INFLUENCE OF SOME OPERATING VARIABLES ON THE CHARACTERISTICS OF GRANULES OBTAINED IN A HIGH SHEAR GRANULATOR

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1. ABSTRACT

This paper examines the influence of the operating variables in a high-shear granulator on granule size distribution and the hardness of the resulting granules. The influence of the water addition method used on resulting granule size distribution was analysed first, it being observed that the results of addition by spraying and by direct pouring were similar. The effect of granule moisture content during granulation time in the different stages: nucleation, growth, and rupture, was also analysed. It was found that when agglomerate moisture content remained constant the growth mechanism prevailed throughout the test. To optimise the operating variables for appropriate granule size distribution in the ceramic production process, a design of experiments (DOE) was performed for two types of rotors. The following variables were studied: rotor speed, barrel speed, and granulation time. The DOE allowed the effect of each variable on final granule size distribution to be determined and the optimum conditions for the granulation of a mixture of red-firing clays in the studied apparatus (moisture content 14,5 %, PIN 1, rotor speed 4800 rpm, barrel speed 35 rpm, and time 8 minutes) to be established.

2. INTRODUCTION

The dry milling and granulation process used to obtain the granulate for pressing ceramic tile bodies enables the characteristic high water and energy consumptions of the wet preparation process to be reduced [1]. The adoption of powder granulation systems is aimed at providing the powder with sufficiently high flowability for appropriate charging in the press mould in order thus to obtain tiles with homogeneous bulk density distribution. This is of vital importance because it assures tile dimensional stability and minimises defects in the fired tiles [2].

Two little studied aspects of the granulation stage are the influence of process variables on granule size distribution (GSD) and hardness. This work was therefore undertaken to examine the influence of a series of operating variables on the development of the granulation stage of a composition for manufacturing ceramic tiles. The study was conducted using a high shear granulator and the effects studied included rotor type, rotor speed, barrel speed, and granulation time.

The purpose was to determine the process variables that allowed maximisation of the granule size fraction between 200 and 500 μm and yielded granules with a similar flowability to that of the granules obtained by spray drying the suspensions.

The influence of the apparatus variables were studied using design of experiments (DOE) as a mathematical tool. This tool allowed the study to be conducted with relatively few experiments, and trends to be obtained that showed the variation of the studied properties with the selected input variables.

3. EXPERIMENTAL

3.1. MATERIALS AND EQUIPMENT

The study was conducted on a red-firing clay mixture customarily used to manufacture glazed stoneware tile. The chemical composition of the mixture is detailed in Table 1.

Composition	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	TiO_2	MnO	L.O.I.
(%)	62.2	17.5	5.70	1.65	1.68	0.16	4.29	0.74	0.04	5.89

Table 1. *Chemical composition of the clay mixture.*

3.2. EXPERIMENTAL PROCEDURE

3.2.1. DEFINITION OF THE INPUT AND OUTPUT VARIABLES

The study was carried out with a high-shear mixer–granulator (Figure 1). In this type of granulator it is very difficult to predict the characteristics of the end product, mainly because the granulation mechanisms (nucleation, growth, consolidation, and rupture) take place simultaneously and more than one can predominate at the same time [3-5]. In addition, during the granulation, there are a wide range of shear and impact velocities in the barrel that affect the material in different zones in different ways [7]. On the other hand, high-speed stirring of the material raises temperature and, consequently, causes the material to dry rapidly. It is necessary, therefore, to determine the effect of the process variables on the resulting granulate, for which a design of experiments with two types of rotors, PIN 1 and PIN 2, was considered (Figure 2).

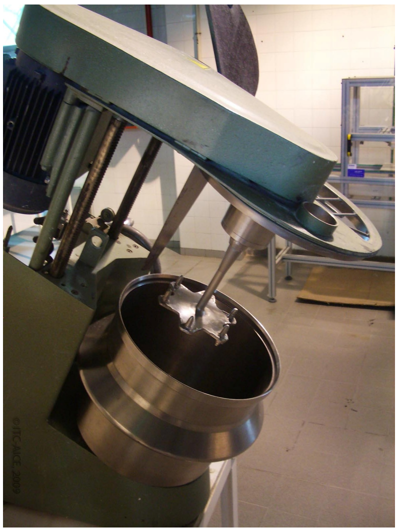

Figure 1. *High-shear granulator*

Figure 2. *PIN 1 (top) and PIN 2 (bottom) rotors*

Using design of experiments enabled a study with relatively few tests to be performed, which allowed trends to be identified that showed the variation of the studied properties or output variables with the selected input variables.

Owing to the great number of variables that influence the process, it was decided to focus the study on the following input variables:

- Type of rotor: PIN 1 and PIN 2 (Figure 2).
- Mixer or rotor tool speed. Six speeds between 900 and 4800 rpm
- Barrel rotation speed: 35 and 70 rpm
- Granulation time

The following response variables were used:

- Statistical granule diameter (D'), corresponding to the diameter below which there was 63.2% of the total mass.
- Uniformity index (n), corresponding to the width of the sample agglomerate size distribution.

The results were analysed by the ANOVA analysis of variance method using Statgraphics software [9].

3.3. CHARACTERISATION OF THE GRANULES

3.3.1. GRANULE SIZE DISTRIBUTION

Granule size distribution (GSD) was determined by dry sieving with a mechanical vibratory screen, using a series of sieves with different mesh apertures and a bottom collector. Sieves with the following mesh apertures were used: 125 µm, 200 µm, 300 µm, 500 µm, and 750 µm. Based on the granule size distributions, obtained by the Rosin-Rammler-Sperling curve fit (RRS correlation [10]), the following parameters characterising the size distribution were obtained:

- D' corresponds to the statistical size of the agglomerate
- Uniformity index n.

RRS fitting equation:

$$\log\left(\ln\frac{100}{R'}\right) = n \cdot \log D - n \cdot \log D'$$

3.3.2. DETERMINATION OF GRANULE HARDNESS/YIELD PRESSURE

Granule hardness was indirectly determined in an INSTRON universal testing machine at a crosshead speed of 5 mm/min. The test consists of forming a test piece by pressing, recording the pairs of values: applied load and powder bed height. Plotting bulk density versus the logarithm of pressure yields a graph from which two parameters directly related to granule hardness and mechanical strength are obtained:

- Bed density (horizontal segment). Bed density is related to granule density by means of the following expression:

$$\rho_{bed} = \Phi \cdot \rho_{granule}$$

- The compactness (Φ) of a spherical material of a single size (in this case agglomerates sized between 300 and 500 µm), after vibration, is constant. Consequently, an increase in bed density in the tests performed would correspond to a higher granule density, owing to lower granule internal porosity.
- Yield pressure (Pf) this being the intercept between the two segments of the graph. Yield pressure depends on several factors, including agglomerate moisture content and microstructure [8].

4. RESULTS AND DISCUSSION

4.1. INFLUENCE OF THE WATER ADDITION METHOD

First a series of tests were conducted to determine how the method of adding the water required for granulation affected this owing to the great influence this has according to the literature [4]. Two extreme conditions were studied: direct pouring or spraying of the water, the samples being extracted from the granulator at two residence times: 1.5 and 6.0 minutes.

Figure 3 shows the granule size distribution obtained at short granulation times (1.5 minutes), while Figure 4 shows this at long times (6.0 minutes). It may be observed that granulation time had an important effect on granule size distribution. As granulation time increased, the largest granule size fraction (>710 µm) and the smallest granule size fraction (< 120 µm) decreased, yielding a narrower granule size distribution.

With regard to the water addition method, no important differences in the results were observed, not even at short granulation times, in which the mixing of both phases might not have been complete. The little influence of the water addition method must be related to the high kinetic energy provided by the granulator rotor, which rapidly mixed the clay composition with water.

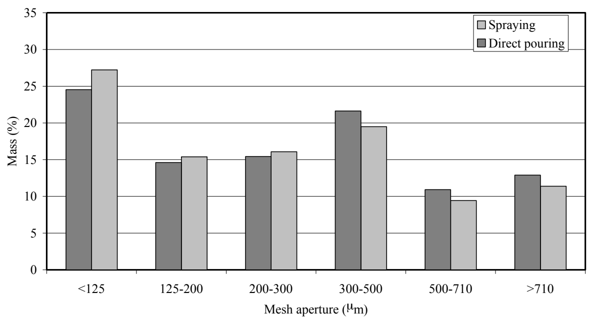

Figure 3. *Comparison of the water addition method at a granulation time of 1.5 min.*

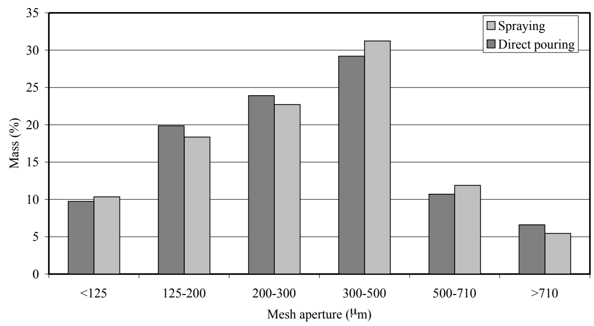

Figure 4. *Comparison of the water addition method at a granulation time of 6 min.*

4.2. INFLUENCE OF GRANULE MOISTURE CONTENT DURING THE GRANULATION PROCESS

Granulation is a complex process that, according to the literature, consists of several stages: wetting and nucleation, granule consolidation and growth, and granule rupture. This section studied the influence of granulation time with the resulting granule size distribution with a view to identifying the mechanisms mentioned.

Since granules dry progressively during the granulation process owing to their movement inside the granulator and the increase in temperature that takes place inside it as a result of friction, experiments were conducted at variable and at constant granule moisture contents.

Figure 5 shows the evolution of the granule size fractions considered with granulation time when the initial water content was 15%. The following was observed:

- The quantity of granules sized larger than 710 µm formed at the start of the test, about 10.4% at 1.5 min., indicated the rapidity with which the liquid was distributed in the clay material, owing to the high stirring speeds that the mixture underwent. At the same time, the great quantity of material that remained below 125 µm (30.2%) indicated that there was still much non-granulated clay material.
- During the first 6 minutes of the test, the fraction below 125 µm decreased dramatically. This indicated that granule growth was taking place; the smaller-sized agglomerates disappeared to form granules of intermediate size fractions, between 200 and 710 µm. The fraction above 710 µm decreased slightly in this stage, augmenting the intermediate size fractions.
- At stirring times between 6 and 9 min., a certain stability in the resulting size distributions was observed, except in the fraction above 710 µm, which continued to decrease until it minimised at 9 minutes (2.3%). The smallest size fraction minimised at 7 minutes (8.0%), after which it increased. The most significant fraction, corresponding to that between 300 and 500 µm, reached its maximum value at about 9 minutes (35.8%).
- Between 9 and 16 minutes, the percentage of smaller-sized granules increased: the fraction below 125 µm grew dramatically, practically in linear form, until the end of the test. The fraction between 200 and 300 µm stabilised at its maximum value during this period, whereas the fraction between 125 and 200 µm continued to increase.
- After 16 minutes, up to 22 minutes stirring, the quantity of granules below 200 µm increased, mainly owing to rupture of the granules between 300 and 500 µm, as the larger-sized granules had almost disappeared.
- After 22 minutes, in addition to the decrease in the fraction between 300 and 500 µm, the fraction between 200 and 300 µm also decreased. The percentage of the fraction below 125 µm continued to increase.

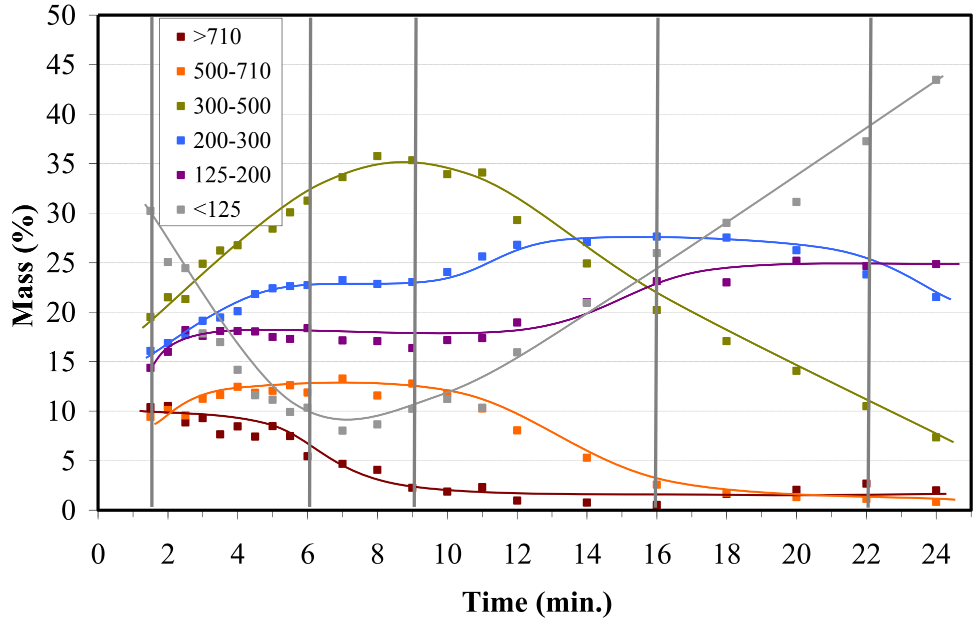

Figure 5. *Evolution of granule size fractions with granulation time at variable granule moisture contents*

The results indicate that, during the first 6 minutes of stirring, the growth mechanism prevailed, as practically all the granule size fractions increased except the size fraction below 125 μm, which logically decreased. Rupture also occurred of the largest agglomerates (fraction >710 μm) which, on forming at the start of the granulation process, had not consolidated and were therefore weak, as will be seen below.

During 6 and 9 minutes, the growth and rupture mechanisms practically took place identically, providing a certain stability in all granule fractions except the granule fraction above 710 μm, which continued to decrease.

After 9 minutes, owing to continuous drying of the material as a result of the increasing temperature in the mixture caused by high-speed stirring, the rupture mechanism prevailed. Thus, the granule fraction below 125 μm increased and the fractions between 300 and 710 μm decreased.

With a view to eliminating the influence of granule drying, a second series of tests were conducted in which granulate moisture content was kept constant. To do so, the moisture content of granules extracted at different times was initially determined (Figure 6), which allowed the water quantity to be determined that needed to be added to the granulator at certain intervals to keep its moisture content constant.

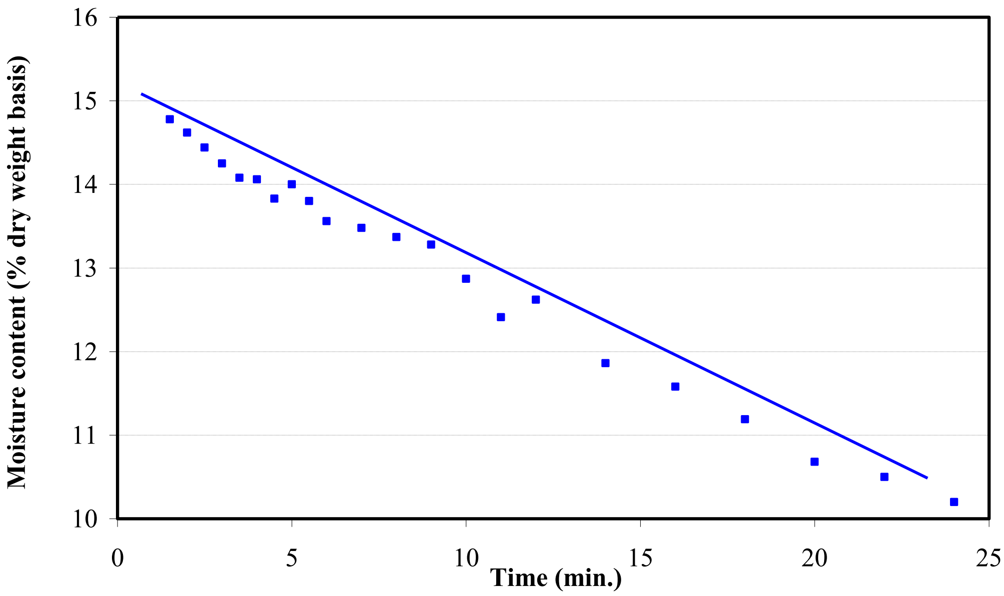

Figure 6. *Evolution of granule moisture content with granulation time*

The results of the evolution of granule size during the granulation process, keeping the moisture content constant, are shown in Figure 7.

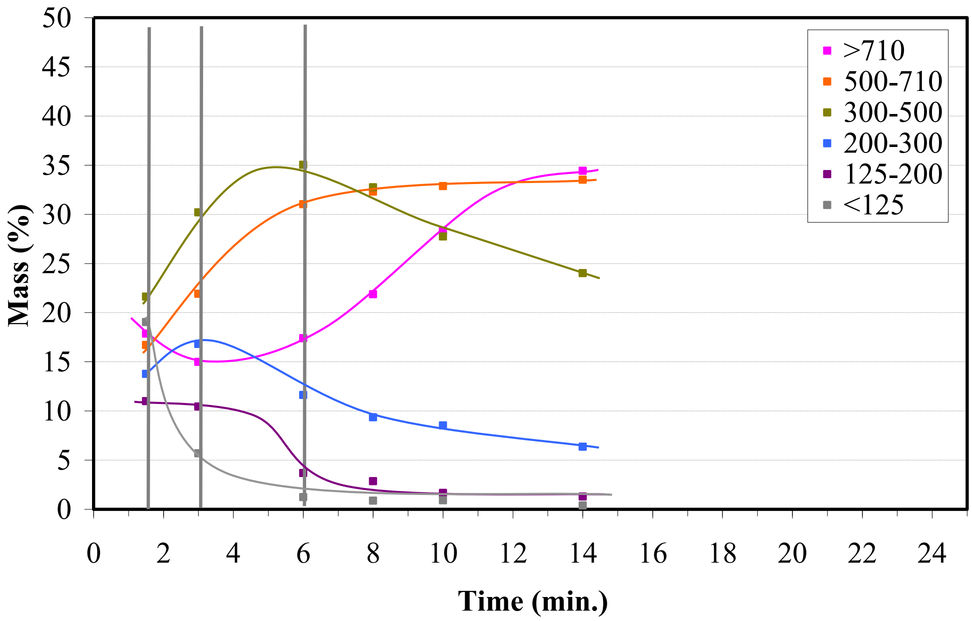

Figure 7. *Evolution of granule size fractions with granulation time at constant granule moisture content*

Comparison of the results obtained in both tests revealed the following differences:

- In the test at constant moisture content, the granules sized below 125 µm almost disappeared at 6 minutes and did not increase at long times, whereas in the test at variable moisture content, this fraction reached its minimum mass at 8 minutes, after which it increased.
- The fraction between 125 and 200 µm also decreased with stirring time in the test in which moisture content was kept constant to form larger-sized granules. This fraction practically disappeared after 10 minutes. In contrast, when drying of the material occurred, this fraction was quite steady throughout the test, exhibiting values of about 20%.
- The quantity of agglomerates of the fraction between 200 and 300 µm increased gradually in the test at variable moisture content, whereas in the test at constant moisture content, its mass decreased after 3 minutes.
- The evolution with time of the fraction between 300 and 500 µm was similar in both tests, but in the test at constant moisture content the maximum value was reached after a shorter time, namely 4 minutes.
- The agglomerate size fraction between 500 and 710 µm increased progressively with stirring time in the test at constant moisture content, whereas in the test at variable moisture content, the value remained practically constant for 10 minutes after which the fraction progressively disappeared.
- The agglomerate size fraction above 710 µm tended to decrease in the test at constant moisture content owing to drying of the material, whereas it increased when further water was added in the test at constant moisture content.

Finally, the evolution of granule bed density (Figure 8) and of granule hardness (Figure 9) with granulation time was determined. In Figure 9, the hardness of the granules obtained by spray drying is indicated with a horizontal line. The results for the tests at variable and constant moisture content were qualitatively similar. It was observed that, as granulation time lengthened, granule hardness increased markedly up to 3-4 minutes, indicating that, during this time, granule consolidation was taking place as a result of the high process energy. After 4 minutes, granule hardness stabilised, indicating that the consolidation process had ended. The increase in granule hardness was due to increased granule density, as evidenced by the evolution of granule bed density, which followed a parallel trend to that of granule hardness.

As may be observed in Figure 9, the hardness of the granules obtained with the high-shear granulator was greater than that of material obtained by spray drying, after stirring times above 2 minutes.

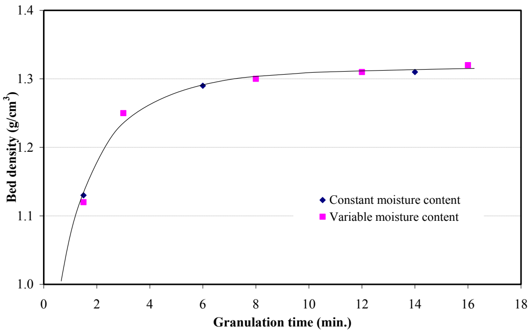

Figure 8. *Bed density*

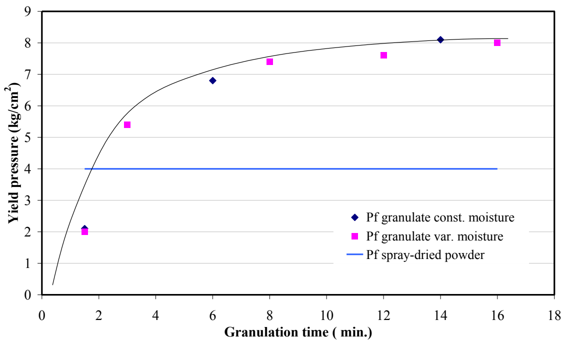

Figure 9. *Yield pressure*

4.3. INFLUENCE OF THE OPERATING VARIABLES IN A HIGH-SHEAR GRANULATOR

The results obtained in the design of experiments conducted with the most important high-shear granulator operating variables according to the literature are set out below [6,7].

Table 2 and Figures 10 and 11 show the results of the Fisher distribution probability for both rotors (PIN 1 and PIN 2) obtained in the design of experiments. All the effects were significant except rotor PIN 1 speed on n (GSD uniformity index).

Input variable	Rotor 1. *Fisher distribution*		Rotor 2 *Fisher distribution*	
	Effect on D'	Effect on n	Effect on D'	Effect on n
A: time	0.0455	0.0002	0.0038	0.000
B: rotor speed	0.0000	0.1081	0.0000	-
C: barrel speed	0.0000	0.0000	-	0.020
BxB	0.0000	-	0.0135	-
BxC	0.0009	0.0026	-	-

Table 2. *Values of the Fisher distribution probability.*

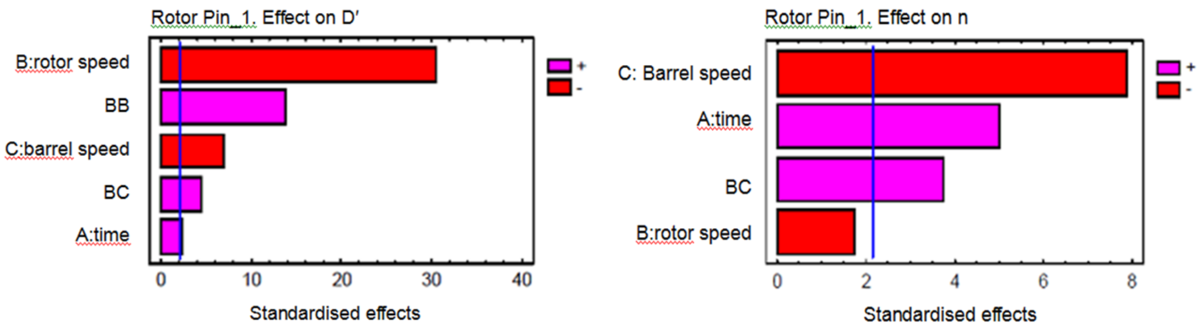

Figure 10. *Significant effects on D and n: Rotor PIN 1*

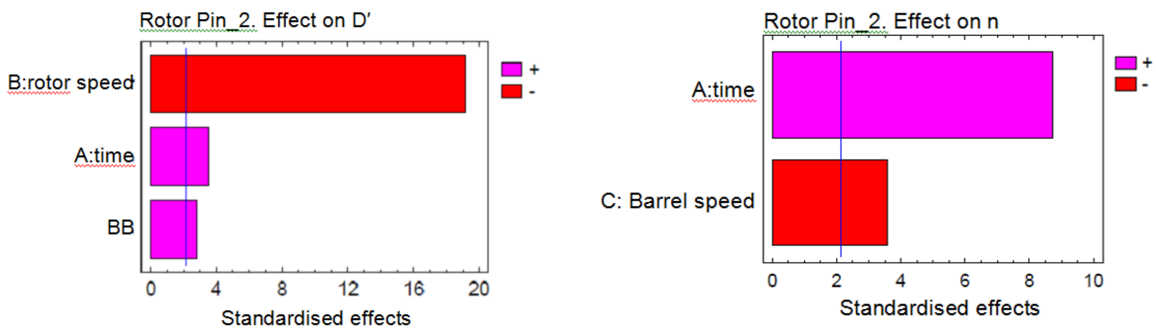

Figure 11. *Significant effects on D and n: Rotor PIN 2*

Figure 10 shows that, for **PIN 1**, the simple effects that most influenced D' (characteristic diameter) were rotor speed and barrel speed, both doing so negatively. Rotor speed was the simple effect that most influenced characteristic size D', such that the greater the granulation rotor speed, the thinner was the resulting agglomerate. In comparison, stirring time hardly influenced average granule size. Barrel speed and granulation time were the simple effects that most influenced the uniformity index: at greater barrel speed, the value of n decreased dramatically and granule size distribution broadened, whereas at shorter granulation time, a more uniform (narrower) granule distribution was obtained. The cross effect rotor speed x barrel speed was also significant with PIN 1.

Figure 11 shows that, for **PIN 2**, the simple significant effects on D' were rotor speed and granulation time. The former had a strong negative influence. That is, when rotor speed increased, the characteristic diameter decreased gradually in all the range. Granulation time had much less effect, this being positive in the 3–6 minute range. After this time, granule size remained invariable. In the effect of the PIN 2 rotor on n, the most significant variable was stirring time, doing so in a positive sense. That is, the longer the stirring time, the higher was the value of parameter n, and therefore the narrower and more uniform was the size distribution. Barrel speed had less effect, and did so in the opposite sense, so that a more uniform distribution was obtained at low speeds.

After the design of experiments analysis, the influence of rotor speed, barrel speed, and granulation time on granule size distribution was compared for both types of rotor.

As may be observed in Figure 12, ***rotor speed*** hardly influenced the uniformity index obtained with both rotors, though it strongly influenced granule diameter.

- With regard to the variation in granule size with rotor speed, rotor PIN 2 produced granules with a larger size at 1800 and 3000 rpm. However, when rotor speed reached 4800 rpm, the rotor used was not a decisive factor in the resulting statistical diameter, which was about 0.5 mm.
- The figure shows that the distributions obtained with rotor PIN 1 were narrower than those obtained with rotor PIN 2 at a given rotor speed, in all the study range.

Barrel speed influenced granule size distribution width more than granule diameter (Figure 13).

- The size of the agglomerates obtained with rotor PIN 2 was slightly larger than that obtained with rotor PIN 1. It may be observed that the influence of the barrel rotation speed used was greater for rotor PIN 1, as for rotor PIN 2 the diameter remained practically invariable at about 1.1 mm.
- Barrel speed was more significant for the uniformity index obtained in the tests conducted with rotor PIN 1 owing to the different movement of the material inside the barrel caused by rotor design.

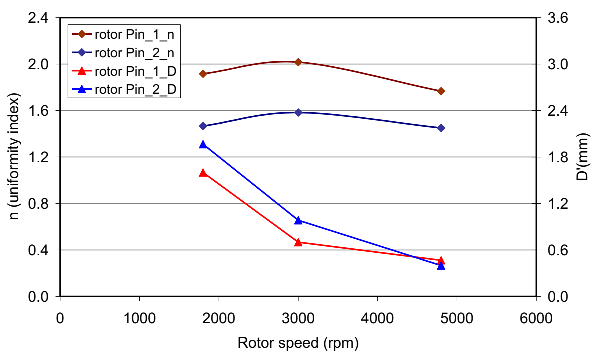

Figure 12. *Influence of rotor speed on granule size distribution*

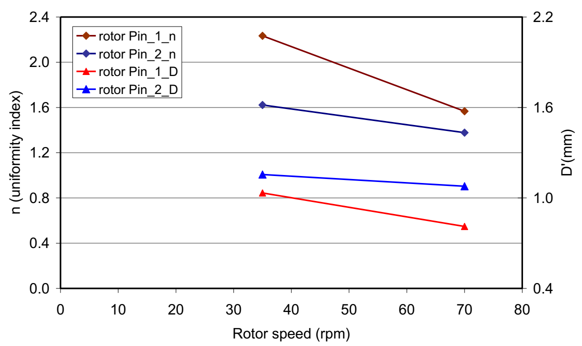

Figure 13. *Influence of barrel speed on granule size distribution*

Finally, ***granulation time*** or stirring time strongly influenced the uniformity of the granule size distribution obtained with both rotors and the granule statistical diameter, as shown in Figure 14.

- The statistical diameters were greater in the tests conducted with rotor PIN 2 between 6 and 9 minutes, while the values obtained in three-minute stirring were practically the same. As granulation time increased, the statistical diameter grew.
- At a given granulation time, under given rotor speed and barrel speed conditions, the agglomerate distribution obtained with rotor PIN 2 was broader and hence less uniform.

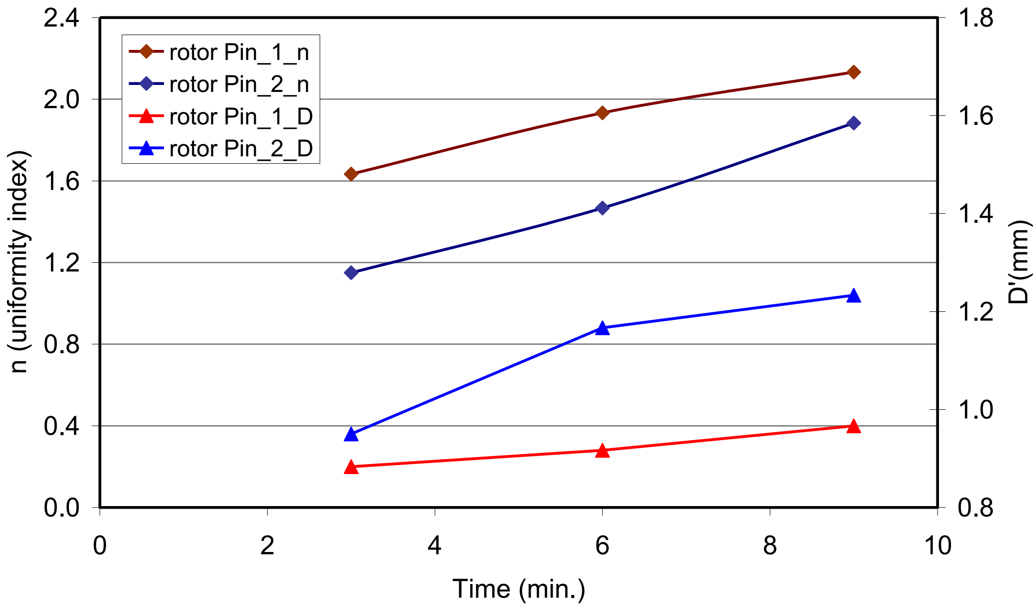

Figure 14. *Influence of granulation time on granule size distribution*

5. CONCLUSIONS

The main conclusions drawn from the study were as follows:

- The water addition method did not influence granule size distribution.
- The following mechanisms were present in the granulation process when water was only added at the start of the process:
  - Granules growth during the first 6 minutes of stirring.
  - Between 6 and 9 minutes, the growth and rupture mechanisms overlapped. In this time period, particle size distribution displayed a certain stabilisation.
  - After 9 minutes, the rupture mechanism prevailed, mainly owing to drying of the material.
- When the agglomerate moisture content remained constant, the growth mechanism prevailed throughout the test.
- During the first 4 minutes, granule consolidation and densification occurred, raising granule hardness and bed density. After this time, both characteristics stabilised.
- The granule size fraction between 300 and 500 μm maximised at about 8 minutes. This maximum practically coincided with the minimum of the fraction below 125 μm under the tested conditions.
- The apparatus parameter that most influenced granule diameter was rotor speed: the greater the rotor speed, the smaller was granule size.
- The apparatus parameters that most influenced the breadth of the granule size distribution were stirring time and barrel speed. Stirring time had a positive influence, so that at longer stirring time, greater size uniformity was obtained. In contrast, barrel speed had a negative effect, as an increase in the highest rotation speed broadened granule size distribution.
- Under given test conditions, the size distribution obtained with rotor PIN 2 was less uniform than that obtained with rotor PIN 1.
- It is possible to obtain granule size curves similar to that obtained with spray-dried powder (D'=0.5 and n=1.93) under the following conditions:

Type of rotor:	PIN 1
Rotor speed (rpm)	4800
Barrel speed (rpm)	35
Granulation water (%)	14.5
Stirring time (min)	8

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