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DEVELOPMENT OF AGGLOMERATED TALC. I. EVALUATION OF FLUIDIZED BED GRANULATION PARAMETERS ON THE PHYSICAL PROPERTIES OF AGGLOMERATED TALC

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ABSTRACT :

Studies were conducted to develop agglomerated talc as a tablet diluent using the fluidized-bed granulation method. A complete 2^3 factorial experiment was run using a Uniglatt fluidized bed granulator to determine the effects of atomizing air pressure (P), inlet air temperature (T), and the quantity of the granulating fluid (V) upon the characteristics of the resultant agglomerated talc. It was found that the atomization pressure was the most prevailing factor for controlling the growth of the agglomerates in the granulation process. With the decrease of the atomizing air pressure, the geometrical mean particle size and flowability of the agglomerates increased. The volume of the binder solution (dilution effect) affects the properties of the agglomerates in the same direction as did the atomizing air pressure. Droplet size distribution of the atomized binder solution was estimated. The result suggested that the dilution effect altered the properties of the product through its adhesivity. The flowability and hardness of talc were significantly improved by the fluidized bed granulation process.

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INTRODUCTION :

Talc is a hydrated magnesium silicate with the chemical formula : $Mg_3SiO_3(OH)_2$(1). The structure of talc consists of layers of unit composition. Each layer contains three planes of arrangements: silicate-brucite-silicate(2). The layers are held to one another by van der Waals forces and are easy to cleave. Talc is nontoxic, less expensive, physiologically inert, and physicochemically inactive; it fullfills all the criteria of being a diluent. But due to the weak van der Waals force being the only force among adjectant layers, talc is a soft mineral. The hardness of talc is one on the Mohs Scale. Adding a large quantity of talc in a tablet dosage form will make the resultant tablet friable and unacceptable. To overcome the tableting deficiency properties of talc, the wet granulation approach can be applied to talc using the fluidized bed granulation method to modify the properties of talc. The resultant talc is called agglomerated talc. This research is designed to develop agglomerated talc using fluidized-bed granulation method.

Fluidized-bed granulation involves the spraying of a binder solution on to a powder suspended in an upward moving stream of air. The agglomerates produced by the fluidized bed granulation method have an excellent flowability and a narrow size distribution of almost spherical particles in which the binder uniformly distributed[(3)]. Aulton and Banks[(4)] classified the possible variables during fluidized bed granulation into three groups; [1] apparatus parameters; [2] process parameters; [3] product parameters. The effectiveness of fluidized-bed granulation is very sensitive to those variables and their interactions and to the properties of the starting materials.

The granulator used in this study is Uni Glatt granulator (Figure 1). Because of its small size, it has a limited solvent evaporation rate for granulation process; therefore the process parameters needed to be carefully controlled. The variables tested in this study are atomizing air pressure, inlet air temperature, and quantity of solvent. Factorial design can be well applied in studying technological processes with many parameters[(5)]. The systematic order of this kind of experimental design allows the construction of the mathematical model which elucidates the influence of the operational parameters examined and the nature of the possible interactions among the individual parameters.

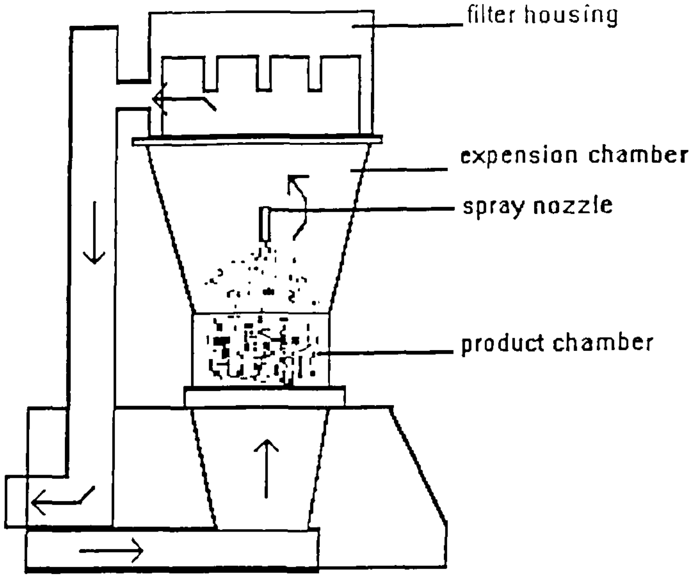

FIGURE 1. UniGlatt fluidized bed granulator.

The aims of this granulation process are to amend the hardness, to enlarge the particle size, and to enhance the flowability of talc. To achieve these goals, a 2^3 factorial experiment on granulation of talc was performed to determine the values of the parameters which significantly influence the characteristics of final agglomerates and to discover the proper conditions for the granulation process.

MATERIAL AND EQUIPMENT :

USP grade talc (Alphafil 400) was supplied by Cyprus Industrial Mineral Company. The binder used is polyvinylpyrrolidone (GAF Corp., k value 29-32). The apparatus used is an UniGlatt granulator (Glatt GmbH) with a binary nozzle (model 970/S4).

EXPERIMENT :

Granulation process :

To reduce the test effort, the following factors of the fluidized-bed granulation were kept constant :

Granulation fluid flow rate : 6.1 ml/min
Fluidizing velocity : 22.5 auf
Drying time : 4 minutes
Bed load : 250 g of talc
Outlet air temperature : 20°C - 30°C
Binder amount : 24.73 g of polyvinylpyrrolidone
Relative humidity of the inlet air : 50 ± 5 %

The three test variables were studied at low (-1) and high (+1) levels. The coded variable is used to preserve orthogonality and to permit direct comparison of the magnitude of the regression coefficients. The two levels of each variable and the treatment combinations evaluated in this design are shown in Table 1 and Table 2 respectively.

The binder solution was prepared freshly and was sprayed for 3 minutes; then the spraying was stopped for 2 minutes. At the end of 5 minutes, the spraying was started again and the granulation process was performed in a continuous manner until the desired amount of binder solution was charged. Then the granules were allowed to fluidize for an additional 4 minutes. The shaker for the exhaust filter was activated from time to time during the process to prevent accumulation of fine powders on the filter surface. The resultant agglomerated talc was subjected to the following tests :

(a) Tapped density :

The tapped density was determined using the Vanderkamp Tap Density Tester. The volume was periodically recorded at specified time intervals. Each sample was tapped till the volume remained constant to ensure maximum packing. The tabulated value is the average of three determinations.

(b) True density :

True density of the samples was determined using a helium pycnometer (Micromeritics Instrument Corp., Model 1330).

(c) Percent tapped porosity :

% tapped porosity = (1 - tapped density/true density) * 100

TABLE 1. The coded value and levels for tested variables.

	+1	-1
Atomizing air pressure (P)	1.0 bar	0.8 bar
Inlet air temperature (T)	90°C	80°C
Volume of the solvent (V)	200 ml	150 ml

TABLE 2. Treatment combinations in the 2^3 factorical design.

Treatment combination	Atomizing air pressure (P)	Inlet air temperature (T)	Volume of the solvent (V)
(1)*	-1	-1	-1
P**	+1	-1	-1
T	-1	+1	-1
V	-1	-1	+1
PT	+1	+1	-1
TV	-1	+1	+1
PV	+1	-1	+1
PTV	+1	+1	+1

* : the treatment combination for all variables is at their low level.

** : the treatment combination with the variable P at the high level and the other two at the low level.

(d) Loss on drying :

One gram of accurately weighed talc was dried in a 105°C oven for three hours.

Loss on drying (%) = weight loss * 100 / sample weight

(e) Hopper flow rate :

A recording powder flowmeter was used for evaluating the flowability of the samples. The aluminum funnel used as the hopper had 20 cm top diameter, 28 cm length, 1.5 cm orifice diameter. One hundred grams of powder were poured into the funnel with the orifice closed. When the orifice was opened , the powder flowed into a pan and a trace was obtained on a 10 inch strip chart recorder.

(f) Particle size determination :

Granule size distribution was estimated using sieve analysis. 100 g granules were placed on the top of a series of U.S. standard sieves and shaken by a Ro-Tap sieve shaker (W.S. Company) for 5 minutes. After sieving, the weight of the material remaining on each sieve was determined. The data were evaluated using a particle size analysis program based on a log-probability plot of the data[6]. From this evaluation, the geometric mean particle size for the granulation was determined. The percent fines is defined as the percentage of the agglomerated talc passed through number 200 U.S. standard sieve (< 74 μ).

Making compacts and hardness measurement :

Compacts were compressed on a Carver press at 2000 lbs force, using a one-half inch, flat-faced punch and die set. The compression rate was constant. Compact weight was maintained at 800 ± 10 mg. After allowing one day for elastic recovery, the hardness of the agglomerated talc compacts was measured by a Schleuniger hardness tester.

Surface tension and viscosity :

The surface tension and viscosity of the binder solution were measured using Fisher Autotensiomat and Brookfield Digital Viscosmeter (Model DV-II) respectively.

Droplet size analysis :

A modification of the method described by Schafer and Worts [7] was used to study the droplet size distribution of the sprayed binder solution. A slide covered with parafilm (American National Can) was manually passed across the spray cone at a distance of about 24.5 cm below the nozzle orifice. A photomicrograph (Polaroid 667 film) of the droplet sample was then taken immediately. Six photographs were sampled for a single droplet size analysis. The droplet size distribution was characterized by counting a total of approximately 500 droplets.

RESULTS AND DISCUSSION :

The flow rate, mean particle size, percent fines, loss on drying and percent tapped porosity of each batch of agglomerated talc are shown in Table 3. A complete 2^3 factorial design allows estimation of all the effects and interactions with no confounding; but it can only estimate linear effects and interactions. The properties of the granule obtained using fluidized-bed granulation process have been suggested to have linear relationship with the processing variables[8].

The effects of changes in granulation characteristics due to changes in inlet air temperature (T), volume of the solvent (V), and atomizing air pressure (P) were described using a linear regression model. Student t-tests were conducted for each variable in the full model and a reduced model containing only the significant variables (at $\alpha = 0.05$ significance level) was determined. The testing of the data was done using the SAS package (SAS Version 6, SAS Institute). The corresponding regression equation of the full model for this design is :

$$Y_i = \beta_o + \beta_1 P + \beta_2 T + \beta_3 V + \beta_4 PT + \beta_5 TV + \beta_6 PV + \beta_7 PTV$$

Where Y_i is the measured responses, such as flow rate, mean particle size, percent fines, loss on drying and percent tapped porosity, β_o to β_7 are regression coefficients and P, T, V are the coded level of the independent variables.

TABLE 3. The physical properties of the agglomerated talc evaluated in this experimental design.

Treat -ment	Flow rate (g/sec)	Mean size (μ)	Percent fines (%)	L.O.D. (%)	% Tapped porosity
(1)**	11.3 (0.6)	131.0 (1.1)	14.3 (0.2)	1.32 (0.14)	79.7 (0.4)
P***	6.3 (0.3)*	88.4 (0.2)	35.2 (3.1)	1.27 (0.08)	79.2 (0.1)
T	10.3 (0.2)	126.2 (3.4)	15.3 (2.5)	1.33 (0.02)	81.3 (0.2)
V	12.0 (0.5)	127.2 (1.2)	14.9 (0.5)	1.16 (0.07)	78.4 (0.4)
PT	8.1 (0.5)	93.0 (1.4)	34.1 (1.7)	1.14 (0.03)	79.0 (0.2)
TV	11.2 (0.8)	105.2 (0.4)	20.8 (1.1)	0.75 (0.05)	77.9 (0.2)
PV	7.6 (1.3)	79.3 (1.8)	47.9 (1.6)	0.60 (0.05)	73.9 (0.4)
PTV	8.9 (0.6)	73.8 (1.3)	52.3 (1.9)	0.71 (0.04)	75.6 (0.1)

* : Mean and standard deviation of three determinations.

**: the treatment combination for all variables is at their low level.

*** : the treatment combination with the variable P at the high level and the other two at the low level.

Table 4 summarizes the results of the regression analysis and gives the reduced model for each measured response. The correlation coefficients, F ratios, and model significance level were acceptable in all cases.

As indicated by the magnitude of the regression coefficients in Table 4, the atomization air pressure (P) had the greatest effect on the mean particle size, percent fines, and flow rate of the resultant talc agglomerates. As the atomization air pressure is increased, a reduction in mean particle size, granule flow rate, loss drying value, and percent tapped porosity occurs, whereas the percent fines of the agglomerates is increased.

TABLE 4. Summary of regression results for the measured responses.

Yi / Coefficient	Flow rate	Mean size	Percent fines	L.O.D.**	% Tapped porosity
β_0	9.46	103.01	29.52	0.01	78.13
β_1	- 1.74	- 19.38	12.85	$-1*10^{-3}$	-1.2
β_2	*	-3.47	*	$-5*10^{-3}$	0.32
β_3	0.47	- 6.65	4.44	$-2*10^{-3}$	-1.68
β_4	0.63	3.23	*	$5*10^{-4}$	*
β_5	*	-3.41	*	*	*
β_6	*	*	3.28	$-5*10^{-4}$	-0.48
β_7	*	*	*	$8*10^{-4}$	0.51
F	72.16	395.37	103.15	58.03	434.25
P	0.0001	0.0001	0.0001	0.0001	0.0001
R^2	0.92	0.99	0.96	0.95	0.99

where * indicates that the regression coefficient is not significant at $\alpha = 0.05$.

** : Loss on drying (%).

The regression equation of the full model is :

$$Y_i = \beta_o + \beta_1 P + \beta_2 T + \beta_3 V + \beta_4 PT + \beta_5 TV + \beta_6 PV + \beta_7 PTV$$

F : Mean square regression/mean square residual.

P : Model significance level.

R^2 : The square of the multiple correlation coefficient.

The atomization air pressure controls the degree to which the granulating solution is broken up. The droplet size increases with the decrease of the atomizing air pressure[7]. When the atomized droplets reach the fluidized particles, nuclei consisting of two or more primary particles held together by liquid bridges are formed. Since a large droplet can bind more particles together than a small one, decreasing the atomizing air pressure results in larger nuclei and subsequently produces larger granules. In addition, increasing the atomizing air pressure results in smaller and more droplets which gives a more even wetting of the bed and subsequently the percent tapped porosity of the resultant agglomerates is decreased.

As shown by Table 4, the volume of the binder solution (V or dilution effect) affects the properties of the agglomerates in the same direction as does the atomizing air pressure (P). Changes in dilution of the binder solution can alter two parameters for granulation process, e.g. solution viscosity and binder adhesiveness. The former parameter can directly influence the droplet size distribution, the latter parameter governs the adhesivity within particles.

The surface tension and viscosity of the binder solutions prepared with different quantity of solvent are listed in Table 5. There is no significant difference in surface tension of the two solutions. However, the viscosity of the binder solution with a lower volume of solvent is higher (at $\alpha = 0.05$) than that of the binder solution with a higher amount of solvent. To verify the effect of changes in atomizing air pressure and volume of the solvent on the droplet size distribution of the binder solution, droplet size analysis was performed. The mean droplet size of the four treatment combinations is listed in Table 6 and the droplet size distribution plot is shown in Figure 2. Student t tests were used to compare the mean droplet size for the four different combinations of these two variables. The result of the test showed, at the same atomizing air pressure, that the changes of the mean droplet size due to the changes of the volume of the solvent are not statistically significant. However, decreasing the atomizing air pressure can significantly increase the mean droplet size.

It is shown in Table 4 that with an increase in quantity of solvent, there is a reduction in the final mean granule size. This was attributed to a decrease in the binder solution's tackiness or adhesiveness. Moreover, increasing the

TABLE 5, Viscosity and surface tension for binder solution.

Volume of the solvent (V)	- 1	+ 1
Viscosity (cps)	11.3 (0.5)	7.4 (0.8)
Surface tension (dynes/cm)	59.3 (0.5)	59.2 (1.0)

TABLE 6. Mean droplet size of binder solution sprayed at different levels of atomizing air pressure and volume of the solvent.

Atomizing air pressure (P)	- 1		+ 1	
Volume of the solvent (V)	- 1	+ 1	- 1	+ 1
Mean droplet size (μ)	160.58 (95.28)	149.46 (78.95)	123.70 (89.85)	114.59 (83.75)

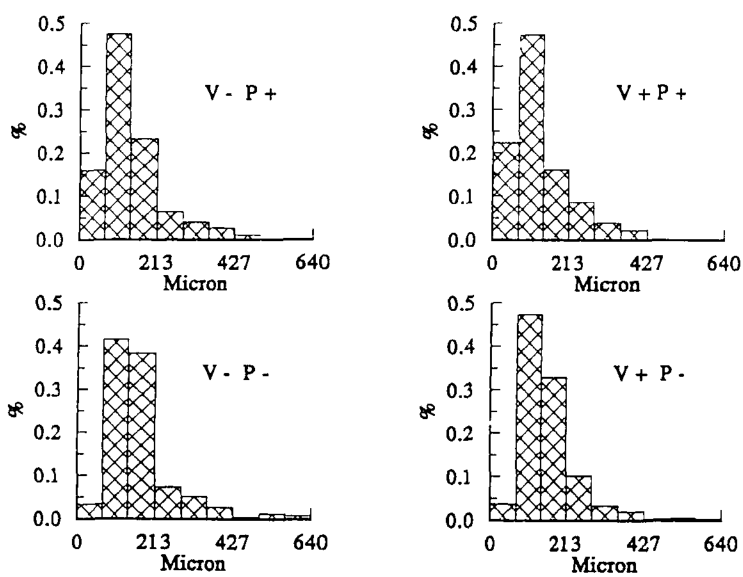

FIGURE 2. The droplet size distribution of the binder solution sprayed at different atomizing air pressure (P) and different dilution level (V).

quantity of the solvent increases penetration and wetting of the fluidized solids by the more dilute solution and produces less porous and more flowable granules.

Inlet air temperature has a significant effect on the degree of agglomeration which is a function of the equilibrium moisture content of the granulation solid bed. For determination of the drying rates, a well known equation[9] for heat transfer is as follows:

$$dw/dt = h * (A/\Delta H) * \Delta T$$

where dw/dt is the rate of mass transfer for the evaporation of the liquid, h is the heat transfer coefficient, ΔH is the latent heat of vaporization of the solvent, A is the surface area of the drying solid, and ΔT is the temperature difference between the drying air and the product. Increasing the inlet air temperature can increase the temperature difference between the drying air and the product[10], thus increasing the drying rate and shortening the time available for the primary particles to adhere to the nuclei before the wet binder dry out. Therefore the moisture content and mean granule size decrease with the increase of the inlet air temperature.

Basic properties of powders that are related to their flow properties include surface area, chemical composition, particle size distribution of the powders and the structure, size, shape, porosity, and surface texture of the individual particle porosity. The flow rate of the agglomerated talc can be increased by increasing the quantity of the solvent and decreasing the atomizing pressure. Inlet air temperature has no significant effect on the flowability, but it interacts with atomizing air pressure. The + sign of the PT interaction term in the reduced model for flow rate means that the flow rate increases with the decrease or increase of the two factor values at the same time.

Comparisons of the flowability and compact hardness were made between the talc powder and the eight batches of agglomerated talc. It was found that the talc powder can not freely flow through the funnel and the hardness of the talc compact is approximately 1.0 kp. In contrast, the agglomerated talc can flow freely and has an average flow rate 10 g/sec; also the hardness of the agglomeratec talc compacts compressed at the same pressure are within 13.5 ~ 16.5 kp. This indicates that the flowability and bonding strength of talc are significantly improved by the fluidized bed granulation process.

CONCLUSION :

This study has demonstrated the feasibility of granulating talc using the fluidized bed granulation method. The 2^3 factorial experimential design is able to construct the mathematical model to evaluate the magnitude and direction of the influence of the atomizing air pressure, inlet air temperature, and volume of the solvent on the properties of the resultant agglomerated talc. The mean particle size and moisture content of the agglomerated talc decrease with the increase of atomizing air pressure, inlet air temperature and volume of the solvent. The result of this study can assist to monitor the adequate granulation condition for the development of the agglomerated talc.

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