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STUDIES ON GRANULATION IN A FLUIDIZED BED III. CALCULATION OF THE FEED RATE OF GRANULATING LIQUID

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In granulation in a fluidized bed, the appropriate selection of the feed rate of the granulating liquid is very important since this parameter, in addition to its effect on the formation of the granulates, considerably influences the capacity of the apparatus. The results of experiments on the effect of the feed rate of granulating liquid on the physical properties of the granulates formed in a fluidized bed (average particle size, the relative amount of the particles not granulated, wear strength, and the inhomogeneities of the binder distribution) are given. On the basis of the heat and liquid balances of the process, correlation is given for the maximum and equilibrium liquid feed rates.

A very important process parameter of the batch granulation in a fluidized bed is the feed rate of the granulating liquid. To attain the optimum average particle size, a well defined quantity of the given binder solution is needed. The liquid can be sprayed in the fluidized bed at different feed rates, i.e. during various lengths of time. Thus the appropriate choice of the feed rate of the granulating liquid, basically determines the duration of the granulation and hence the capacity of the apparatus.

In the first paper of this series [1] - where the relationships between the amount of the binder and the physical properties of the formed granulates were dealt with - the effect of the feed rate on the physical properties of the granulates was also touched upon to ensure the completeness of the paper. The results of other authors - RANKELL and all. [2], MÖBUS [3] and DAVIES and GLOOR [4] were also given. It was pointed out that the results were evaluated in literature in two different ways. Some authors changed the feed rate while keeping the duration of the spraying at a constant value [2, 3], while others kept the quantity of the granulating liquid constant [4] - hence the results are difficult to compare.

On the basis of the experimental results it was deduced that the average diameter and average porosity and the particle size distribution of the formed granulates were only slightly influenced by the variation within certain limits of the feed rate of the granulating liquid, provided that the overall amount of the binder was kept constant. Therefore, the influence of the feed rate was neglected in addition to those parameters that had greater effects on the above mentioned properties of the granulates.

In the present paper the influence of the feed rate of the granulating liquid on the average particle size, the relative amount of the particles not granulated, the wear strength and the homogeneity of the binder distribution will be dealt with in details. On the basis of the heat and liquid balances of the process, correlations were derived for the upper limit of the liquid feed rate and for the approximately optimum feed rate.

EXPERIMENTAL APPARATUS AND METHODS

The experimental laboratory granulating apparatus and the applied experimental methods will not be described here in detail, since they do not depart from those described in the previous paper of this series [1].The 0.1-0.2 millimetre fraction of quartz sand was used as basic material and an aqueous gelatine solution of a concentration of $c' =$ 60 kilograms per cubic metre was used as the granulating liquid. In the experiments dealt with in this paper, the concentration of the granulating liquid, the mass of the material to be granulated, the relative expansion of the fluidized bed, the temperature of the air at the inlet and the distance between the atomizer and the underplate were kept at constant values, in addition to the characteristics of the apparatus and the basic material and the binder. The air feed rate of the atomizer was increased with the feed rate of the granulating liquid to ensure a nearly identical liquid dispersion throughout the measurements, so that the specific air consumption was always about 2.5 kilograms air per kilogram liquid.

The test and calculation methods for the physical properties of the granulates formed were summarized in the previous paper of this series [1] while the test methods were detailed in the first paper [5].

In some cases the relative humidity of the air at the outlet and the liquid content of the bed were determined several times during the granulation at various liquid feed rates. The liquid content of the bed was determined by drying the samples taken from the bed in a drying oven until constant weight. The relative humidity of the air at the outlet was measured by an ASSMANN psychrometer redesigned to this purpose. These measurements had two objectives: they provided data for the establishment of the heat and liquid balances of the process and helped in deciding whether the change of state of the air flowing through the apparatus is really adiabatic as suggested in literature [2, 6].

At the two extreme values of the feed rate of the granulating liquid, the concentration of the binder was determined as a function of the size of the granulates. The average binder content of the granulates was determined by dissolving NaCl in the granulating liquid. Having concluded the granulation, the dry product was fractioned and from the average samples of identical mass, the labelled binder was extracted by hot distilled water. The conductivity of thermostated solutions of identical volumes of the samples were measured by bell-shaped electrodes. In the knowledge of the relationship between the conductivity and concentration of the NaCl solution, the binder content was determined from the measured conductivity.

EXPERIMENTAL RESULTS

The effects of the feed rate on the physical properties of the granulates were studied by altering the feed rate at four different values of the relative amount of the granulating liquid (V'/V = 5; 10; 20; 30 vol per cent) while keeping the other parameters at constant values. The studied feed rates were as follows: $w' = (2.5;\ 4.2;\ 5.9;\ 7.6$ and $9.2) \times 10^{-5}$ kilogram per second. When the feed rate is changed while the relative amount of the granulating liquid is kept constant, the duration of the granulation also changes,with smaller feed rate it is longer, and with greater feed rate is shorter. Because of the repeatability of the experimental results [1] three parallel experiments were carried out in every case so the dots in the following Figures correspond to the average value of three parallel experiments.

The average diameter of the granulates is plotted against the feed rate of the granulating liquid in Fig.1. The average particle size tends to be decreased by the increase of the feed rate, though the dots are rather scattered.(The straight lines in the Figure do not show anything more than the tendencies of the changes!) When the relative amount of the granulating liquid is greater, the decrease of the particle size is greater. While at a relative liquid amount of

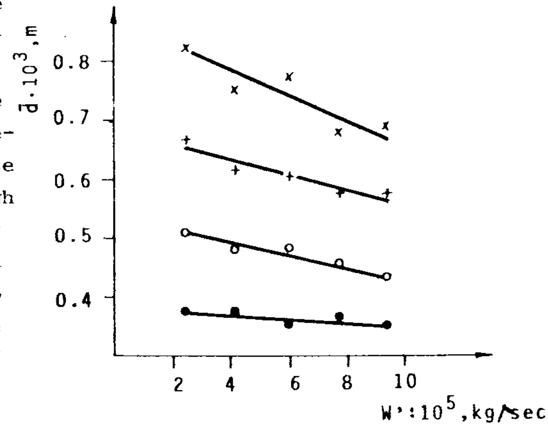

Fig.1. x – V'/V = 30 vol. per cent
+ – V'/V = 20 vol. per cent
o – V'/V = 10 vol. per cent
• – V'/V = 5 vol. per cent

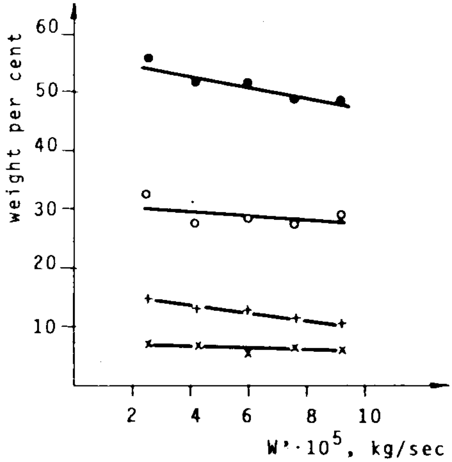

Fig.2. d = (0.1 - 0.2)x10^{-3} m
•-V'/V = 5 vol. per cent
o-V'/V = 10 vol. per cent
+-V'/V = 20 vol. per cent
x-V'/V = 30 vol. per cent

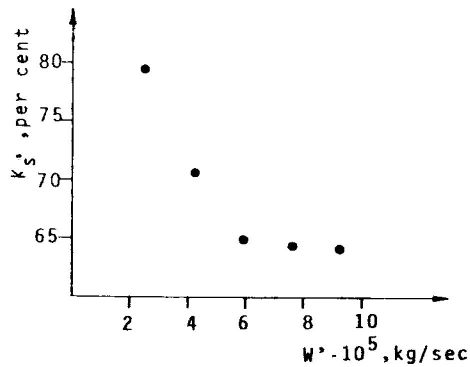

Fig.3. V'/V = 20 vol. per cent

V'/V = 5 vol.per cent, the increase of the feed rate by about four times decreases the average particle size by only about 7 per cent at a relative liquid amount of V'/V = 30 vol. per cent this change is almost 16 per cent.

In Fig. 2 the weight fraction of the particles not granulated is plotted against the feed rate at four different values of the relative amount of the granulating liquid. With the increase of the feed rate there is a slight decrease of this weight fraction. The greatest effect was found at the smallest liquid amount (V'/V = 5 vol. per cent), where the amount of the particles not granulated decreases by about 7 weight per cent when the feed rate is increased from 2.5x10^{-5} kilogram per second to 9.2x10^{-5} kilogram per second, which represents a relative decrease of 12.5 per cent.

In Fig. 3 a more profound change is shown, the wear strength is plotted against the feed rate of the granulating liquid. After an initial sharp drop, the wear strength approaches a lower limit with the increase of the feed rate, despite the fact that the amount of the binder does not vary.

In Fig. 4 the binder content is plotted against the particle size at two different feed rates. The tendencies are identical in both cases, the binder content at first increases with the particle size, but later it attains a nearly constant value. From the Figure it is clear that if a given amount of the binder solution is sprayed in the bed with high velocity, that is during a short time, there are greater differences between the binder content of the particles of different size than in the case of granulation with small feed rates.

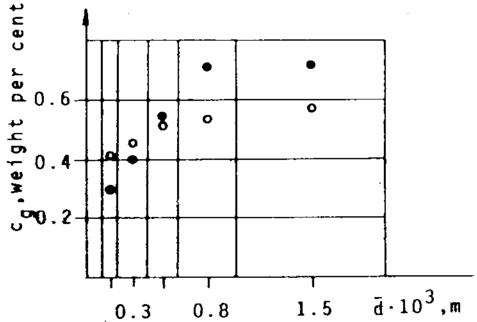

Fig. 4. • – v' = 8.4×10^{-5} kilogram per second; o – v' = 3.4×10^{-5} kilogram per second; V'/V = 20 vol. per cent

The temperature and relative humidity of the air was measured at the inlet and at the outlet. In the Mollier diagram of the humid air, the point corresponding to the measured relative humidity of the air at the outlet was determined from the conditions of the air at the inlet, supposing an adiabatic drying process. The dry bulb temperature corresponding to this point is the "adiabatic temperature", ($T_a^{"}$). The comparison of $T_a^{"}$ and the measured temperature at the outlet makes it possible to see how the process approaches the ideal adiabatic one. The differences between the measured temperatures and those obtained from the psychrometric diagram are plotted against the measured temperatures in Fig. 5. The discrepancies between the temperature of the air at the outlet

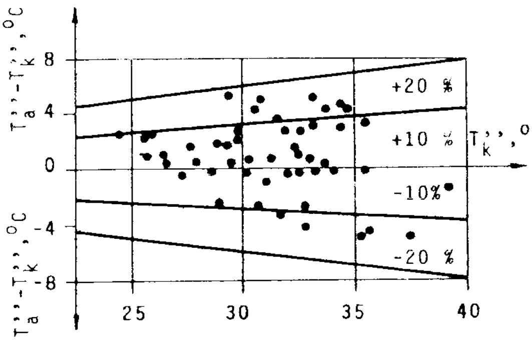

Fig. 5

and the adiabatic temperature were in every case less than ± 20 per cent and in 75 per cent of the cases the differences were not greater than ± 10 per cent.

DISCUSSION

According to Fig. 1, 2, 3 and 4 the changes in the physical properties of the granulates due to the increase of the feed rate of the granulating liquid are not of the type that would make it possible to determine a well defined optimum value of the feed rate. The increase of the feed rate is undesirable on the one hand, because of the worse binder distribution and because of the decrease in the wear strength, while it is desirable on the other hand, because of the greater capacity and because a slight decrease in the amount of the particles not granulated.

The feed rate of the granulating liquid cannot be selected arbitrarily. Both the experimental results and the theoretical considerations show that there is a maximum value of the feed rate above which granulation in a fluidized bed cannot be realized.This greatest feasible value of the feed rate is determined by the air velocity corresponding to the required bed expansion and by the air temperature at the inlet; after a certain feed rate, the heat content of the air flow is insufficient for the removal of the liquid sprayed in. In such cases, the amount of the granulating liquid retained in the bed continuously increases and at a critical liquid content characteristic of the given system, the fluid movement of the bed cannot be maintained even by a further increase of the air velocity.

The maximum liquid feed rate can easily be determined from the liquid mass balance of the process, in the knownedge of the critical liquid content of the bed.

<u>The liquid mass balance</u>:

$$w't'(1 - \frac{c'}{\rho'}) - F\rho''(x_k'' - x_b'') \int_0^{t'} u''(t)dt - V_p''t'(x_k'' - x_b'') = x_r G \quad (1)$$

Equation (1) can be reduced by the introduction of the average gas velocity (integral mean); the air flow of the atomizer should be expressed as the product of the liquid feed rate and the specific air requirement of the atomizer:

$$w't'(1 - \frac{c'}{\rho'}) - F\rho''(x_k'' - x_b'') \bar{u}''t' - kw't'(x_k'' - x_b'') = x_r G \quad (2)$$

The results on the changes of the velocity of the air required for the movement of the fluidized bed, with respect to time, and the method for the determination of the average air velocity will be published in a following paper on the fluidization properties of the granulates.

Taking the critical liquid content as the liquid content of the bed ($x_r = x_{rk}$), Equation (2) gives the maximum feed rate:

$$w_M' = \frac{x_{rk} G + F\rho''(x_k'' - x_b'') \bar{u}''t'}{t'(1 - \frac{c'}{\rho'}) - kt'(x_k'' - x_b'')} \quad (3)$$

The liquid content of the bed attains the critical value at which the movement of the bed stops, just by the end of the spraying (during a time interval of t') if the feed rate is that corresponding to Equation (3). The correlation helps in finding the feasible range of feed rates, that extends in theory from a feed rate of zero to w'_M. In practice the lower limit is the lowest feed rate where the precondition of the formation of the liquid bonds does not exist because of the very small wetting. In such cases the atomized granulating liquid becomes partly dry before getting in the bed (drying with atomization), and partly it dries on the surface of the particles without the formation of either liquid or solid bonds.

The heat balance of the batch granulation in a fluidized bed with spraying can be used for the determination of the equilibrium liquid feed rate (w'_e). The latter is the feed rate at which the heat content lost by the flowing gas is equal to the heat required for the evaporation of the solvent sprayed in.

The heat balance:

$$F\rho''c''_p(T''_b - T''_k)\int_0^{t'} u''(t)dt = [w't'(1 - \frac{c'}{\rho'}) - G\int_0^{t'} x_r(t)dt]r' + Q_v \qquad (4)$$

The term on the left hand side of Equation (4) can be reduced by the introduction of the average gas velocity ($\bar{u}''$); moreover, the Q_v heat loss can be neglected in the case of appropriate heat insulation, since the temperature of the bed does not differ significantly from the room-temperature.

$$F\rho''c''_p(T''_b - T''_k)\bar{u}''t' = [w't'(1 - \frac{c'}{\rho'}) - G\int_0^{t'} x_r(t)dt]r' \qquad (5)$$

The liquid feed rate can be equated to that of the equilibrium liquid feed rate defined above. In that case there is no accumulation of the liquid in the bed, so the equation reduces once again:

$$F\rho''c''_p(T''_b - T''_k)\bar{u}''t' = w'_e t'(1 - \frac{c'}{\rho'})\, r' \qquad (6)$$

From Equation (6) the equilibrium feed rate of the granulating liquid is:

$$w'_e = \frac{F\rho''\bar{u}''c''_p(T''_b - T''_k)}{r'(1 - \frac{\rho'}{\rho'})} \qquad (7)$$

The practical use of this equation is facilitated by the fact that the temperature of the air at the outlet (T''_k) can be aproximated well with the help of the psychrometric diagram, in the knowledge of the characteristics of the air at the inlet, since $T''_k \approx T''_a$ (see under the heading "Experimental Results").

The applicability of Equations (3) and (7) were checked by the solution of several problems. The data needed were substituted into Equations (3) and (7) in the case of the studied model and the following values were obtained: $w'_e = 6\times10^{-5}$ kilogram per second; $w'_M = 10.5\times10^{-5}$ kilogram per second.

The feed rates obtained by the equations are quite feasible and they are compatible with the experimental observations. The calculated maximum feed rate was somewhat higher than the value of 9.2×10^{-5} kilogram per second found to be just employable. But is has to be remembered that at this feed rate an appreciable experiment was carried out so the critical liquid content of the bed was not attained. Hence the higher limit of the feasible feed rates can be well approximated by Equation (3). The equilibrium feed rate calculated by Equation (7) is a good mean value in the feasible interval. Lower rates should be applied for the granulation only if the wear strength of the granulates is to be increased or if small amounts of some additives are to be introduced and distributed in the bed (e.g. in the pharmaceutical industry). The decrease of the feed rate according to Fig. 3. and 4. increases the wear strength of the granulates and helps in the relatively homogeneous distribution of materials fed in small amounts with the granulating liquid, independently of the particle size.

SYMBOLS USED

- c' concentration of the granulating liquid (kilogram per cubic metre)
- c_g the binder content of the granules (weight per cent)
- c_p'' heat capacity of the gas (kilocalories per kilogram per Degree Centigrade)
- d particle size (metre)
- $\bar{d}$ average particle size (metre)
- F cross section area of the apparatus (square metre)
- G the mass of the bed (kilogram)
- k the specific air requirement of the atomizer (kilograms per kilogram)
- K_s wear strength (per cent)
- r' latent heat of the solvent (kilocalories per kilogram)
- Q_v heat loss (kilocalories)
- T_a'' air temperature at the outlet from the psychrometric diagram (Degree Centigrade)
- T_k'' air temperature at the outlet (Degree Centigrade)
- t time (second)
- t' the duration of granulation (second)
- u'' gas velocity (metres per second)
- $\bar{u}''$ average gas velocity (metres per second)
- V the overall volume of the particles to be granulated (cubic metre)
- V' the volume of the granulating liquid (cu.metre)
- V'/V the relative amount of the granulating liquid (vol. per cent)
- V_p'' air flow of the atomizer (kilogram per second)
- w' feed rate of the granulating liquid (kilogram per second)
- w_e' equilibrium feed rate of the granulating liquid (kilograms per second)
- w_M' maximum feed rate of the granulating liquid (kilograms per second)
- x_b'' absolute humidity of the air at the inlet (kilograms per kilogram)

x''_k absolute humidity of the air at the outlet (kilograms per kilogram)

x_r liquid content of the bed (kilograms per kilogram)

x_{rk} critical liquid content of the bed (kilograms per kilogram)

ρ' density of the granulating liquid (kilograms per cu.metre)

ρ'' density of the gas (kilograms per cu.metre)

REFERENCES

1. ORMÓS, Z., PATAKI, K., CSUKÁS, B., Hung. J. Ind. Chem. 1, 307 (1973)
2. RANKELL, A.S., SCOTT, M.W., LIEBERMAN, H.A., CHOW, F.S., BATTISTA, J.V., J. Pharm. Sci. 53, 320 (1964)
3. MÖBUS, W., Ceskoslow. farm. 18, 109 (1969)
4. DAVIES, W.L., GLOOR, Jr. W.T., J. Pharm. Sci. 60, 1869 (1971)
5. ORMÓS, Z., Hung. J. Ind. Chem. 1, 207 (1973)
6. SCOTT, M.W., LIEBERMAN, H.A., RANKELL, A.S., BATTISTA, J.V., J. Pharm. Sci. 53, 314 (1964)

РЕЗЮМЕ

При грануляции, происходящей в псевдоожиженном слое, очень важен выбор соответствующей скорости подачи гранулирующей жидкости, поскольку этот параметр не только влияет на образование гранул, но и в значительной мере определяет производительность установки.

Авторы рассматривают влияние, оказываемое скоростью подачи гранулирующей жидкости на физические свойства (средний диаметр частиц, относительное число несгранулировавшихся частиц, износостойкость, негомогенное распределение связующего вещества) образующихся гранул. На основании уравнений материального и теплового баланса была выведена зависимость для определения максимального и равновесного значения скорости подачи жидкости.