Influence of temperature on the compaction and the mechanical strength of an organicpowder

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INFLUENCE OF TEMPERATURE ON THE COMPACTION OF AN ORGANIC POWDER AND THE MECHANICAL STRENGTH OF TABLETS

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ABSTRACT

Classical works made on powder compaction have been developing studies of parameters like the maximum pressure reached, the punch velocity, the duration of isobaric stage, on tablets properties. But the influence of temperature during the compression has been neglected in most cases. Now, most of pharmaceutical and chemical products, in particular organic products, are sensitive to temperature. In this work, the role of temperature on the compressibility of an organic powder commonly used in the industry has been examined. The consequences on the structure and on the mechanical behaviour of the tablets have been studied. The evolution of the porosity, the tablet strength and the fracture aspects have been followed and then analysed during and after the compression.

KEY-WORDS

Compression ; temperature ; organic powder ; porosity

INTRODUCTION

Classical works made on powder compaction consist in studying parameters like the maximum pressure reached, the punch velocity, the duration of isobaric stage, on tablet properties. But the study of the influence of temperature during the compression has been forgotten or the influence neglected in most cases [1, 2]. As a matter of fact, the behaviour of mineral compounds is not significantly modified by a temperature increase, because of their high melting point and of the low mobility of their atoms or ions at relatively low temperatures. Fragmentation, plasticity and viscosity of mineral powders are not modified by an increase of a few tens of degrees. In the opposite side, most of pharmaceutical and chemical products, in particular organic products, are sensitive to temperature and exhibit quite low melting points. Therefore plasticity and viscosity of such products can easily be modified by an increase of temperature.

The goal of this work is to examine the influence of the temperature on the compressibility of an organic powder commonly used in the pharmaceutical or chemical industry. The consequences on mechanical properties and on the structure of the pellets have been studied.

EXPERIMENTAL PROCEDURES

To begin, a regulation system of the temperature had to be adapted on the die. Then an experimental protocol has been defined. Different studies have been realized to characterise the evolution of the porosity, the mechanical strength of the pellets, and the modification of the fracture surface of pellets as function of the temperature. Results have been linked with the acoustic activity measured during the compression, as done in previous works with cornstarch and aspirin [4].

1. Experimental device and procedure

To make pellets, a 5 kN press LLOYDS has been used. In the cylindrical die, six cylindrical holes have been regularly placed around the centre hole dedicated to the powder. Six heating rods [5] equipped with thermocouples J are inserted in the holes and linked to a temperature regulator TGC 2000 from Setaram. The rods are resistors, which can each, deliver energy of 250 W. The maximum energy delivered by the regulator is 2000 W under 220 V. The temperature regulator has been calibrated before use. Then the die needs a quite long time to get stabilized. For example, to heat the die and reach a constant temperature of 50°C, one hour is needed. A preheating time of

three hours has been established before the insertion of the powder. Once the product is inside the die, a period of 15 minutes has been imposed before to begin experiments. In this way the homogeneity in temperature in the powder is ensured. The temperature is fixed during the compression cycle that is divides in several stages [5].

2. Analyses

2.1. Study of the porosity

Two gauges measure the displacement and the strength applied by the punch on the powder during the compression cycle. These values recorded by the software piloting the press allow calculating the evolution of the porosity of the powder as a function of pressure and of time. The porosity under pressure (n) can be defined by equation 1.

$$n = \frac{Vp}{Vapp} = \frac{Vapp - Vs}{Vapp} = 1 - \frac{m_S}{\pi \text{ x } r^2 \text{x } \rho_S \text{ x } \Delta l} \quad \text{equation 1}$$

V_p: the porous volume, V_{app}: the total volume (pores + grains), V_s: the solid volume, m_s: the weight of powder, r the internal radius of the die (radius of pellets), ρ_s: the density of the powder, Δl: the height of powder in the die.

To get precise values of the porosity, we should know the variations of the density of a product with the temperature, but it is not the case for the product studied. Nevertheless, by considering that such variations present only second order effects, significant conclusions can be deduced anyway.

2.2. Tablet strength

A diametral compression test is realized with a durometer Vanderkamp in order to estimate the mechanical strength of pellets. This test consists in compressing tablets diametrically between two platens with a force increasing up to a maximum F for which the tablet is fractured. The determination of the tensile strength is possible thanks to the relation of Fell and Newton [7] (equation 2). Pellets are considered as cylinders of diameter D and height e. So the value for the maximum tensile stress, R, is constant over the whole of the load diameter and has a magnitude:

$$R = \frac{2F}{\pi x \Delta x e} \quad \text{equation 2}$$

3. Experimental protocol

The organic product used for this study will be called "product A". It shows a visco-plastic behaviour and a melting point close to 150°C. This product of the chemical industry is furnished as a white powder with grains of median diameter 300 µm and a density closed to 1.40 g.cm^{-3}.The goal of this work is to know the influence of the temperature on the compression of the product A. The protocol used is summed up in the table 1.

Table 1. Experimental protocol

Temperature	V_m	P_{max}	t_p	V_d	t_d	T_r
25°C, 60°C and 140°C	10 mm.min^{-1}	50 MPa	30 min	10 mm.min^{-1}	0 min	0 min or 24 h

Other studies have been led to follow the porosity during the pressure increase, n(P), and during the isobaric stage, n(t_p). The mechanical strength, R(T), of tablets has also been measured for tablets made at different temperatures. Then a SEM study on the fractured faces of the pellets has allowed us to observe the evolution of the internal structure of pellets. Finally the XRD analysis has gave us information on the evolution of the degree of cristallinity of the powder.

RESULTS AND DISCUSSION

1. Effects of the compression cycle parameters

At the beginning of the pressure increase, grains organise themselves without noticeable deformation (simple packing). Indeed, the initial volume of powder is quite aerated, so that grains can easily move under the local shear and axial stress to reach a higher compacted state (blocked state). This state is characterised by an important number of contacts per grain. When this state is reached and pressure continues to increase, grains are deformed (elastically or viscously) and/or are fractured. The plastic irreversible deformations appear instantaneously during the pressure increase. During the isobaric stage, the viscous deformations of the granular medium submitted to a constant axial stress are also irreversible. These deformations of the grains provoke more and more important rearrangements of the granular medium. These deformations allow a better cohesion between grains. The packing fraction still increases. During the decrease of pressure, elasticity effects may appear in pellets, sometimes leading to a loss of cohesion of the tablets. This is not the case of the product A.

At the end of the compression cycle, before or after the tablet ejection, the reorganisation of the compact continues. The global volume of tablets generally increases. This phenomenon is considered as a viscous relaxation of the shear stresses.

1.1.Evolution of the porosity during the pressure increase

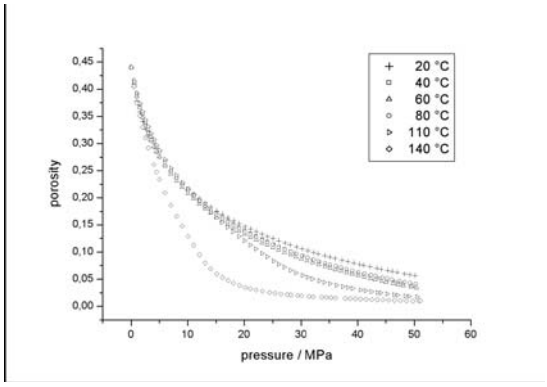


Figure 1. Evolution of the porosity of the product A during the pressure increase for several temperatures.

The temperature at which the compression is realised has a noticeable influence on the compression of the product A (figure 1). Under 100°C, the curves are similar. That allows us to conclude that temperature presents no marked influence during the pressure increase for temperatures less than 100°C. The types of reorganisation must be similar and not modified in this temperature range. On the opposite side, the shape of the curves is completely changed for temperatures higher than 100°C. The explanation probably lies in different types of reorganisation.

1.2. Evolution of the porosity during the isobaric stage

The influence of the temperature on the evolution of the porosity during the isobaric stage is important (figure 2). Up to 80°C, the porosity decreases with the raising temperature. But for temperature higher than 80°C, the shape of the curves is completely modified: the temperature becomes close to the melting point of the powder and this modifies significantly the powder behaviour. To plot curves, equation 1 was applied with the density of the pure product 1.40g.cm^{-3}. But near the melting point, the product may begin to become very ductile and even to melt a little bit in some regions and its density may change. In consequence, the porosity calculated with the equation 1 for temperature higher than 100°C does present higher uncertainties.

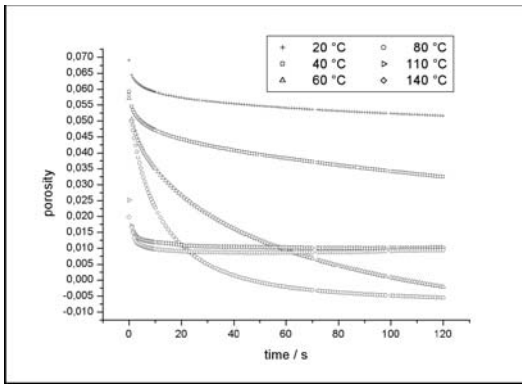


Figure 2. Evolution of the porosity of the product A during the isobaric stage for several temperatures.

2. Influence of the temperature on the mechanical strength

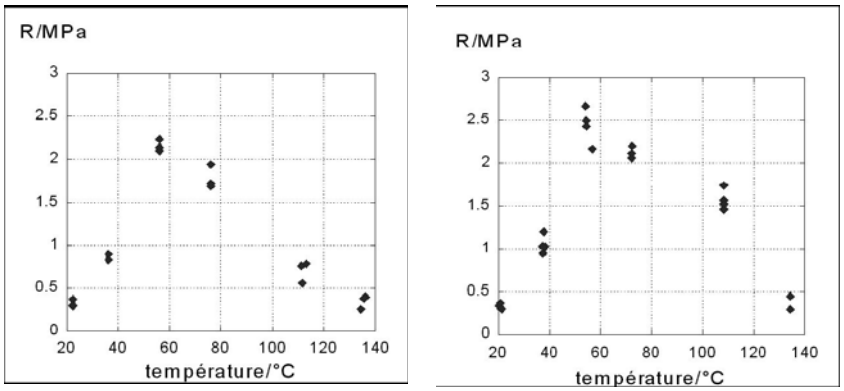
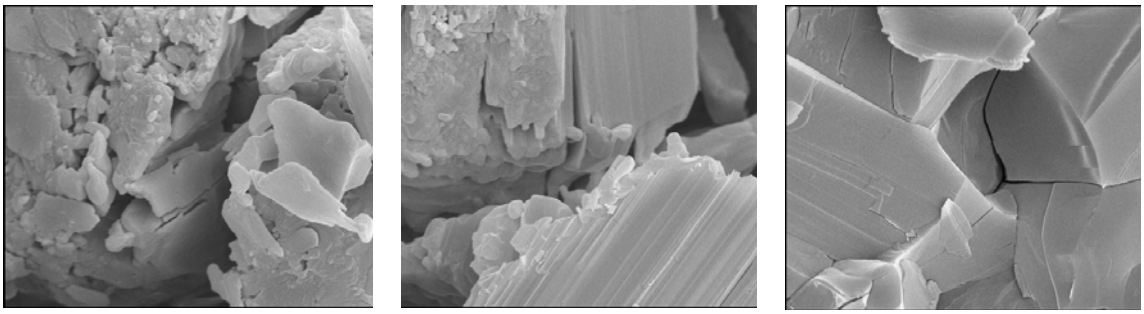


Figure 3. Results of mechanical strength obtained by diametral compression tests after the ejection of pellets (t_d =0) and a relaxation time (t_r=24 hours)

Figure 3 shows a great influence of the temperature on the tablet tensile strength. Whatever the relaxation time after the ejection is, the tensile is maximal around 60°C.
Between 20 and 80°C, the evolution of the porosity under pressure (figure 1) does not depend on the temperature in opposition to the evolution of the mechanical strength that increases with the temperature (figure 3). The A.E., strongly connected to the rate of fragmentation, also increases with the temperature [5]. In fact, the growing number of fragmentations creates new areas of contact between grains, which improve the mechanical strength. The relaxation time has only a small influence. Whatever the temperature is, the tensile strength needed to break a pellet is increased by the relaxation time.

3. Evolution of tablet cohesion

Fractured pellets have been analysed by SEM to observe their internal structure. The photo 1a shows us grains of product A well visible at the fracture surface of a pellet made at room temperature. Any strong deformation can be seen in this typical intergranular fracture. A zoom on the grain joint (photo 1a) shows that grains are not completely stuck together (high porosity). The photo 1a shows that grains are constituted of crystallites whose mean size is about 10 µm.



a b c

Photo 1. Fractured face of a pellet made at 20 °C (a), 60 °C (b), 140 °C (c).

In pellets made at 60 °C, the grains are less visible (photo 1b). A more transgranular type fracture has occurred inside the grains, between crystalline planes. This observation is confirmed by the photo 1b where stripes can be seen. The temperature increase has favoured the grains cohesion and then fractures have to happen inside grains and not between the grains. This could be an explanation to the increase of the tablet tensile strength.

In pellets made at 140°C, grains have disappeared at the benefit of a continuum of matter (photo 1c). There is no more free space between grains and porosity becomes low enough. Fracture is now clearly transgranular. The facets on photo 1c confirmed this type of fracture. The higher the temperature, the higher the cohesion and more grains are transformed in a continuum of matter. But during the cooling (and after ejection of the pellet), cracks may appear and propagate inside pellets. As a consequence, pellets become brittle and the mechanical strength decreases. Cracks are visible on photo 1c.

4. Evolution of the structure

Diffractograms of pellets made at room temperature and at 140°C are similar. Their lines are identically placed (showing a same crystalline structure) but their intensities are different. Seeing no influence of the compression temperature on the crystalline structure of tablets, we have decided to realise the XRD analysis directly on the powder. This time, a variation can be noticed in the intensity of the peaks (orientation effect) and in their positions. When temperature increases, the lines slightly move either to the left or the right depending on the Miller indexes when temperature increases. But these displacements are not important enough to conclude to a new crystal lattice. The only phenomenon explaining these observations seems to be a very small dilatation of the elementary cell of the crystallites when the temperature increases. A refinement of the elementary cell parameters developed by a least square method for diffractograms obtained at different temperatures don't show great changes in the values of the elementary cell.
The hypothesis made in the first part of this work supposing that the variation of the true powder density with temperature is therefore verified.
The maximum in the mechanical strength cannot be explained by this small and monotonous modification of the lattice.

CONCLUSIONS

Several characterisations have been made during the study of the compression of product A at different temperatures.
Then the study of the pressure increase stage of the compression cycle shows a reduction of the porosity for temperatures higher than 100 °C. Whereas the diametral compression test notes a maximum of tensile strength around 60°C. Moreover the evolution of porosity versus time during the isobaric stage depends strongly on temperature.
In addition, the SEM analysis of the fracture faces of pellets shows a transformation of the tablet behaviour from individual grain structure and intergranular type rupture (T=20°C) to a structure

of matter continuum and intragranular type fracture for temperatures higher than 60 °C. This modification of the fracture type influences the mechanical strength of pellets. Indeed, on the first hand raising the temperature allows a gain in plasticity and then in cohesion and hardness. On the other hand, for temperature close to the melting point, at the ejection of the die a hard cooling creates cracks in tablets and a decrease of the mechanical strength occurs.
This study shows that an optimum temperature exists to get a better cohesion in tablets of organic materials. The temperature of 60 °C for the product A seems to be the good choice to enhance cohesion and good mechanical strength.

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