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International Journal of Pharmaceutics 123 (1995) 257–264

international journal of pharmaceutics

Droplet size measurement: II. Effect of three independent variables on parameters describing the droplet size distribution from a pneumatic nozzle studied by multilinear stepwise regression analysis

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Received 1 December 1994; accepted 20 February 1995

Abstract

The influence of three independent variables (atomizing air pressure, flow rate of binder solution and polyvinylpyrrolidone concentration) on the parameters describing the droplet size distribution from a pneumatic nozzle (volume of droplets under 18.9 μm, median and 90% fractile of droplet size) was studied using a normal 3^3 factorial design. The droplet size measurement was carried out by laser diffractometry. The dependence of the response variables on the independent variables was studied by a multilinear stepwise regression analysis. On the basis of this study, it was concluded that a high atomizing air pressure led to an increased volume of small droplets. Thereafter, the polyvinylpyrrolidone concentration and the binder flow rate affected this response variable inversely. Increasing the atomizing air pressure resulted in a fall in the droplet size. A larger droplet size was obtained with increased binder flow rate and polyvinylpyrrolidone concentration. In addition to main and quadratic effects, the regression analyses revealed some interactions between independent variables. For example, the atomizing air pressure had a stronger effect on the median of droplet size when the polyvinylpyrrolidone concentration was lower. This was supposed to be due to changes in viscosity.

Keywords: Atomizing air pressure; Binder solution flow rate; Polyvinylpyrrolidone concentration; Droplet size, median; Droplet size, 90% fractile; Multilinear stepwise regression analysis; Small-droplet volume

1. Introduction

So far, droplet size studies have revealed that the mean droplet size from pneumatic nozzles is primarily affected by the following factors:

(i) The nozzle construction (Gretzinger and Marshall, 1961; Kim and Marshall, 1971; Aulton and Banks, 1979);

(ii) The spray angle (corrected by the setting of the air dome) (Schæfer and Wørts, 1977);

(iii) The liquid orifice (Schæfer and Wørts, 1977);

(iv) The liquid flow rate and air-to-liquid mass ratio (Gretzinger and Marshall, 1961; Kim

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SSDI 0378-5173(95)00082-8

and Marshall, 1971; Schæfer and Wørts, 1977);

(v) The atomizing air pressure (Kim and Marshall, 1971; Aulton and Banks, 1979; Yliruusi et al., 1992);

(vi) The temperature of the granulating solution (Aulton and Banks, 1979);

(vii) The addition of surfactants (Aulton and Banks, 1979; Yliruusi et al., 1992).

The air-to-liquid mass ratio is often used to

Table 1
Measured values for parameters describing the droplet size distribution from a pneumatic nozzle

Amount of PVP (%) in solution	Atomizing air pressure (bar)	Flow rate of binder solution (g/min)		Volume of droplets (%) under 18.9 μm $x \pm \epsilon$ [a]	Fractiles of droplet size (μm)	
		Target	Real		50% $x \pm \epsilon$ [a]	90% $x \pm \epsilon$ [a]
0	1.0	100	102	18.6 ± 1.0	37.1 ± 0.9	68.7 ± 2.6
0	1.5	100	105	41.1 ± 1.0	24.3 ± 0.3	43.3 ± 0.5
0	2.0	100	101	61.8 ± 0.5	15.4 ± 0.2	33.6 ± 0.2
0	1.0	150	156	16.3 ± 0.1	41.6 ± 0.2	104.7 ± 0.7
0	1.5	150	154	30.9 ± 0.3	28.6 ± 0.2	56.4 ± 0.2
0	2.0	150	156	54.5 ± 0.2	17.7 ± 0.1	37.9 ± 0.1
0	1.0	200	210	13.0 ± 0.4	45.1 ± 0.3	194.8 ± 1.8
0	1.5	200	207	27.2 ± 0.1	31.5 ± 0.1	72.2 ± 0.5
0	2.0	200	210	48.7 ± 0.4	19.4 ± 0.2	46.3 ± 0.4
10	1.0	100	104	14.3 ± 0.3	40.5 ± 0.2	75.6 ± 0.7
10	1.5	100	106	25.3 ± 0.3	31.2 ± 0.1	52.6 ± 0.1
10	2.0	100	105	35.9 ± 0.8	24.7 ± 0.4	43.3 ± 0.4
10	1.0	150	166	13.9 ± 0.3	45.9 ± 0.3	177.9 ± 11.0
10	1.5	150	169	23.5 ± 0.8	35.7 ± 0.3	75.0 ± 0.4
10	2.0	150	167	30.5 ± 0.6	28.4 ± 0.4	62.1 ± 4.2
10	1.0	200	208	10.0 ± 0.2	56.0 ± 0.5	277.4 ± 0.7
10	1.5	200	210	21.1 ± 0.9	39.4 ± 0.4	124.1 ± 5.4
10	2.0	200	209	26.0 ± 0.1	32.4 ± 0.2	96.4 ± 0.2
20	1.0	100	117	16.3 ± 1.7	41.4 ± 0.4	79.0 ± 7.9
20	1.5	100	126	21.6 ± 0.3	35.5 ± 0.2	60.5 ± 0.4
20	2.0	100	119	25.2 ± 0.9	31.9 ± 0.3	52.1 ± 1.3
20	1.0	150	147	14.9 ± 1.2	45.5 ± 0.7	166.2 ± 19.0
20	1.5	150	151	19.2 ± 0.4	37.5 ± 0.5	99.2 ± 14.0
20	2.0	150	145	21.8 ± 0.7	34.7 ± 0.7	113.7 ± 3.7
20	1.0	200	196	9.1 ± 0.5	59.5 ± 3.5	336.5 ± 21.0
20	1.5	200	195	15.2 ± 0.3	43.8 ± 0.7	198.5 ± 8.5
20	2.0	200	194	18.8 ± 0.4	38.9 ± 0.7	144.8 ± 2.4
Corner point parallel tests						
0	1.0	100	103	16.8 ± 1.2	38.0 ± 0.2	68.7 ± 1.2
0	2.0	100	103	58.2 ± 0.4	16.2 ± 0.1	33.9 ± 0.1
0	1.0	200	209	11.0 ± 0.1	45.3 ± 0.2	230.7 ± 13.0
0	2.0	200	191	46.5 ± 0.1	21.0 ± 0.1	41.7 ± 0.1
20	1.0	100	99	12.6 ± 0.2	42.4 ± 0.0	80.4 ± 0.3
20	2.0	100	101	20.6 ± 0.2	31.4 ± 0.1	49.4 ± 0.4
20	1.0	200	203	9.0 ± 0.2	51.4 ± 0.8	264.8 ± 8.4
20	2.0	200	212	19.2 ± 0.2	34.1 ± 0.3	67.5 ± 0.9
Central point parallel tests						
10	1.5	150	160	19.7 ± 3.1	35.2 ± 0.8	65.0 ± 2.2
10	1.5	150	162	20.1 ± 0.8	34.8 ± 0.7	65.2 ± 1.4
10	1.5	150	163	20.2 ± 3.3	34.0 ± 1.0	65.8 ± 3.3

[a] x is the mean and ϵ denotes the maximum error calculated as $1/2(\max - \min)(n = 3)$.

describe atomization (Kim and Marshall, 1971; Schæfer and Wørts, 1977). For calculation of the ratio the atomizing air flow rate must be known. The droplet size is also affected by the surface tension and viscosity of the liquid (Kim and Marshall, 1971; Yliruusi et al., 1992) and by the density of the liquid and the atomizing air (Kim and Marshall, 1971).

In the accompanying paper (Juslin et al., 1995), the 3^3 factorial design was used to study the influence of three independent variables (atomizing air pressure, flow rate of binder solution and polyvinylpyrrolidone concentration) on the droplet size distribution from a pneumatic nozzle. In the present paper, the effect of different variables was studied by multilinear stepwise regression analysis in order to determine to what extent different variables influence parameters describing the droplet size distribution (volume of droplets under 18.9 μm, median and 90% fractile of droplet size). The volume of droplets under 18.9 μm was chosen as a response variable because it describes the volume of small droplets which was thought to be a critical parameter in granulation. The median and 90% fractile of droplet size were chosen to describe the average size of droplets and the size of large droplets, respectively.

2. Materials and methods

2.1. Study design

The study was based on a 3^3 factorial design. The levels of independent variables and the matrix of experiments are presented in the preceding paper (Juslin et al., 1995).

2.2. Regression analysis

Multilinear stepwise regression analysis was used to study the dependence of response variables (volume of droplets under 18.9 μm, median and 90% fractile of droplet size) on the independent variables atomizing air pressure (P), flow rate of binder solution (Q) and PVP concentration (C). The real flow rates (flow rates were impossible to keep at the target values) were taken into account when creating the regression model but for practical reasons ignored when drawing the response surfaces. In principle extrapolation is not acceptable in this kind of experimental model. However, it was believed that because the response surface plots acted quite regularly, no significant error was made. The analysis was performed by backward selection technique. The general form of the regression equation has been given earlier (Juslin et al., 1995). Only the terms of a significance level of about 5% were accepted. Modelling was performed by Design-Expert Software (v. 3.0.6c, Stat-Ease, Inc., USA). The surface plots were drawn by Graftool (v. 3.3, Graphical Analysis System, 3-D Visions Corp., USA). Analyses of variance were performed using the SYSTAT statistical package (v. 5.0, Systat Inc., USA) to evaluate the levels of significance.

2.3. Materials

The binder solutions studied were purified water and 10 and 20% aqueous dispersions of polyvinylpyrrolidone (PVP) (Kollidon® K25, BASF, Germany).

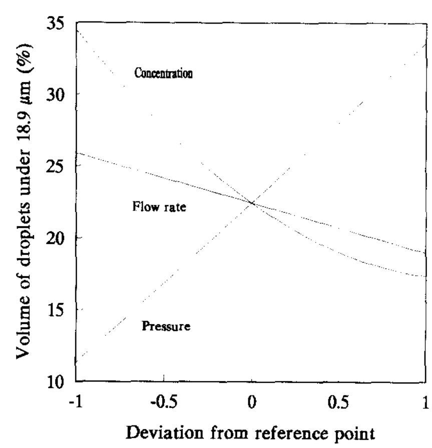

Fig. 1. Effect of independent variables on the volume of droplets under 18.9 μm. Each variable moves from the chosen reference point while the other two variables are kept constant at the reference value (coded zero level of each factor).

2.4. Droplet size measurement

Droplet sizes from a pneumatic nozzle were measured by laser diffractometry (Malvern 2600C Droplet and Particle Sizer, Malvern, UK). A detailed description of the method used for the determination of droplet sizes and droplet size distributions has been given earlier (Juslin et al., 1995).

3. Results and discussion

3.1. Effect of different variables on the volume of droplets under 18.9 μm

Table 1 shows the measured values of the parameters describing the droplet size distribution. Real flow rates are also presented. To facilitate the interpretation of the results, the effect of different variables on the response variables was studied by multilinear stepwise regression analysis. The volume of droplets under 18.9 μm ($V_{18,9\ \mu m}$) is described by the following equation (coded factors) calculated by the regression analysis:

$$V_{18.9\mu m}(P,Q,C) = 22.5 + 11.2P - 3.44Q - 8.57C - 7.36PC + 1.16QC + 3.50C^2 \quad (1)$$

where P is the atomizing air pressure, Q denotes the flow rate of binder solution and C is the PVP concentration. The same equation presented in terms of actual factors is:

$$V_{18.9\mu m}(P,Q,C) = -7.20 + 37.0P - 0.0921Q + 0.302C - 1.47PC + 0.00232QC + 0.0350C^2 \quad (2)$$

The former equation is useful because it immediately shows the effect of different variables on the response variable. In this study, the latter equation was used when creating the three-dimensional response surface. The squared multiple regression coefficient (R^2) was 0.973, indicating a good explanation degree. This means that about 97% of the variability in the volume of droplets under 18.9 μm can be explained by the model. It is observed that there are first-order interactions between the pressure and the concentration, and the flow rate and concentration. The former interaction which has a quite high absolute regression coefficient, 7.36 (Eq. 1), is important.

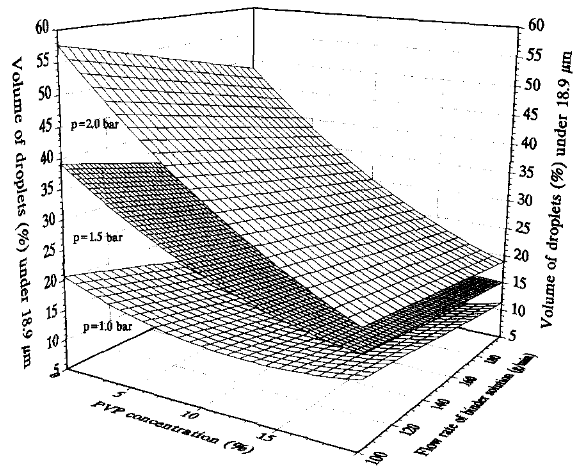

Fig. 2. Three-dimensional surface plot showing the effect of different variables on the volume of droplets under 18.9 μm (atomizing air pressure constant on each surface).

It is seen (Fig. 1) that only the PVP concentration has a quadratic effect on the $V_{18.9\ \mu m}$ (curved line). This perturbance plot shows how the response variable ($V_{18.9\ \mu m}$) changes as each variable moves from the minimum to the maximum value while the other two variables are kept constant at the reference value (coded zero level of each factor). It is observed that changing the atomizing air pressure from −1 (1.0 bar) to +1 (2.0 bar) increases linearly the volume of droplets under 18.9 μm. Increasing the flow rate from −1 (100 g/min) to +1 (200 g/min) causes an opposite effect although this change is not so appreciable: the absolute value of regression coefficient is 3.44 for Q and 11.2 for P (Eq. 1).

The three-dimensional surface plots (Fig. 2) were drawn on the basis of Eq. 2 by giving constant values to the atomizing air pressure. It can be seen that increasing the PVP concentration decreases the volume of small droplets, but not linearly (slightly curved response surfaces). It is also observed that the atomizing air pressure has

Table 2
Analysis of variance: effect of atomizing air pressure (P), PVP concentration (C) and target flow rate of binder solution (Q^t) on the median of droplet size.

Source	Sum of squares	DF	Mean square	F ratio	p
P	2036	2	1018	236	0.000
C	753	2	377	87	0.000
Q^t	431	2	216	50	0.000
PC	119	4	30	7	0.005
PQ^t	54	4	13	3	0.061
Q^tC	19	4	5	1	0.394
PQ^tC	17	8	2	0.5	0.842
Error	48	11	4		

a greater effect on $V_{18.9\ \mu m}$ when the PVP concentration is lower. This is probably due to the effect of viscosity: increasing the amount of PVP in binder solution results in higher viscosity; hence, the same dynamic force of atomizing air is not able to disperse 20% PVP solution as effectively as water. Also, with a low PVP concentration the flow rate affects the volume of small droplets more than with a high concentration because of the change in viscosity. The volume of small droplets varies from about 10 to 60% depending of the levels of independent variables (Fig. 2). It is supposed that a very large number of small droplets may result in the poor granulation of starting materials. This can be explained by the fact that small droplets dry more easily than large ones in the air before reaching the bed, and they can become elutriated (Maroglou and Nienow, 1985).

3.2. Effect of different variables on the median of droplet size ($d_{50\%}$)

According to this study, the atomizing air pressure which has the highest F ratio is the most significant factor affecting the median of droplet size (Table 2). The analysis of variance shows that the flow rate of binder solution and the PVP concentration are also highly significant ($p < 0.001$). To facilitate the interpretation of the results, a reduced quadratic model was constructed to describe the effect of different variables on the median of droplet size ($d_{50\%}$):

$$d_{50\%}(P,Q,C) = 34.5 - 9.22P + 4.16Q + 5.60C - 1.55PQ + 2.29PC + 0.990QC + 2.44P^2 - 1.74C^2, \quad (3)$$

where P is the atomizing air pressure, Q represents the flow rate of binder solution and C is the PVP concentration. The equation in terms of actual factors is:

$$d_{50\%}(P,Q,C) = 60.1 - 43.0P + 0.156Q - 0.0742C - 0.0621PQ + 0.458PC + 0.00198QC + 9.75P^2 - 0.0175C^2. \quad (4)$$

The squared multiple regression coefficient was 0.971, indicating again a good explanation degree. This model shows that all variables studied affect the $d_{50\%}$ and that all factors except the flow rate have a quadratic effect on the median of droplet size. The equations reveal that there are interactions between all variables in decreasing order (absolute regression coefficient values) $PC > PQ > QC$.

Increasing the flow rate and the PVP concentration increases the median of droplet size (Fig.

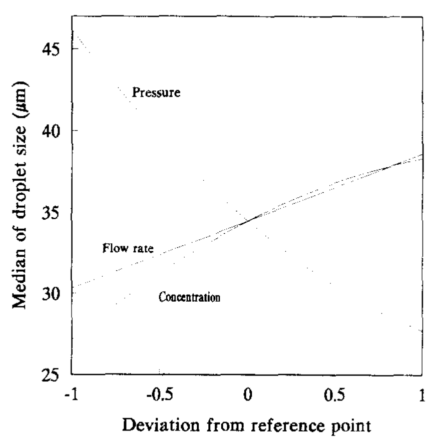

Fig. 3. Perturbation plot illustrating the effect of different variables on the median of droplet size.

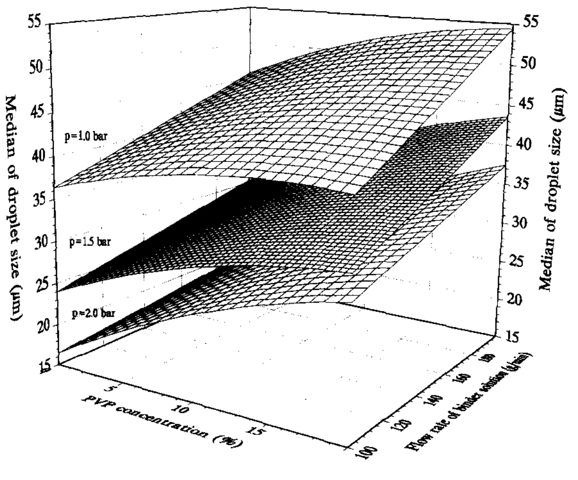

Fig. 4. Dependence of the median of droplet size on the independent variables.

Table 3

Levels of significance according to analysis of variance

Source	$V_{18.9\,\mu m}$	$d_{50\%}$	$d_{90\%}$
P	c	c	c
C	c	c	c
Q^t	c	c	c
PC	c	b	–
PQ^t	–	–	c
Q^tC	–	–	–
PQ^tC	–	–	–

Q^t: the target flow rate of binder solution.
Level of significance: [c] $p < 0.001$, [b] $p < 0.01$, [a] $p < 0.05$ – non-significant ($p > 0.05$).

3). An explanation for the former is the dynamic force of the pressurized air which is not able to atomize the fast flowing liquid so effectively because the liquid flow increases within a certain time. The latter result is in accordance with Schæfer and Wørts (1977) and is due to increasing viscosity with increasing PVP concentration.

The response surfaces (Fig. 4) are slightly curved because of the quadratic terms P^2 and C^2. It is observed that with increasing atomizing air pressure the median of droplet size falls clearly; this fall in mean droplet size has been reported earlier (Kim and Marshall, 1971; Aulton and Banks, 1979; Yliruusi et al., 1992). It can be explained as being due to an increase in the dynamic force of atomizing air with increasing pressure. The response surfaces at each constant pressure are not close to each other describing the dominating effect of the pressure on the median of droplet size. The pressure has more effect on the median of droplet size when the PVP concentration is lower. Fig. 4 also shows that the median of droplet size varies from about 16 to 54 μm in this procedure. Many studies have proved that there is a correlation between droplet and granule size (Thurn, 1970; Schæfer and Wørts, 1978; Waldie, 1991). Hence, it could be assumed that the largest granules will be obtained when the highest PVP concentration and flow rate and the lowest atomizing air pressure are used.

3.3. Effect of different variables on the 90% fractile of droplet size

The effect of different variables on the 90% fractile of droplet size was also studied by the regression model. The following equation in terms of coded factors ($R^2 = 0.913$) was created:

$$d_{90\%}(P,Q,C) = 72.0 - 46.6P + 52.2Q + 25.3C - 37.9PQ + 21.2QC + 36.1P^2, \quad (5)$$

where P is the atomizing air pressure, Q denotes the flow rate of binder solution and C is the PVP

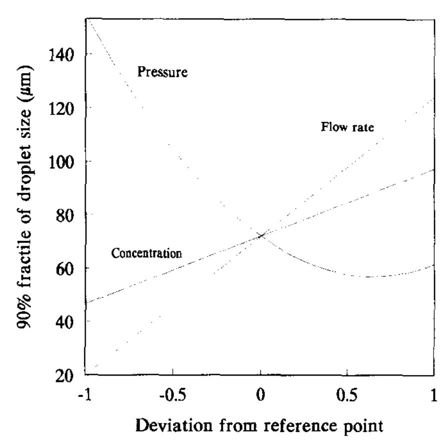

Fig. 5. Perturbation plot illustrating the effect of different variables on the 90% fractile of droplet size.

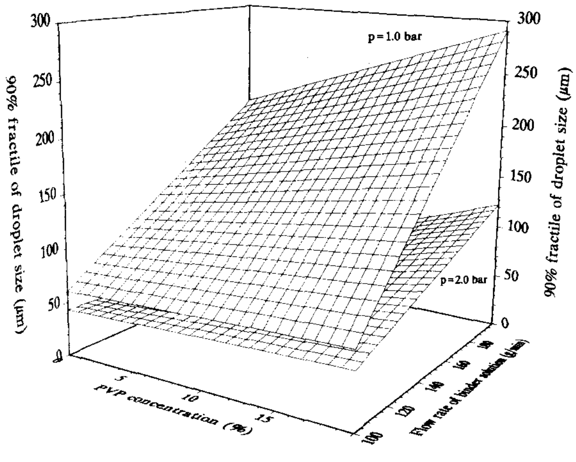

Fig. 6. Dependence of the 90% fractile of droplet size on the independent variables.

concentration. When actual factors are used instead of coded ones, the equation has the following form:

$$d_{90\%}(P,Q,C) = 77.8 - 300P + 2.89Q - 3.82C - 1.51PQ + 0.0424QC + 145P^2. \quad (6)$$

The equations show that only the pressure has a quadratic effect on the 90% fractile of droplet size distribution. There is an important interaction between pressure and flow rate (highly significant, Table 3) and some interaction between flow rate and concentration.

It is seen (Fig. 5) that with increasing pressure the 90% fractile of droplet size decreases in a non-linear way. Increasing the pressure near its highest value does not change the $d_{90\%}$ any further. Both the flow rate and the PVP concentration affect the $d_{90\%}$ linearly: with increasing values the $d_{90\%}$ increases.

Only the lowest and the highest pressures were selected on the response surface plots (Fig. 6) showing the effect of different variables on the 90% fractile of droplet size. The intermediate pressure of 1.5 bar was rejected from Fig. 6 in order to avoid intersecting of the response surfaces. This intersecting might be due to the fact that the model is fitted to measurement results so that the model covers the results well on average or because some extrapolation was made with flow rates. Fig. 6 shows that $d_{90\%}$ varies from about 40 to 290 μm in this study. It is observed that increasing the amount of PVP in binder solution at low flow rate affects only slightly the 90% fractile of droplet size, while at higher flow rate the effect is marked. Not even the effect of pressure is so clear when flow rate is low.

Table 3 summarizes the effect of independent variables on the parameters describing the droplet size distribution according to analysis of variance. It can be noted that some interactions are not significant according to analysis of variance although their regression coefficients are large. This is probably due to the fact that the real flow rates could not been taken into account in the analysis of variance.

Acknowledgements

This study was supported by the Technology Development Centre in Finland (TEKES). The authors wish to thank Jyri Menna B.Sc. (Eng.) for help.

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