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RESEARCH PAPERS

DEVELOPMENT OF AGGLOMERATED TALC II : OPTIMIZATION OF THE PROCESSING PARAMETERS FOR THE PREPARATION OF GRANULATED TALC

Kangwen Lin[1] and Garnet E. Peck[2]

1 Procter & Gamble Pharmaceuticals, Norwich, NY 13815

2 Department of Industrial and Physical Pharmacy

Purdue University, West Lafayette, IN 47907

ABSTRACT :

Agglomerated talc was prepared by the wet granulation method using a planetary mixer and a high speed mixer. The effect of the amount of water added and granulation time for the planetary mixer on the physical properties of the agglomerated talc were investigated by a response surface design. The speed of the agitator and granulation time for the high speed mixer were selected as the two independent variables to study the granulation processing using the same statistical design. Several characteristics of the product prepared by a planetary mixer or high speed mixer were well expressed as a quadratic function of the two independent variables studied in coded level. By superimposing contour plots of granule friability and percent fine of granule, a region was obtained where the requirements of friability and percent fines could be satisfied by controlling the processing variables. The optimal granulation condition was chosen from that region. The values of the measured responses of the agglomerated talc produced using the optimal granulation condition agreed well with the predicted values obtained from the regression equations.

2 To whom the correspondence should be addressed.

INTRODUCTION :

Granulation and agglomeration are terms generally used to describe the process of size enlargement. The purpose of granulation is to improve the properties , such as flow, handling, bulk density, compressibility, and appearance[1], of the powder. Since the granulation process is complicated, processing variables must be carefully controlled and optimized in order to obtain reproducibility from batch to batch. The important processing variables for mixing granulation are amount of binder solution, mixing speed, and mixing time. In this study, wet granulation techniques were applied to talc by using a planetary mixer and a high speed mixer to produce the agglomerated talc with enhanced flowability and binding strength. The purpose of this study is to investigate the effect of processing variables of these two mixing granulators on the physical characteristics of the resultant granules by means of the response surface method.

MATERIAL AND EQUIPMENT:

USP grade talc (Alphafil 400) was supplied by Cyprus Industrial Mineral Company. The binder used was polyvinylpyrrolidone (GAF Corp., k value 29-32). The apparatus used were a Hobart planetary mixer and a high speed mixer (Fukae Powtec Corporation, Model LFS-GS-1J). The high speed mixer was equipped with two ammeters to monitor the current (ampere) consumed during the granulation process for the agitator and chopper respectively.

EXPERIMENT :

Granulation process for planetary mixer :

Adequate weight of PVP (to produce 7% w/w PVP-containing agglomerated talc) was dissolved in different amounts of solvent (distilled water) and served as binder solutions. With continuous low speed mixing (65 rpm) for 9 minutes, the binder solution was added to 600g talc powder at the rate of 21 ml/min. Then the wet mass was mixed at a higher speed (125 rpm) for a fixed time.

Granulation process for high speed mixer :

The binder solution was prepared by dissolving 37.2 g PVP (to produce 7% w/w PVP-containing agglomerated talc) in 110 ml distilled water. With continuous low speed mixing (agitator speed 200 rpm, chopper speed 450 rpm) for 6 minutes, the binder solution was added to 500 g talc powder at the rate of 25 ml/min. Then the chopper speed was increased to 1200 rpm and the wet mass was mixed at a predetermined agitator speed for a certain time.

The discharged wet mass was passed through a number 12 U.S. standard sieve. After storing in a 15% relative humidity, 20^{o}C room for 24 hours, the granules were passed through screen number 16. The moisture content of all granules was lower than 1.5% (w/w). The following characteristics of the resultant agglomerated talc were determined :

(a) Bulk and tap density :

The bulk density was the quotient of weight to the volume of the sample. The tapped density was determined using the Vanderkamp Tap Density Tester.

(b) Hopper flow rate :

A recording powder flowmeter was used for evaluating the flowability of the samples. One hundred grams of agglomerated talc 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.

(c) Particle size determination :

Granule size distribution was estimated using sieve analysis (Ro-Tap sieve shaker). The geometric mean particle size for the granulation was determined using a particle size analysis program based on a log-probability plot(2).

(d) Percent fine :

The percent fines is defined as the percentage of the agglomerated talc passed through a number 200 U.S. standard sieve (< 74 μ).

(e) Granule friability :

Twenty plastic balls and 10 grams of granule which passed through number 40 mesh screen and retained on number 50 screen were placed on the number

50 screen. The diameter and weight of the plastic balls were 0.95 cm and 530 ± 10 mg respectively. The screen was shaken using a Ro-Tap sieve shaker for 5 minutes. The friability of the granule was determined by dividing the weight loss by the original weight of the granule.

EXPERIMENTAL DESIGN :

The experimental design selected to model the effect of independent variables on the various response parameters was a two factors central composite design. This design assured that the entire factor space was adequately covered, so that accurate estimates of the regression coefficients in the mathematical models could be made. The mathematical model for this study was a second order response function. Each variable was measured at five levels, i.e., -d, -1, 0, 1, d where d was the distance from the center point to each of the four extreme points. The d value in this design was computed to be 1.44 [3]. The two independent variables studied were granulation time (A1) and amount of solvent (B1) for the granulation process using a planetary mixer. The granulation time (A2) and speed of agitator (B2) were the two independent variables studied for the granulation process using a high speed mixer. The levels and coded value for the independent variables, A1, B1 and A2, B2, were listed in Table 1 and Table 2 respectively.

Central composite design is the first order factorial design augmented by additional points to allow estimation of the coefficients of a second order surface. The treatment combinations for this design are listed in Table 3 and each treatment combination has two replicate measurements. The first four runs are the usual factorial points for fitting a first order model. The fifth run is the center point and the four remaining points are the axial point.

The effects of changes in granulation characteristics due to changes in granulation parameters were described using a second order 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

TABLE 1. The levels and coded value for the two independent variables, A1 and B1, for the granulation process using a planetary mixer.

Coded level	-1.414	-1	0	+1	+1.414
Mixing time (A1)	1.2 min.	2.0 min.	4.0 min.	6.0 min.	6.8 min.
Amount of solvent (B1)	116 ml	120 ml	130 ml	140 ml	144 ml

TABLE 2. The levels and coded value for the two independent variables, A2 and B2, for the granulation process using a high speed mixer.

Coded level	-1.414	-1	0	+1	+1.414
Mixing time (A2)	1.2 min.	2.0 min.	4.0 min.	6.0 min.	6.8 min.
Agitator speed (B2)	459 rpm	500 rpm	600 rpm	700 rpm	741 rpm

TABLE 3. The nine treatment combinations for the two variables central composite experimental design.

Run	Ai	Bi
1	1	1
2	-1	1
3	1	-1
4	-1	-1
5	0	0
6	0	1.414
7	1.414	0
8	0	-1.414
9	-1.414	0

Version 6, SAS Institute). The corresponding regression equation of the full model for this design is :

$$Yj = \beta_o + \beta_1 Ai + \beta_2 Bi + \beta_3 AiBi + \beta_4 Ai^2 + \beta_5 Bi^2$$

where Yj is the measured responses, such as bulk density, tap density, flow rate, geometrical mean particle size, percent fines, and granule friability. β_o to β_7 are regression coefficients. Ai and Bi are the coded level of the independent variables; i = 1 when the product was prepared by planetary mixer and i = 2 when the product was produced by high speed mixer.

RESULT AND DISCUSSION:

Granulation process usinfg a planetary mixer :

The variations of the characteristics of the agglomerated talc are shown in Table 4. The granulation time (A1) and amount of water (B1) strongly affected the response variables. The geometrical mean particle size and friability of the agglomerated talc showed a wide range from 293 μ to 409 μ and 14.3% to 25.0% respectively. Functional relationships between the measured responses A1 and B1 were analyzed. Table 5 summarizes the results of the regression analysis and gives the reduced regression model for each measured response. As illustrated by the three dimensional surface graph of the reduced regression model (Figure 1a), bulk density is a hyperbolic function of A1. Geometrical mean particle size is a second order function for both variables (A1 and B1) with its maxima value at A1 = -0.40 and B1 = 0.0 coded level (Figure 1b). Figure 1c shows the response surface as a function of A1 and B1 for granule friability. If a smaller friability value is desirable, the combination of higher A1 and higher B1 should be taken. However, this condition may lead to a larger percent fine for the agglomerated talc (Figure 1d).

When contour plots of granule friability and percent fines were superimposed, the influence of A1 and B1 was clearly elucidated. The optimal zone is defined where both friability and percent fine of the granule are as small as possible. From the optimal zone in Figure 2, a point at A1 = 0.5 (4

TABLE 4. Physical properties of agglomerated talc prepared by a planetary mixer.

Run	Bulk density (g/ml)	Tap density (g/ml)	Mean size (μ)	Flow rate (g/sec)	Friability (%)	Percent fines (%)
1	0.720	0.904	293.2	31.4	14.3	15.16
2	0.661	0.846	368.3	25.0	25.0	11.78
3	0.720	0.920	342.4	21.7	21.1	15.16
4	0.653	0.904	361.4	12.2	19.6	17.56
5	0.687	0.849	409.6	26.4	19.9	10.04
6	0.665	0.837	380.2	30.1	14.7	8.61
7	0.734	0.920	319.8	30.3	15.2	12.32
8	0.681	0.889	381.3	22.2	25.0	13.67
9	0.667	0.899	399.5	11.4	21.0	15.22

TABLE 5. Summary of the result of regression analysis for granulation using a planetary mixer.

Co-efficient	Bulk density (g/ml)	Tap density (g/ml)	Mean size (μ)	Flow rate (g/sec)	Friability (%)	Percent fines (%)
β_o	0.680	0.862	409.67	25.73	19.54	11.74
β_1	0.028	0.013	-25.86	5.33	-2.17	*
β_2	*	-0.019	*	4.11	-2.01	-1.617
β_3	*	*	*	*	-3.03	1.446
β_4	0.011	0.026	-32.25	-2.67	*	1.739
β_5	*	*	-21.66	*	*	*
F	84.2	24.3	12.7	36.5	15.7	9.9
r	0.96	0.92	0.85	0.94	0.88	0.84

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

F : Mean square regression/mean square residual.

r : The multiple correlation coefficient.

Full model : $Yj = \beta_o + \beta_1 A1 + \beta_2 B1 + \beta_3 A1B1 + \beta_4 A1^2 + \beta_5 B1^2$

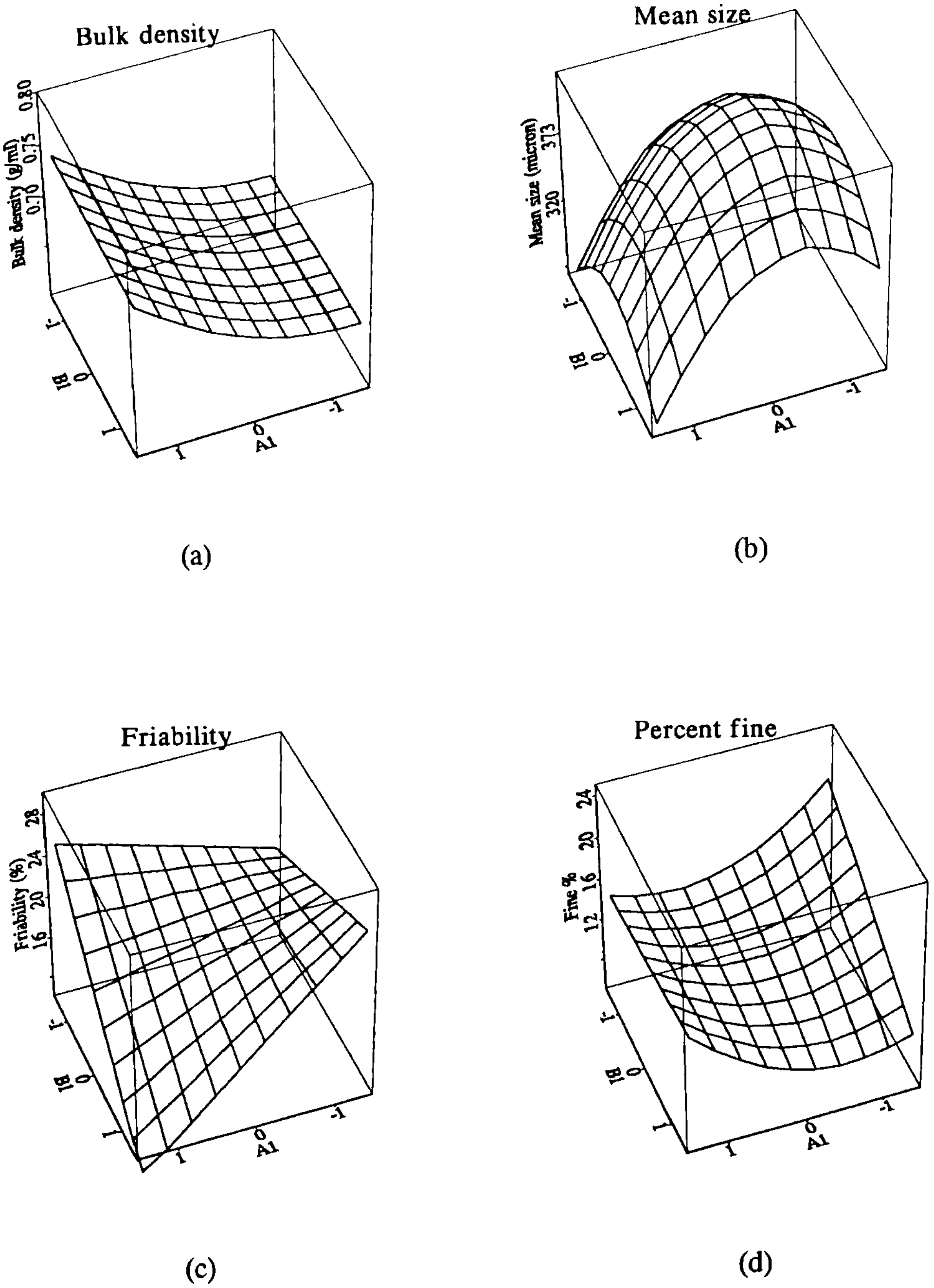

FIGURE 1. Three dimensional surface graphs of the characteristics of the agglomerated talc prepared by a planetary mixer.

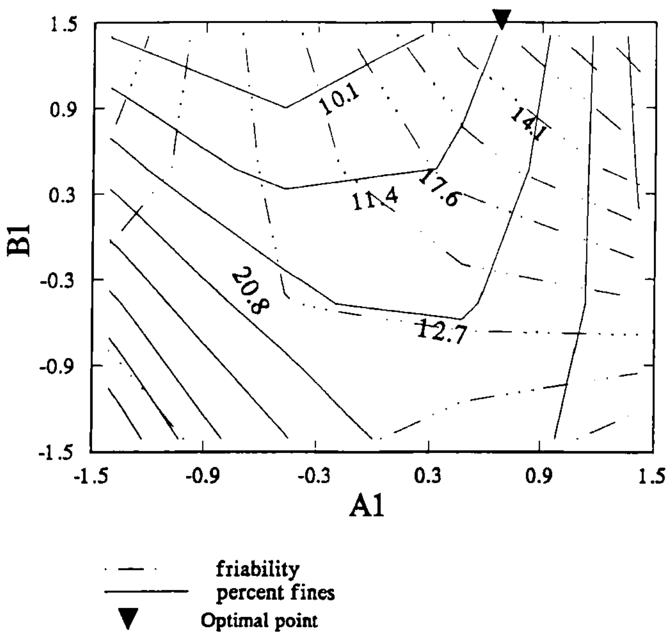

FIGURE 2. Overlapping of the contour plots for granule friability and percent fines.

miniutes) and B1 = 1.414 (144 ml) was chosen as the optimal granulation condition. Experiments were run using this optimal granulation condition. The predicted value for each measure response obtained by using the reduced regression model and the experimental outcome were listed in Table 6. It was found that the experimental value agreed very well with the predicted value.

Granulation process using a high speed mixer :

From the result of the previous study, it was found that with the increase of the amount of solvent, the resultant granules had lower friability and lower percent fine. However, due to the limitation of the instrument, there is a maximum for the amount of solvent added. In this study, the amount of solvent applied was close to the maximum for this system and the tested variables were granulation time (A2) and speed of agitator (B2).

TABLE 6. The predicted value and measured value of each measured response for using the optimal granulation condition at A1 = 1.414, B1 = 0.5 coded level.

Measured response	Predicted value	Measured value
Bulk density (g/ml)	0.697	0.690
Tap density (g/ml)	0.893	0.851
Flow rate (g/sec.)	33.54	30.30
Mean size (μ)	345.37	357.28
Friability (%)	13.74	14.6
Percent fines (%)	8.87	8.10

The physical properties of the agglomerated talc and the results for regression analysis are listed in Table 7 and Table 8 respectively. The regression model for two measured responses, bulk density and tap density, was not statistically significant (at α=0.05). As shown in Table 8, granulation time (A2) strongly influenced all of the characteristics of the product.

The stages of wet granulation include agglomeration, agglomeration breakdown, reagglomeration and paste formation(4). The end point of the granulation process has always been an important issue and has been studied by many scientists. In the past, it was determined by feeling the wet mass in the palm. Recently, Travers(5) determined the end point by measuring the torque on the mixer blade as a function of mixing time or liquid level. Leuenberger(6) used the power consumption curve to determine the end point. Terashita(7)(8) studied the agitation-granulation process of the Fukae Powtec high speed mixer by measuring the power consumption curve. They found that regardless of the granulator model or formulation, the granulation process can be divided into four phases : phase I, formation of agglomerates and nucleic particle; phase II, growth into granular particles; phase III, granular refinement; phase IV, granulation completed. The corresponding power consumption (p) for these four phases was : in phase I, p grows; in phase II, p fluctuates; in phase III, p declines, and in phase IV, p displays a constant Ps value. The granulation end

TABLE 7. Physical properties of agglomerated talc produced by a high speed mixer.

Run	Bulk density (g/ml)	Tap density (g/ml)	Mean size (μ)	Flow rate (g/sec)	Friability (%)	Percent fines (%)
1	0.808	0.999	522.4	36.8	12.1	1.26
2	0.792	0.977	424.7	36.4	14.4	1.72
3	0.769	0.966	522.3	36.1	11.9	1.21
4	0.782	0.962	375.0	35.5	14.2	3.92
5	0.792	0.976	459.7	36.0	12.2	1.50
6	0.769	0.951	424.7	36.1	17.2	1.82
7	0.800	0.983	532.3	36.3	11.9	1.11
8	0.769	0.956	448.7	35.8	16.9	1.71
9	0.762	0.945	342.0	35.9	16.8	5.63

TABLE 8. Summary of the result of regression analysis for granulation using a high speed mixer.

Coefficient	Mean size (μ)	Flow rate (g/sec)	Friability (%)	Percent fines (%)
β_o	450.20	36.10	12.85	1.50
β_1	64.30	0.21	-1.44	-1.19
β_2	*	0.25	*	*
β_3	*	*	*	0.56
β_4	*	*	*	0.80
β_5	*	*	1.50	*
F	169.7	16.2	14.2	24.0
r	0.95	0.82	0.81	0.92

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

F : Mean square regression/mean square residual.

r : The multiple correlation coefficient.

Full model : $Yj = \beta_o + \beta_1 A2 + \beta_2 B2 + \beta_3 A2B2 + \beta_4 A2^2 + \beta_5 B2^2$

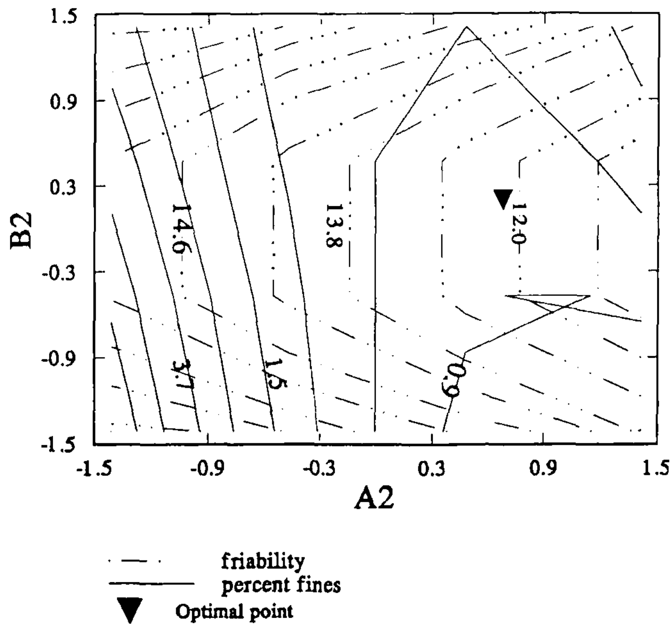

FIGURE 3. Overlapping of the contour plots of granule friability and percent fines.

point(7)(8) that produced a high yield of spherical well-compacted granules was found in the area where a constant value of power consumption was obtained (initial stage of phase IV).

In this study, the optimal granulation condition was determined using the response surface method. By measuring the characteristics of the product prepared using the predetermined treatment condition, the outcome for the entire experimental space can be estimated and predicted. The overlapping of contour plots of granule friability and percent fines is shown in Figure 3. The optimal zone is defined where both friability and percent fines of the granule are as small as possible and a point at A2 = 0.74 (5.48 minutes) and B2 = 0 (600 rpm) was chosen as the optimal granulation condition. Experiments were run using this optimal granulation condition. The predicted value obtained using the reduced regression model and the experimental outcome for each measured response are very close (Table 9).

TABLE 9. The predicted value and measured value of each measured response for using the optimal granulation condition at A2 = 0, B2 = 0.74 coded level.

Measured response	Predicted value	Measured value
Flow rate (g/sec.)	36.3	36.0
Mean size (μ)	497.8	505.2
Friability (%)	11.8	12.0
Percent fines (%)	1.06	0.93

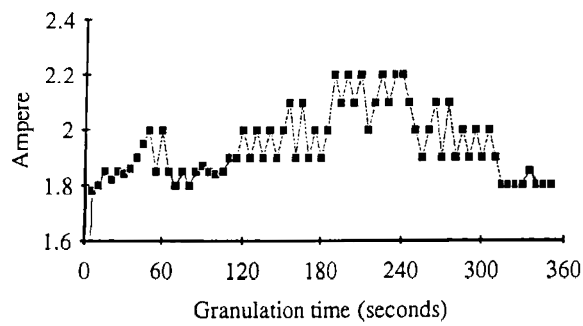

FIGURE 4. Power consumption vs. granulaton time plot for a high speed mixer.

In order to verify the validity of this experimental design for determination the end point of granulation, it was of interest to locate this optimal granulation time point on the power consumption curve plot. Experiments were run using the optimal impeller speed (600 rpm). The power consumption curve for both the agitator and chopper during the process was recorded from the ammeters every five seconds and plotted as Figure 4. The ampere consumed for the chopper remained constant for the whole process. It was found that the optimal end point (5.48 minutes or 328 seconds) determined using the response surface method is very close to the end point at the initial stage of phase IV described by Terashita[7].

The speed of an agitator has been reported as an important processing variable. With the increase of agitator speed, intragranular porosity and fines of the granule decreased[9][10]. As indicated in Table 8, speed of agitator (B2) affected the percent fines through the interaction with granulation time (A2) and the square of B2 was proportional to the granule friability. However, A2 was a more important factor, as far as the granule size distribution was concerned.

CONCLUSIONS:

Physical characteristics of the granules were strongly influenced by the processing variables. In this study, the effects of processing variables on the physical properties of the resultant agglomerated talc were successfully described using the second order regression equations obtained through the response surface method. By superimposing contour plots of granule friability and percent fines, the optimal granulation condition was chosen. The experimental value obtained using the optimal granulation condition for the measured response showed good agreement with predictions for both planetary mixing and high speed mixing processes.

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