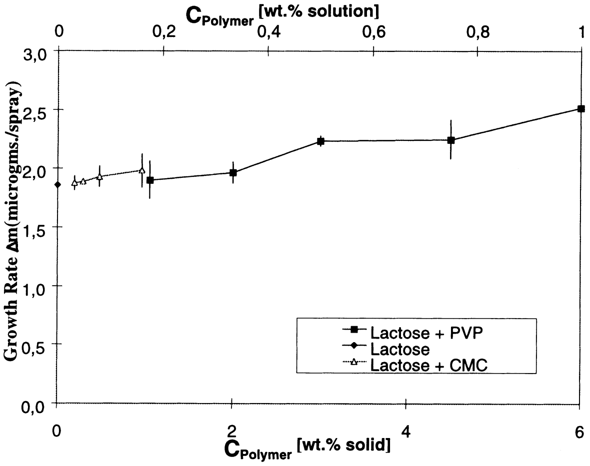

Fig. 5. Effect of viscosity of spraying liquid (by adding a binder material) on particle growth rate (fluidization temperature 80°C, droplet velocity 74 cm/s).

3.2. Addition of binders

Addition of binders changes the viscosity of spraying liquid. A change in the amount of droplet deposition on the particle is therefore expected. PVP was used as binder material to lactose. The dependence of growth rate on concentration of binder materials may be seen in Fig. 5. It is clearly seen that the addition of polymer substances increases the droplet deposition slightly. The cause could be the increase in viscosity of spraying solution due to addition of polymers. Link [5] showed that the critical velocity of droplets above which the droplets bounce back from the surface, is dependent on surface-contact angle δ, viscosity of droplet and diameter of droplet. An increase in the viscosity of solution, increases the critical velocity and thereby increases energy of dissipation. To prove this, CMC is added with aqueous solution of lactose to increase its viscosity. Slightly more deposition of material is observed (Fig. 5). Addition of binder changes physical properties of material like surface energy significantly as observed by Reyza [6].

Present experiments show that the growth rate increases almost linearly with increase in polymer concentration. With a maximum concentration of PVP (C_{PVP}=1% with viscosity 1.8 cp at 20°C) or CMC (C_{CMC}=0.3% with viscosity 28 cp at 20°C) in the solution of aqueous lactose, the viscosity of solution is increased by a factor of 1.2 or 20 (over aqueous lactose 16%), respectively. Although the increase in viscosity by CMC is significantly high, the slope of growth rate is small. This concludes that the addition of PVP also causes a change in surface properties. To clearly understand the effect of viscosity on growth rate, experiments related to physical properties of materials (specifically surface properties like surface tension, dynamic contact angle) are essential in order to formulate a mathematical model.

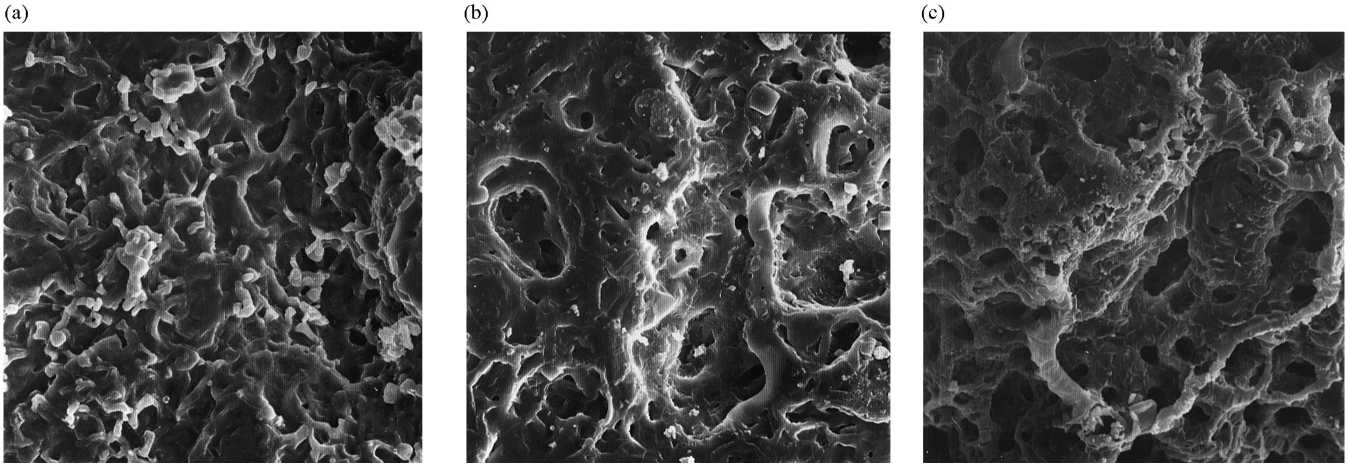

Fig. 6. Morphology of particles (sodium chloride coated — left: at 30°C; middle: at 50°C; right: at 80°C, with a droplet velocity of 74 cm/s in both cases) (magnification: 120 μm).

3.3. Particle morphology

Effects of process parameters on surface morphology of granulated particles are observed under SEM. Granulation at lower temperature shows crystalline structure of lactose whereas at higher temperature the morphology is amorphous. In the case of sodium chloride (Fig. 6), strong porous structure is observed which changes with drying conditions. The porosity increases with higher drying temperatures.

This is in close resemblance with the results of Link [5] as at higher temperature the surface tension increases the growth rate as revealed in Fig. 3a. Conversely, granulation at lower temperature does not show any enhancement of porosity.

4. Conclusion

The experimental results show that the droplet deposition is specifically dependent on physical properties of the material. The drying temperature and droplet velocity influence the granulation kinetics. These results for a single particle are useful in case of fluidized bed spray granulation. Addition of binders changes the physical properties of the solution and also influences droplet deposition. Change of surface tension properties also play an important role in coating process. The effect of viscosity of solution on the spray-granulation process is supposed to be less significant. These results are useful to select the operating variables and to scale up the process.

Acknowledgements

The first author wishes to acknowledge the financial assistance provided by DAAD in carrying out this research work.

References

[1] H. Uhlemann, Kontinuierliche Wirbelschicht Spruehgranulation, Chemie-Ingenier-Technik 62 (1990) 822–834.
[2] F. Loeffler, Staubabscheiden, Georg Thieme, Stuttgart, New York, 1988, p. 171.
[3] H. Schubert, Grundlagen des Granulierens, Chem. Ing. Technol. 51 (4) (1979) 266–277.
[4] E. Kleinbach, Th. Riede, Coating of solids, Chem. Eng. Process. 34 (1995) 329–337.
[5] K. Link, Wirbelschicht Spruehgranulation: Untersuchung der Granulatbildung an einer frei schwebenden Einzelpartikel, Dissertation, Universitaet Karlsruhe (TH), Karlsruhe, 1996.
[6] J.A. Reyza, Reibungsdruckverlust bei der Gas-Fluessigkeits-Zweiphasenstro-emung in waagerechten Rohren mit kreisfoermigem und ovalem Quer-schnitt, Dissertation, Universitaet Karlsruhe (TH), Karlsruhe, 1985.