Particle Growth in Fluidised Bed Granulation

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A series of batch experiments are carried out in a fluidized bed granulator with malic acid to understand the growth mechanism and the growth rate of particles with respect to the operating parameters such as the flow rate of the spray solution, temperature and flow rate of the fluidizing air, the concentration of the solution and the seed particle diameter. The increase in flow rate or the increase in concentration of the spray solution is found to increase the growth rate of particles. The increase in temperature and flow rate of the heating medium does not affect the growth rate of particles, however it indirectly helps the process by facilitating to operate at higher liquid flow rates. The seed particle diameter has a major role to play on deciding the particle growth mechanism. The larger seed particles favour coating process, whereas the smaller particles favour agglomeration. Since smaller seed particles tend to agglomerate and grow faster, higher solute throughput could not be achieved. Hence a special type of agitator is designed to promote coating phenomena with smaller seed particles. Thermal efficiency of around 80 % is achieved.

Keywords:
Fluidization, granulation, particle growth, coating, agglomeration

Introduction

Fluidized bed granulation drying has established itself as a thermal treatment process for granular solids due to its improved heat and mass transfer characteristics. Fluidised bed granulation is a process of converting liquid products such as suspensions, solutions or melts into granular solids. The process of granulation is governed either by coating or agglomeration, which depends upon the operating conditions and the physico-chemical properties of the raw material. The coating process is mainly utilised for modifying the surface properties of the material, which finds wide application in pharmaceutical industries. The second operation agglomeration serves the purpose of granulating fine powders with the use of binders there by reducing dust and explosion risks, to improve the flowing properties and to provide ease of handling, which finds wide application in industries, ranging from the pharmaceutical to fertilizer and mineral-processing industries.

The liquid to be granulated is usually sprayed with a jet into a fluidized bed composed of hot solid particles, whereby a portion of the liquid forms a precipitate on the particles. The spraying can occur in the fluidized bed from the top down, from the bottom up or sideways with a jet submerged in a chosen position.

The mentioned earlier, particle growth is governed by two mechanisms: coating and agglomeration. During coating, the solute in the solution is deposited on the surface of solid particles forming a thick layer. This mechanism is observed when the wetted particles can be dried before their collision or when the cohesive strength is weak. During agglomeration, large particles or granules are produced by smaller particles adhering to one another via liquid bridges. The solidification of liquid bridges through drying leads to the formation of solid granules.[1]

Control of fluidized bed granulation process is difficult as it involves simultaneous wetting, drying and mixing of particles. If the liquid is excessive or if it is maldistributed, large region of the bed may defluidize and particles stick together as large wet lumps, called wet quenching. On the other hand, if excessive particle growth occurs the minimum fluidization velocity (which is proportional to square of the particle diameter) will exceed the operating velocity, which leads to defluidization, called dry quenching. A thorough understanding of the particle growth mechanism in fluidised bed granulation process is very important to control the process effectively.

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Number of work has been reported in literature covering the various aspects of fluidised bed granulation process. *Smith* and *Nienow*[2,3] reported that the spray liquid form a cold wetting zone only at the spraying site, and that this zone exchanges particles with the surrounding hotter mixing zones, whereby, a heating up zone was discovered directly above the air distribution plate. *Maronga* and *Wnukowski*[4] observed the gas humidity and temperature to be the important parameters influencing the coating process based on experimental results for a top spray particulate coating, using a single non-submerged spray nozzle. *Pont* et al.,[5] studied the effects of physicochemical properties, such as the viscosity and wettability of the granulating liquid on solid particles surfaces, on the agglomeration kinetics of solids particles and reported the dominant force in granulation process to be the capillary forces. The viscosity of solution was reported to have less effect on growth of agglomerates than interfacial tension and contact angle. *Becher* and *Schlunder*[6] reported significant reduction in the agglomerate formation, if a two fluid nozzle spraying upwards, is used. *However Vinter*[7] reported that the nozzle at the top of the bed favoured coating. *Legler*[8] reported that atomizing air does not influence as long as proper fluidization is achieved. However *Schaefer* and *Worts*[9], *Davis* and *Gloor*[10] reported decrease in the granule diameter with increase in atomization air pressure. *Scott* et al.,[11] and *Davies* and *Gloor*[10] reported a larger granule diameter with increase in the binder addition rate.

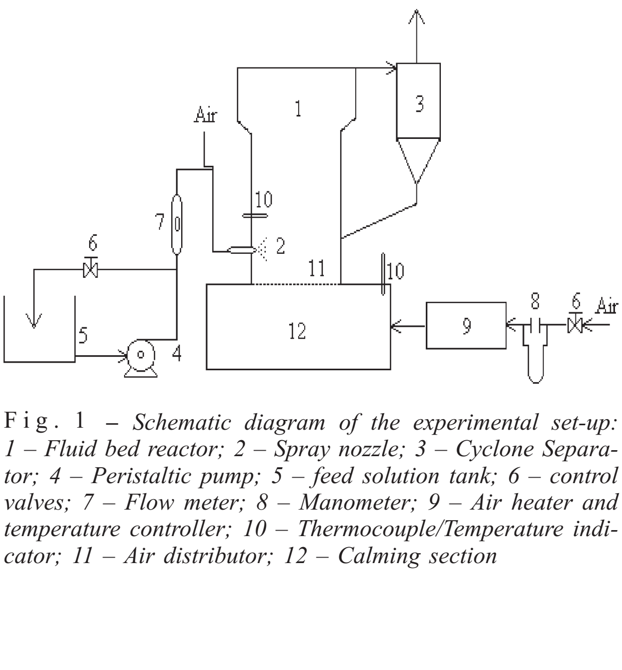
In general, bulk of the earlier research was focused on various factors that influence the growth mechanism and kinetics, in which a solution is sprayed on to a inert material of (sand, glass bead etc.) or material other than the solute with the help of binders. Fluidised bed granulation, in particular to granulating a solution on the same material as that of solute, has received far less attention from the scientific community, in spite of its unique capability to bridge crystallization, centrifuging and drying into a single unit operation.

Malic acid is one of the popularly used food acids, which is also called apple acid, finds wide application in food industries as preservative and taste enhancer. The high solubility of malic acid leads to low temperature crystallization and recycling of large volumes of low concentration mother liquor. An attempt has been made to utilize fluidised bed granulation process as an alternative downstream processing for production of malic acid. The concentrated malic acid solution from the concentrator is sprayed on to the malic acid crystals previously obtained from crystalliser. The main objective of present study is to replace the conventional crystalliser with a batch fluidised bed agglomerator, which necessitates achieving high throughput ratio (quantity of material obtained after granulation to the initial quantity of seed material taken). This requires the coating granulation process to be predominant so, that excessive particle growth can be arrested and high throughput ratio could be achieved.

In order to achieve the above goal, it is essential to understand the mechanism of particle growth, of malic acid in batch-fluidised bed. Hence the effects of various parameters such as the flow rate of spray liquid, fluidizing gas temperature, fluidizing gas velocity, concentration of spray liquid and seed particle diameter on the growth rate of particles is studied. Further to identifying conditions that favour coating rather than agglomeration, the study develops a special agitator, which aids the particle growth mechanism to follow coating rather than agglomeration.

Experimental

The experimental set-up (Figure 1) consists of a fluidization column of 148 mm internal diameter and a height of 800 mm. The column is made of Perspex materials 5 mm thick. Fluidization column is connected to a cyclone separator where the entrained fines are separated from the fluidizing air and returned to main section. A distributor plate made of stainless steel with 2 mm perforation and 13 % free area is fitted at the bottom of the fluidization column. The perforated plate is covered with fine wire mesh to avoid fine particle chocking in the perforation holes. Calming section is provided at the bottom of the fluidization column for proper air distribution. A pneumatic spray nozzle (two fluid nozzle) is fitted through the sidewall at a height of



F i g . 1 – *Schematic diagram of the experimental set-up: 1 – Fluid bed reactor; 2 – Spray nozzle; 3 – Cyclone Separator; 4 – Peristaltic pump; 5 – feed solution tank; 6 – control valves; 7 – Flow meter; 8 – Manometer; 9 – Air heater and temperature controller; 10 – Thermocouple/Temperature indicator; 11 – Air distributor; 12 – Calming section*

60 mm from bottom of the distributor plate. Air heater with 8, 12, 20 kW heating coils and a temperature controller is provided along the airline. The temperature of the heating medium can be controlled within ± 0.5 °C. The pressure drop across the bed is measured using pressure tapings. The exit gas temperature from the bed is obtained using a Cr–Al thermocouple connected to a digital thermometer (Doric 450). The measurements are obtained at several points from the centre of axis of the bed to the bed wall. At those points where fluctuations are observed, both the minimum and maximum values, are recorded. Based on the inlet air humidity and temperature, the corresponding wet bulb temperature is read from the psychrometric chart, which served as a guide to estimate the thermal efficiency of the bed.

As discussed in the earlier section, new type of agitator (Figure 2) has been used in the present investigation to reduce the particle agglomeration and to enhance the coating phenomena. The agitator assembly consists of a central rod fitted with limbs (cylindrical pieces of 5 mm diameter and 40 mm length), which are placed at distance of 10 mm from each other. Limbs of same dimensions are welded along with the bottom air distributor plate at a different angle so that the direct impingement of the spray is avoided. Clearance of 5 mm is expected when agitator is rotated between the limbs fitted with the bottom air distributor plate.

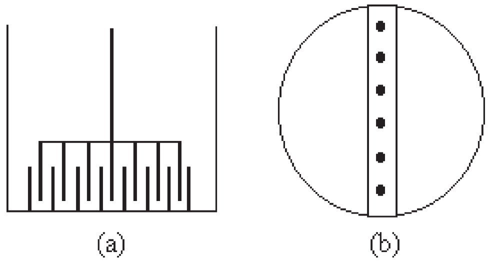


Fig. 2 – *Schematic diagram of agitator assembly a) Agitator; b) Fluidising air distributor plate*

A known quantity and diameter of seed particles is charged into the fluidized bed. Air at desired temperature and flow rate is admitted in the calming section. The bed particles are preheated before the liquid solution is sprayed on it. The liquid solution is sprayed over the bed particles with the help of spray nozzle. The atomization air and liquid solution flow rates are adjusted to get fine spray. Samples are collected at regular intervals of time, subjected to sieve analysis to obtain the particle size distribution, and the mean size is calculated from the distribution. Good fluidization is maintained throughout the processing time.

Results and discussion

The pressure drop across the fluidized bed is related to bed density using the fluidisation equation,

$$\Delta p = G/A = m\,g/A = 1 \cdot (1 - \varepsilon)\,(\rho_s - \rho_g) \cdot g \quad (1)$$

Equation (1) and a simple mass balance, from the amount of solution sprayed, are utilized to monitor the increase in solid content of the bed during granulation. At the end of each experiment, solids in the bed are weighed to cross check the prediction using equation (1).

The bed material is allowed to attain equilibrium temperature with the inlet fluidizing air, before the liquid solution is sprayed on to particles in the bed. The solution is sprayed on to the bed material continuously until the fluidizing air is not sufficient to fluidise the particles in the bed (dry quenching). However, some of the authors reported intermittent spraying in order to maintain proper fluidization.[8] The bed is observed to fluidise well with continuous spraying of solution in the present study, until beginning of defluidisation due to excessive particle growth.

The increase in flow rate of liquid solution reduces the exit temperature of heating medium there by increasing the thermal efficiency of the bed. A thermal efficiency of up to 80 % could be achieved maintaining good fluidisation condition. Thermal efficiency is defined as,

$$\eta = (T_i - T_o)/(T_i - T_w) \quad (2)$$

Beyond this point, the bed is found to quench due to excessive loading of moisture into the bed (wet quenching).

The growth rate is found to increase with increase in flow rate of the liquid solution (Figure 3).

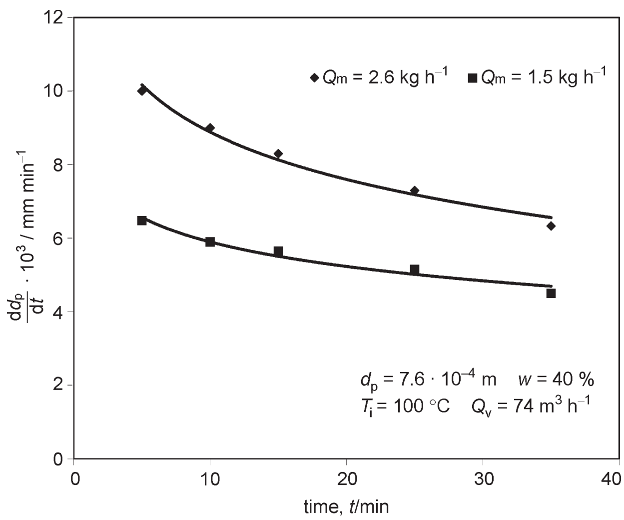


Fig. 3 – *Influence of the flow rate of the spray solution on the growth rate of particles*

The increase in flow rate of the solution leads to higher amount of solids precipitation over the existing solids into the bed, resulting in higher growth rate. Sieve analyses of the samples withdrawn from the experiments show, a remarkable uniformity in size, with 90 to 95 % of the solids retained in single size range.

Scanning Electron Micrography (SEM) studies were conducted on the granulated particles by many authors,[5,6] and reported that a layered growth granulated material to have a uniform and spherical surface, as compared to non-uniform agglomerate, in case of agglomerated product. In the present study as seed particles are sufficiently large enough, physical observation of the product is found sufficient to identify, the growth mechanism. The samples show a layered growth granulation mechanism rather than agglomeration. Moreover, the growth mechanism is substantiated using simple material balance, taking into account the amount of solution sprayed into the bed and the growth of particles, assuming uniform distribution of the spray on all the particles and the coated part to have same density as that of the original material.

Neither increase in flow rate of the heating medium nor the temperature of heating medium has any effect on growth rate of the particles as the growth rate is purely governed by material balance, which is proportional to the amount of liquid solution pumped and growth mechanism in the bed. The higher temperature and flow rate of the heating medium facilitate to operate the bed at higher liquid flow rates without wet quenching.

Figure 4 shows an increase in growth rate of the particles with increase in concentration of the spray solution. The increase in concentration of solution reduces the quantity of water to be evaporated and increases the amount of solute coated over the particles. Physical observation of the samples for spray solution mass fraction of 30 % as well as 40 % shows a layered growth granulated product rather than agglomerates.

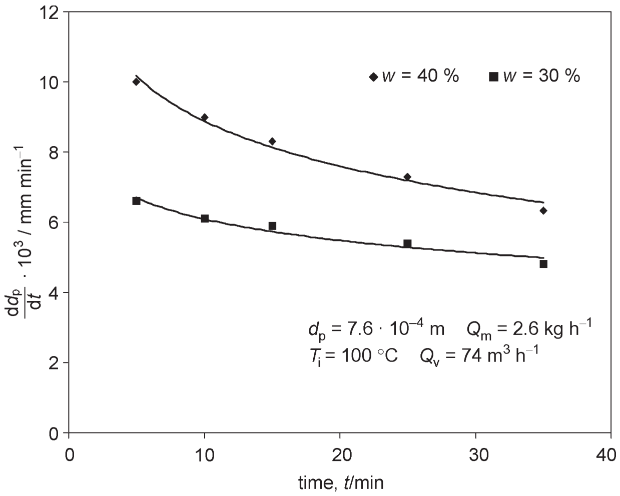


Fig. 4 – *Influence of the initial mass fraction of the spray solution on the growth rate of particles*

The higher seed diameter of the particles favor layered growth (Figure 3–4) rather than agglomeration as the binding forces between the particles is weak. This is favorable to achieve high throughput ratio, as the growth rate is much smaller as compared to agglomeration. However, a smaller initial seed diameter is favorable, since much higher throughput ratios can be obtained, as the surface area provided by smaller particles is much higher than the bigger particles, provided layer growth mechanism is maintained.

Figure 5 shows higher growth rate for smaller particles, as compared to bigger particles. If the layered growth mechanism is predominant, growth rate of the smaller particles have to be much lower than the bigger particles as the surface area provided by equivalent quantity of smaller particles is much higher. [Since binding force required for keeping particles together for smaller particles is much lower than that required for bigger particljes, the smaller particles tend to agglomerate easily and grow at much faster rate than bigger particles.]

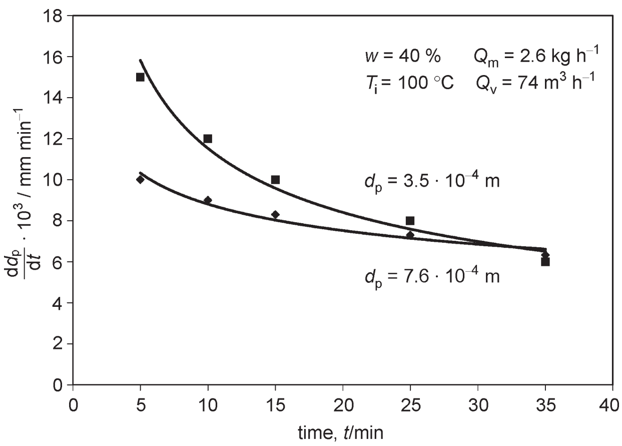


Fig. 5 – *Influence of the seed particle diameter on the growth rate of the particles*

In order to utilize the advantage of using smaller particles as starting material for achieving higher throughput ratio, an agitator has been designed (Figure 2). The agitation is introduced into the bed, primarily to break the agglomerates formed as and when it is formed.

Figure 6 shows lower growth rate for smaller particles as compared to bigger particles with the agitator. An agitation rate of 180 rpm is used. This was arrived after extensive preliminary experiments, in which a too low rpm result in agglomerated product or two high rpm result in a wide size distribution of the product. The growth rate is to-

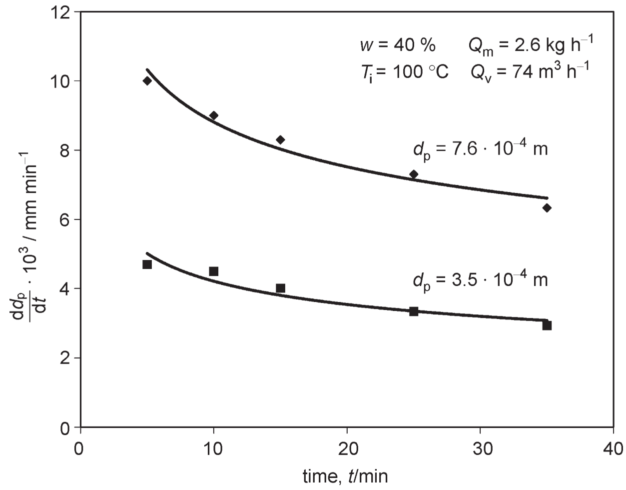


Fig. 6 – *Influence of agitation on the growth rate of the particles*

tally contrary to figure 5 where the growth rate of smaller particles is much higher than the bigger particles. The reverse trend in figure 6 clearly indicates that the agglomeration pattern, which is more predominant without agitator, is replaced by layered growth mechanism with the use of agitator. It can be construed that the agitation breaks the particles (agglomerates) rapidly, such that new surface is created, which takes up the precipitation of solute, there by controlling the growth rate of the seed material. Further, the physical observation of the product indicates the particles to be stronger and more spherical, as the layered growth mechanism is predominant. Hence with the smaller seed particle size, it is possible to achieve the maximum throughput ratio without excessive particle growth.

Conclusion

Experiments are conducted to understand the particle growth mechanism and kinetics of malic acid crystals in a batch fluidised bed granulator covering wide range in operating condition. An increase in the flow rate or an increase in the concentration of the spray solution increases the particle growth rate. Neither the increase in the flow rate nor the temperature of the fluidising air is found to alter the growth rate, however, they facilitate to operate the bed at high liquid flow rates without wet quenching. The seed particle diameter is found to have major influence on the growth mechanism of the particles. The larger particles favour layered growth, whereas the smaller particles favour agglomeration. In order to maximize the total solute throughput to the bed, smaller seed particles are favoured, as it provides larger surface area. Since smaller seed particles tend to agglomerate and grow faster, higher solute throughput could not be achieved. Hence a special type of agitator is designed to promote coating phenomena with smaller seed particles. Thermal efficiency of around 80 % is achieved.

Notations

A – cross sectional area of the fluidized bed, m^2

d_p – particle diameter, m^3

w_i – mass fraction of spray solution, mass of solute in mass the solution, kg kg^{-1}

Q_m – mass flow rate of spray solution, kg h^{-1}

Q_v – volumetric flow rate of fluidizing air, m^3 h^{-1}

$\frac{dd_p}{dt}$ – growth rate, mm min^{-1}

l – height of fluidized bed, m

m – mass of solids in the bed, kg

ρ_g – density of fluidizing air, kg m^{-3}

ρ_s – density of solids, kg m^{-3}

η – thermal efficiency

T_i – inlet temperature of fluidizing air, °C

T_o – outlet temperature of fluidizing air, °C

T_w – wet bulb temperature corresponding to the inlet air temperature and humidity

G – weight, N

g – gravity acceleration, m s^{-2}

p – pressure drop, N m^{-2}

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