Differences between eccentric and rotary tablet machines in the evaluation of powder densification behaviour

Giovanni F. Palmieri [a,*], Etienne Joiris [b], Giulia Bonacucina [a], Marco Cespi [a], Annalisa Mercuri [a]

[a] *University of Camerino, Department of Chemical Sciences, via S. Agostino 1, 62032 Camerino, Italy*
[b] *University of Lille II, Faculty of Pharmacy, Department of Biopharmacy and Clinical Pharmacy, Rue du Professeur Laguesse, Lille, France*

Abstract

Differences in the dynamics of powder densification between eccentric and rotary machine were pointed out by compressing at different compression pressures microcrystalline cellulose, lactose monohydrate and dicalcium phosphate dihydrate and recovering the corresponding stress/strain data in both machines equipped to monitor punches displacement and compression forces. Heckel plots were then obtained from these stress/strain data.

Curves obtained in the rotary machine possess a narrower zone of linearity for the calculation of P_Y and D_A. The effect of the different compression mechanism of the rotary machine on the shape of the Heckel plot is more noticeable in a non-deforming material such as dicalcium phosphate. The effect of the longer dwell time of the rotary machine on the porosity reduction occurring after the maximum pressure has been reached, is more noticeable in a ductile material such as microcrystalline cellulose.

Heckel parameters obtained in the rotary press are in some cases different from those recovered in the eccentric machine because of the longer dwell time, machine deflection and punch tilting occurring in the rotary machine, although theoretically they could better describe the material densification in a high speed production rotary machine.
© 2005 Elsevier B.V. All rights reserved.

Keywords: Compression; Compaction; Tableting; Mechanical properties; Rotary tablet machine; Densification; Heckel; Stress; Strain

1. Introduction

Tablets are the most common pharmaceutical dosage form but the compression of powdered or granular material into a cohesive mass is a complex and irreversible dynamic process, in contrast to its apparent simplicity.

Mechanically, the process consists of imposing a progressive strain on the powder confining it to a certain final volume and porosity.

The dimensional constraints imposed by the punches and die are then removed and the compact is allowed to relax.

* Corresponding author. Tel.: +39 0737 402289; fax: +39 0737 637345.
E-mail address: gianfilippo.palmieri@unicam.it (G.F. Palmieri).

The compacted material exerts a certain stress against the punches and die during this process in response to the imposed strain.

Generally, a solid material can be characterised from a rheological point of view using a creep test (Tsardaka and Rees, 1989; Celik and Aulton, 1996) and dynamic mechanical analysis (Radebaugh et al., 1989).

Besides, useful information concerning the fundamental structure of tablets can be drawn from compression stress/strain data and researchers have accomplished many efforts in order to develop methods of evaluation based on these data. In fact, in this way, the formation of the compact and its subsequent behaviour under stress, inside the die of a tablet machine, can be monitored.

For instance, among these methods, force/displacement curves allowing the evaluation of the energy expenditure during powder compaction firstly proposed by Nelson et al. (1955) and the plot of minus logarithm of tablet porosity versus compression pressure (Heckel, 1961a,b) have been widely applied.

In particular, this last technique of analysis has recently become the most used due to the fact that it gives a good level of information about the dynamics of powder densification in the die.

The Heckel equation is:

$$\ln\left[\frac{1}{1-D}\right] = KP + A$$

where D is the relative density and $(1 - D)$ denotes the pore fraction, P the applied pressure, K the slope of the straight linear portion of the plot and the reciprocal of K is the mean yield pressure (P_Y), and A is the intercept of the prolonged linear portion of the plot with the Y axis and is the sum of two densification terms:

$$A = \ln\left[\frac{1}{1-D_0}\right] + B$$

where D_0 is the initial relative density and B is the densification due to the slippage and rearrangement of primary and fragmented particles.

So, the relative density at point A is $D_A = 1 - e^{-A}$ and the increase of relative density due to slippage and rearrangement is $D_B = D_A - D_0$.

The use of this method requires the measurement of the force as it varies during the punch penetration inside the die.

Compaction simulator represents a powerful mean to collect these data, particularly when material characterisation is needed, since it can reproduce different compression kinetics (Rees et al., 1972; Celik and Marshall, 1989; Muller and Augsburger, 1994).

Otherwise, single station eccentric presses are still used to acquire the stress/strain data. In fact, in this type of machine, only the upper punch penetrates the die to compress the material whilst the position of the lower punch does not change (except deflection) during the tablet formation. For this reason, eccentric presses can be easily instrumented to measure the axial forces exerted by both punches and the distance moved by the upper punch.

Anyway, an eccentric press cannot reproduce the compression conditions occurring on a rotary multi-station press in which the lower and upper punches both move and penetrate inside the die.

Moreover, there can be considerable differences in relative punch speed penetration between the low velocities encountered on eccentric presses and the higher velocities possible on the rotary presses. Since compact formation is based on time-dependent viscoelastic properties, the speed of the process can have marked effects on compactibility and tendencies to lamination, capping and picking.

It is easy to instrument a rotary press to measure changes in punch force during a compression cycle, but the measurement of the punch displacement poses some formidable problems due to the difficulties of retrieving signals from the moving punches and turrets.

Punch displacement of rotary tablet machines was calculated by equation (Rippie and Danielson, 1981; Charlton and Newton, 1984) as a function of time, from the profile of the punch head, machine dimensions, turret velocity and punch position relative to the compression roller. Unfortunately, in this way, punches and machine deflections cannot be taken into account.

Upper and lower punches of a rotary press were then instrumented to measure both force and displacement (Walter and Augsburger, 1986). Two linear variable displacement transducers were mounted in the empty punch and die sockets adjacent to the station holding the instrumented upper and lower punches. These transducers were linked to the punches through rigid brass linkages. Data were collected from the rotating turret through an eight-channel mercury swivel system. Dis-

placement data were obtained by an umbilical cord that was designed to break away following compression.

A similar device was then mounted on one station of a rotary machine, although in this case the signal acquisition was performed with the aid of a modular radiotelemetric device (Muller and Augsburger, 1994). Brackets were constructed to affix the telemetry unit and transmitter to the underside of the upper turret.

More recently, it was demonstrated (Matz et al., 1999) by using a partially modified portable device that the correct value of punch displacement is also influenced by punch tilting and this phenomenon should be minimised or corrected by the use of two transducers per punch for a precise measurement of punch displacement. Punch tilting was maximal before the appearance of the force and also at the beginning of the increase of the force.

Nowadays, there are several devices (even portable) available on the market. These are telemetric devices with transducers mounted beside punches in extra holes or instead of punches by occupying free stations of the rotary press.

A commercial device (Ronchi) (Fig. 1) was mounted on one of the 10 stations of a rotary tablet machine (Ronchi, Cinisello Balsamo (MI), Italy). This device does not work telemetrically but transducers signals are directly linked to the relay unit, and then managed by computer software. Transducers do not occupy free stations but are arranged in specific extra holes in the turrets beside those of the displacement-controlled upper and lower punches. These punches were equipped immediately below their head, with a narrow flat steel arm that ended just above the transducer. Since it is a fixed device, once mounted and calibrated, the whole system performs very reproducible analyses. In the specific case, LVDT transducers having an accuracy of $\pm 4\,\mu$m were used.

Transducers holes immediately succeeded punches holes and were very close to them, in order to minimize the effect of punch tilting on the transducers (Ridgway Watt, 1988; Matz et al., 1999). The distance between punch and transducer was 0.4 cm and the steel arm was only 12 mm long.

The aim of this work was not to improve the current knowledge in the rather studied field of powder compaction mechanism, but to highlight and describe some similarities and differences occurring between eccentric and rotary tablet machines, when methods usually
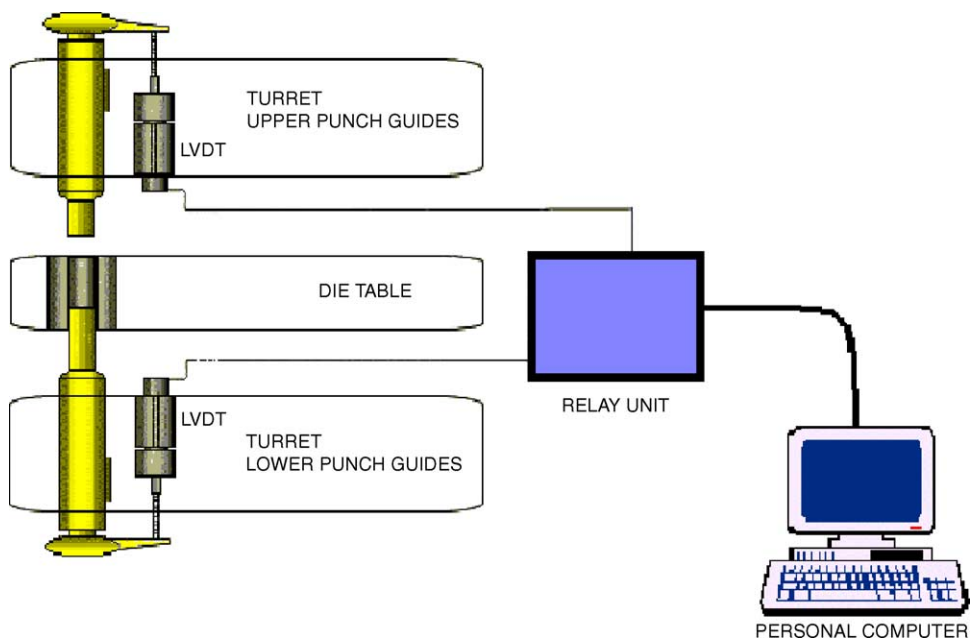

Fig. 1. Device for punch displacement control.

used to study the densification mechanism of solid particulate materials are applied.

So, Heckel analyses were carried out on microcrystalline cellulose, lactose monohydrate and dicalcium phosphate dihydrate using either a rotary or an eccentric machine and experimental parameters as similar as possible between the two machines. These materials were chosen because they are frequently used as fillers in tableting processes and they possess different densification behaviour. Microcrystalline cellulose is known as a deforming material and time is an important parameter in the stress/strain correlation. On the other hand, dicalcium phosphate densification mostly occurs by fragmentation and strain directly depends on the applied stress. Lactose has intermediate properties. Concerning the Heckel equation, in order to make a clearer distinction between densification due to the movement of the original particles and that due to the brittle fracture, D_0 and subsequently D_B were modified by using a relative pre-compression density D_0' term which includes the initial rearrangement of particles (Doelker, 1994). So, D_B' was calculated as follows:

$$D_B' = D_A - D_0'$$

where D_B' is only representative of the densification due to fragmentation.

2. Materials and methods

2.1. Load cells check

Before performing the Heckel analysis for the three materials, the correct load cells calibration was checked in both machines. Dicalcium phosphate was compressed using 11.28 mm flat faced punches at pressures ranging between 25 and 250 MPa, using very similar kinetics of powder-bed reduction. These kinetics were calculated taking into account the punch penetration speed into an empty die for both machines (Fig. 2). In the figure, time 0 is not the beginning of penetration and only the last 110 ms (the most significant) before the maximum penetration are shown. The porosity of the ejected tablets was calculated and reported against the compression pressures for both machines (Fig. 3). In this way, despite an absolute evaluation of the calibration is impossible, if the same trends are obtained, in
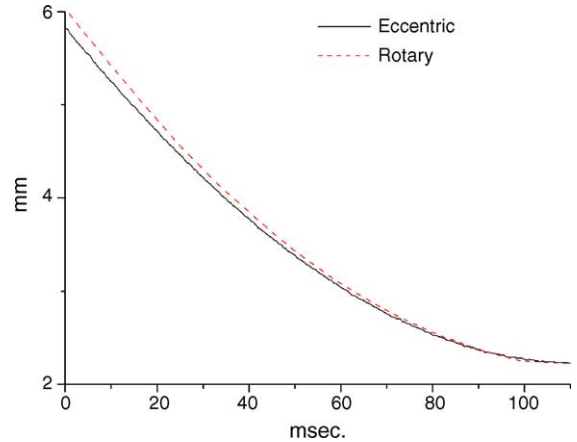

Fig. 2. Kinetics of powder-bed reduction in the eccentric and rotary tablet machines.

high probability the load cells of both machines are well-calibrated. In any case, eventual further differences in Heckel parameters between the two machines cannot be attributed to a different load cell calibration. Emcompress was chosen for this preliminary phase since its densification mechanism mostly depends on the applied pressure.

2.2. Displacement transducers check

Transducers calibration and validation was performed by the Ronchi Company using a micrometer screw (Mitutoyo, Tokyo, Japan) since punch displacement device is directly part of the machine instrumentation. Zero penetration for the lower punch was set at the minimal punch penetration position inside the die.
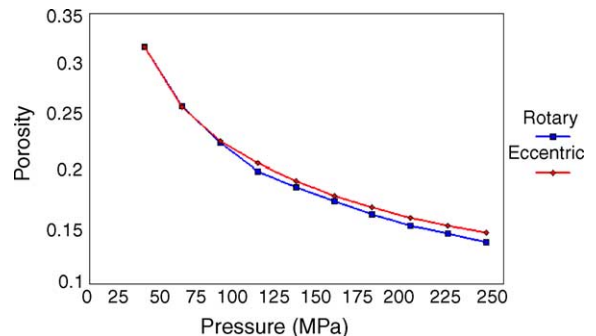

Fig. 3. Porosity of Emcompress ejected tablets at different compression pressures.

Zero penetration for the upper punch was set as the contact point between the punch and a blind steel die at zero force.

In any case, the transducers functionality was checked by the authors using Mitutoyo steel gauge blocks.

2.3. Densification on the eccentric tablet machine

Microcrystalline cellulose (Avicel PH 102, FMC Europe NV, Brussels, Belgium), lactose monohydrate (Pharmatose 50, DMV, Weghel, The Netherlands) and dicalcium phosphate dihydrate (Emcompress, Penwest, Reigate, Surrey, UK) were compressed with an instrumented Frogerais OA single punch tablet machine (Frogerais, Vitry, France) equipped with 6 mm flat-faced punches, by introducing manually the powder into the pre-lubricated die (magnesium stearate slurry in acetone) and adjusting the weight of the samples in order to obtain the desired pressures. Five replicates cycles were performed for the three substances at maximal punch pressure of 100, 150, 200 MPa. For a single compression cycle, both the compression pressures on the upper and lower punches and the displacement of the upper punch were measured and recorded at a frequency of 400 Hz. Correction of the displacement transducer data for machine looseness and punch deformation was carried out (Juslin and Paronen, 1980). Heckel profiles (in die method) were generated from single compression cycles. D_A, D'_0 and D'_B were calculated using a pre-compression pressure value of 1.5 MPa and P_Y was calculated from the right portion of the plots (50–90 MPa). Relative density at the end of the compression cycle (D_{FIN}) was calculated from the last point of the decompression portion of the curve and relative density of the ejected tablet (D_{EJC}) was calculated immediately after the ejection. Each value further presented is the mean of five measurements.

2.4. Densification on the rotary tablet machine

Microcrystalline cellulose, lactose monohydrate and dicalcium phosphate dihydrate were compressed with an instrumented 10 stations Ronchi rotary tablet machine (Ronchi) equipped with 6 mm flat-faced punches, by introducing manually the powder into the pre-lubricated die (magnesium stearate slurry in ace-

tone), after having blinded nine stations and adjusted the weight of the samples in order to obtain the desired pressures. Five replicates cycles were performed for the three substances at maximal punch pressures of 100, 150, 200 MPa and compression speed of 25 rpm. This speed was chosen in order to use a very similar speed of powder-bed reduction between eccentric and rotary machines (Fig. 2). In fact, machine speed influences Heckel parameters since time influences visco-elastic phenomena (Muller and Augsburger, 1994; Gabaude et al., 1999). For a single compression cycle, the compression pressure and the displacement of the upper and lower punches were measured and recorded at a frequency of 400 Hz. Correction of the displacement transducers data for machine looseness was not necessary since the transducers position in the turrets (Fig. 1) enabled the automatic detection of machine deflection. The correction of punch deformation was carried out point by point according to the following equation:

$$D = \frac{FL}{ES}$$

where D is the punch deformation (mm), F the applied force (kN), L the punch length (mm), E the steel rigidity modulus (kN/mm^2) and S is the punch section (mm^2).

The equation is valid below the limit of steel elasticity that is by far higher than the pressures used to perform the analyses.

Besides, for a more precise correction, punch length was divided in two parts: punch stem (20 mm diameter) and punch neck (6 mm diameter).

Heckel profiles (in die method) were generated from single compression cycles. D_A, D'_0 and D'_B were calculated at a pre-compression pressure of 1.5 MPa and P_Y was calculated from the right portion of the plots (50–90 MPa). This interval was chosen because it can be used even when a maximal punch pressure of 100 MPa is applied and because, as further shown, Heckel plot changes a fraction its slope after 100 MPa for the plastic material (Avicel PH 102). Relative density at the end of the compression cycle (D_{FIN}) was calculated from the last point of the decompression portion of the curve and relative density of the ejected tablet (D_{EJC}) was calculated immediately after the ejection. Each value further presented is the mean of five measurements.

2.5. True densities of the materials

True densities of the microcrystalline cellulose, lactose and dicalcium phosphate were calculated using a helium pycnometer (AccuPic 1330, Micromeritics).

3. Results and discussion

Even if the aim of the authors was, first of all, to point out the macroscopic differences on the trend of the Heckel plots obtained on a rotary machine, in comparison with those obtained in the more familiar (for this type of analysis) eccentric machine, they tried to reproduce the same kinetics of powder-bed reduction (strain rate) in both machines, despite the difference in the mechanics. After having fixed the minimal distance (2.25 mm) between upper and lower punches in an empty die in both machines, this was kept constant and the speed of the rotary machine was changed until a strain rate as similar as possible to that of the eccentric machine was found as shown in Fig. 2. In any case, when a material is inside the die, the strain rate would inevitably be a little different, even if exactly the same in an empty die, since rotary machine deflection is different. In this way, any eventual differences in the recovered Heckel parameters between the two machines cannot be ascribed to different compression kinetics and, at the same time, the effect of the longer dwell time typical of the rotary press can be investigated.

The porosity of the preliminarily prepared tablets of Emcompress (Fig. 3) is identical in both machines up until a compression pressure of 75 MPa. At higher stresses, the porosity of the tablets prepared using the rotary machine is a little lower and this difference is softly but gradually more noticeable as the pressure increases. On the basis of these results, the authors decided to use the 6 mm diameter flat punches for the core of the study since in this way it is possible to reach compression pressures as high as 200 MPa without exceeding a compression force of 6 kN. This small difference in porosity may not completely depend on a different calibration of the load cells between the two machines but could be the result of the different dwell time occurring in them. In an eccentric machine, the upper punch after having reached the maximum penetration does not remain in that position but immediately
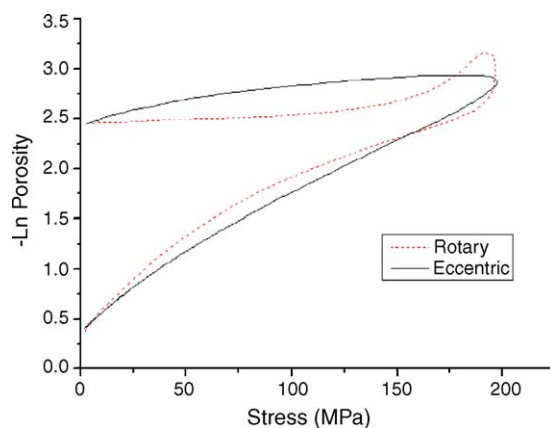

Fig. 4. Heckel plots of Avicel PH 102 in eccentric and rotary machines.

returns back. In the rotary machine, both upper and lower punches penetrate the die and, what is more relevant, they both possess a considerable dwell time. In fact, when a punch reaches the maximum penetration corresponding to the lowest point of the upper roller (for the upper punch) and to the highest point of the lower roller (for the lower punch), it remains in that position until all the flat portion of the punch head has passed the roller. The phenomenon is not neglectable since it takes more or less a quarter of a compression cycle, intended as the time between the appearance and the disappearance of the force. Of course, dwell time mainly influences the porosity of the tablets made of deforming materials but even a filler such as Emcompress may slightly feel the effects of it.

Fig. 4 shows the Heckel plots obtained at 200 MPa by compressing in the die Avicel PH 102, as an example of the different plots between eccentric and rotary machines. As expected, plot obtained by the eccentric machine is that of a typical deforming material and is nearly linear from the beginning until the maximum compression pressure. Then, the usual immediate elastic recovery is shown by the decompression curve.

The plot from the rotary machine is different and in general less linear. The same material appears better deformed in the rotary than in the eccentric machine, but after more or less 100 MPa the curve gradually changes its trend and the slope decreases. Finally, in the range 190–200 MPa, there is a fast porosity reduction. After having reached the maximum pressure, during the first phase of pressure reduction, tablet densification

continues and it is only after a further stress decrease that there is the consequent porosity increase. The densification reduction (elastic recovery) shown by the return curve is more immediate in the rotary than in the eccentric machine, even if the final points of the return curves are similar. Of course, the elastic modulus of a material does not depend on the type of machine and it is difficult to believe that a stress reduction is accompanied by a porosity reduction. Despite a little effect of punch tilting cannot be excluded, these phenomena can be explained by the considerable dwell time present in a rotary tablet machine. In fact, during this time, punch should maintain its position and insist over the material, inducing a certain further deformation on it, due to the stress relaxation of the plastic component (David and Augsburger, 1977; Dwivedi et al., 1991; Hiestand, 1997). It is at this stage that the porosity of the tablet decreases and the pressure decreases too. Indeed, during the dwell time, punches do not hold their position but their penetration inside the die increases by a small amuont. As an example of the occurring phenomenon, the penetration of the lower punch with (Avicel PH 102, 300 MPa pressure) and without powder in the die is shown in Fig. 5. The vertical right segments 'A' and 'B' define, respectively, the beginning and the end of the dwell time. A bar is positioned on the first point of the plateau region of the empty die curve; 'B' bar is positioned on the point of maximum penetration of the full die curve (one instant before recovery occurs). If the effect of punch tilting on the transducers is present, it should be minimal and not influence this difference since this eventual tilting is repeated for both full and empty die and the compression force is
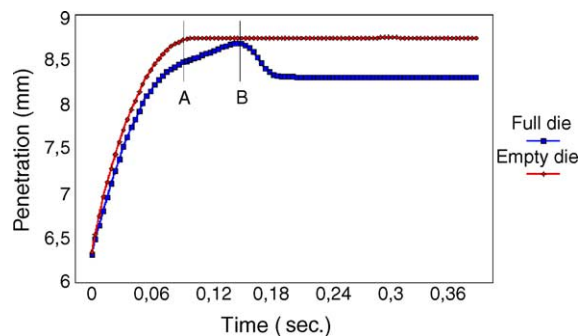
rather low. In any case, since transducer is positioned after the punch, the tilting effect should eventually reduce and not increase the difference between full and empty die.

When the die is empty, there is no further penetration after the segment A since punch head has reached the higher point of the lower roller. When powder is present inside the die, as expected, the plot is different. Powder resistance to penetration causes punch and machine deflection depending on the pressure applied. At point A, the curve is considerably lower but during the dwell time the punch does not remain in the same position; it continues to penetrate but without reaching the same penetration recorded for the empty die. Maybe, at the end of the dwell time, a part of the energy is still stored by the machine (Altaf and Hoag, 1994). The difference in penetration between a full and empty die is less accentuated for the upper punch (not shown curve). However, this depends on the way the machine is built. In other machines, it could be exactly the opposite (Altaf and Hoag, 1994; Walter and Augsburger, 1986; Oates and Mitchell, 1989, 1990). It may depend on what roller the vertically movable one is (to adjust pressure). In our machine, the upper roller is movable. During the dwell time, the energy stored by the deformed punch and machine is gradually given back following the viscous flow of the material in the die. After point B, all the flat portion of the punch head has overcome the maximum of the roller and the immediate elastic component of the compressed material relaxes and tablet height increases pushing punches to remain in contact with the respective roller for a short period of time. Punch penetration then remains at a steady state. Punch withdrawal occurs only when punch head is transported by its sloping guide as a consequence of the rotary movement of the turret.

Also, in this phase the remaining energy still stored by the machine relaxes and the upper and lower rollers move a little bit downwards and upwards, respectively.

This phenomenon, together with tablet expansion, maintains punches in contact with the rollers for a longer time than that observed in an eccentric machine. In this last one, 42 ms elapse between maximum punch penetration and pressure disappearance. In the rotary machine, it takes place after 64 ms. This may explain why in the rotary machine there is only a very small difference of relative density between the endpoint of the Heckel plot and the ejected tablet (Table 1).

Fig. 5. Comparison of lower punch penetration with and without material in the die.

Table 1
Heckel parameters and corresponding standard deviations

	MPa	Avicel PH 102		Pharmatose 50		Emcompress	
		Rotary	Eccentric	Rotary	Eccentric	Rotary	Eccentric
D_0'	100	0.324 ± 0.003	0.329 ± 0.002	0.571 ± 0.005	0.556 ± 0.003	0.490 ± 0.005	0.474 ± 0.001
	150	0.320 ± 0.001	0.330 ± 0.001	0.578 ± 0.006	0.555 ± 0.002	0.490 ± 0.004	0.476 ± 0.002
	200	0.320 ± 0.004	0.330 ± 0.003	0.574 ± 0.006	0.557 ± 0.002	0.488 ± 0.006	0.476 ± 0.002
D_A	100	0.513 ± 0.007	0.436 ± 0.002	0.740 ± 0.002	0.695 ± 0.001	0.709 ± 0.002	0.660 ± 0.002
	150	0.508 ± 0.010	0.452 ± 0.001	0.750 ± 0.003	0.693 ± 0.001	0.722 ± 0.001	0.655 ± 0.001
	200	0.527 ± 0.004	0.456 ± 0.002	0.740 ± 0.004	0.696 ± 0.001	0.724 ± 0.004	0.655 ± 0.001
D_B'	100	0.189 ± 0.009	0.107 ± 0.003	0.169 ± 0.008	0.139 ± 0.003	0.219 ± 0.004	0.186 ± 0.002
	150	0.188 ± 0.010	0.122 ± 0.001	0.172 ± 0.006	0.138 ± 0.002	0.232 ± 0.004	0.179 ± 0.002
	200	0.207 ± 0.009	0.125 ± 0.004	0.166 ± 0.008	0.139 ± 0.002	0.236 ± 0.007	0.179 ± 0.003
D_{MAX}	100	0.852 ± 0.002	0.849 ± 0.003	0.871 ± 0.002	0.865 ± 0.002	0.782 ± 0.003	0.788 ± 0.001
	150	0.926 ± 0.003	0.912 ± 0.001	0.911 ± 0.002	0.901 ± 0.001	0.824 ± 0.004	0.823 ± 0.002
	200	0.971 ± 0.004	0.949 ± 0.002	0.940 ± 0.004	0.926 ± 0.001	0.853 ± 0.002	0.846 ± 0.001
P_Y	100	87.30 ± 1.200	77.70 ± 0.400	238.0 ± 2.300	127.9 ± 1.100	404.0 ± 4.500	207.0 ± 1.700
	150	86.10 ± 1.100	83.00 ± 0.400	244.0 ± 2.100	130.2 ± 0.400	393.0 ± 4.100	195.0 ± 1.400
	200	84.60 ± 0.700	83.80 ± 0.300	245.0 ± 1.800	133.3 ± 0.600	395.0 ± 5.200	200.0 ± 1.400
D_{FIN}	100	0.808 ± 0.003	0.828 ± 0.002	0.850 ± 0.003	0.854 ± 0.002	0.775 ± 0.003	0.786 ± 0.001
	150	0.870 ± 0.002	0.886 ± 0.001	0.877 ± 0.002	0.882 ± 0.001	0.813 ± 0.002	0.813 ± 0.002
	200	0.913 ± 0.002	0.916 ± 0.002	0.904 ± 0.003	0.905 ± 0.001	0.837 ± 0.002	0.835 ± 0.001
D_{EJC}	100	0.807 ± 0.002	0.803 ± 0.002	0.848 ± 0.002	0.843 ± 0.002	0.776 ± 0.002	0.783 ± 0.002
	150	0.868 ± 0.002	0.866 ± 0.002	0.878 ± 0.001	0.875 ± 0.001	0.812 ± 0.003	0.811 ± 0.001
	200	0.912 ± 0.002	0.899 ± 0.002	0.900 ± 0.003	0.898 ± 0.001	0.836 ± 0.001	0.833 ± 0.002

On the other hand, the fast density increase occurring after 190 MPa can be explained with the drastic reduction of the penetration speed of the punches as they approach the maximum of the rollers. When machine deflection is absent (empty die) or not considered, punch penetration speed (Fig. 6) depends on the following equation:

$$V_V = V \cos\alpha \sin\alpha$$

where V_V is the vertical punch penetration velocity, V the rotary velocity of the machine and α is the angle formed by the vertical centre line of the roller and the line passing through the central point of the roller and the point where punch head is in contact with the roller. $V\cos\alpha$ is the transversal velocity V_T. Therefore, as α becomes small punch penetration becomes very slow and in practice depends on the energy stored by the machine rather than by the roller. It means there is a sort of pre-dwell time: no real penetration even if the flat portion of punch head has not reached the minimum (or maximum) of the roller yet. The little penetration at this stage is compensated by the machine deflection. This can explain why the Heckel plot changes the slope after 100 MPa and generally is less linear; part of the energy is stored by the machine and a lower than expected punch penetration occurs. The equation described above contains indeed a very little approximation because it does not take into account the punch head curvature. Anyway, a correction is impossible since the arc which is travelled by the roller on the punch head before reaching the flat portion of the same punch head is variable. In any case, this arc is short and it is for this reason that the approximation is little.

The previous statements are confirmed by Fig. 7, where pressure, $-\ln$ porosity and lower punch penetration for a compression cycle of Avicel PH 102 in the rotary machine are reported against time. Penetration values are subtracted by 5.5 mm to make the figure more comprehensive. The three parameters increase at the same time, but the increase rate of tablet density reduces by a small amount for a very short period of time (between 0.12 and 0.13 s). The same trend can
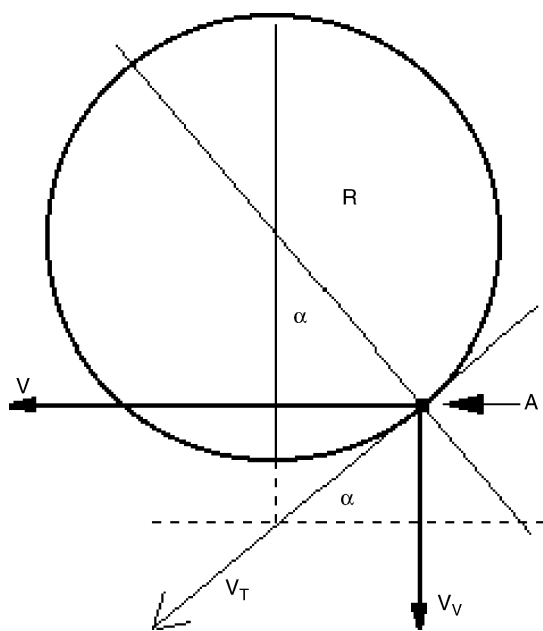
Fig. 6. Representation of the vertical punch penetration in a rotary machine. A is the contact point between roller and punch head.

be remarked at the same time in the punch penetration curve. This is the pre-dwell time where punch penetration is insignificant. Then, the force pulled by the deflected machine overcomes the material resistance. Dwell time begins, pressure reaches the maximum and punches newly penetrate the die a little. As previously noted, it is clearly visible that maximum density is reached a little while before the drastic pressure reduction occurs at the end of the dwell time. After that the elastic recovery takes place.
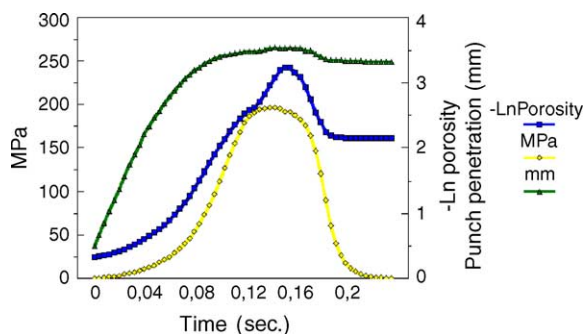

Fig. 7. Pressure, −ln porosity and lower punch penetration relationship for a compression cycle of Avicel PH 102 in the rotary tablet machine.
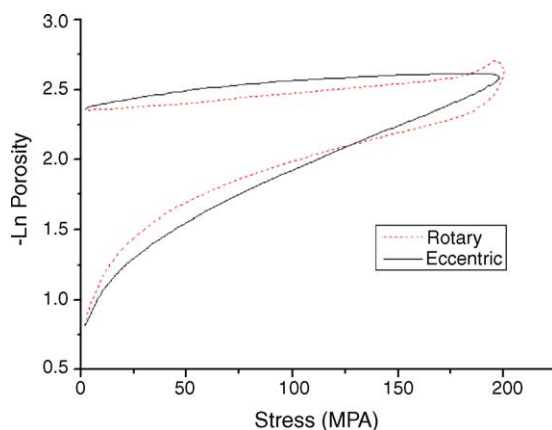

Fig. 8. Heckel plots of Pharmatose 50 in eccentric and rotary machines.

Figs. 8 and 9 show the Heckel plots of lactose and dicalcium phosphate, respectively, obtained at 200 MPa in both the eccentric and rotary machines.

Once again, plots from the rotary machine are less linear and a greater densification by fragmentation and particle rearrangement seems to occur. Indeed, this greater curvature of the initial portion of the plot may depend on the fact that material is pushed from both the upper and lower surfaces and fragmentation, rearrangement and other densification phenomena may take place a little in advance with respect to the eccentric machine. At the same time, the curvature may be increased by punch tilting that is present at the very first stage of compression force detection (Matz et al.,
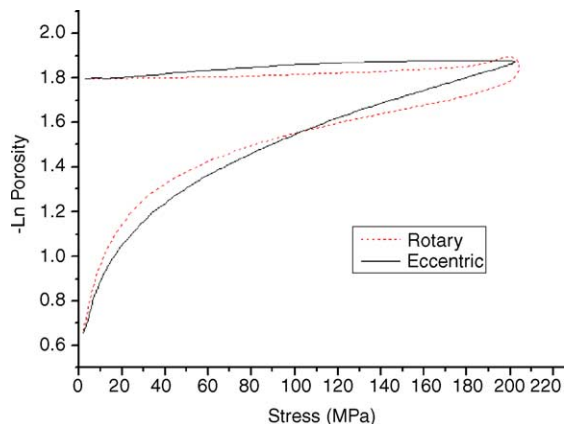

Fig. 9. Heckel plots of Emcompress in eccentric and rotary machines.

1999). Since the transducer succeeds the punch a little higher than real penetration may be measured in this phase. Then, after the right portion of the curve, the temporary increase of density occurring during the dwell time is less relevant than that obtained with the cellulose, particularly for the Emcompress. In practice, dwell time causes a further increase of deformation (viscous flow) as higher as the material is ductile. The final points of the return curves are, also in these cases, similar to those of the corresponding curves obtained by the eccentric machine. Finally, the deformation ability differences from one material to another, as detected by the eccentric tablet machine, are enlarged if the Heckel analysis is performed on a rotary machine. This is probably due to the different compression mechanism occurring in the rotary machine: both punches penetrate the die, machine deflection is remarkable, there is a considerable dwell time and punch tilting is present.

These statements are confirmed by Table 1 where values of D'_0, D_A, D'_B, maximum relative density reached (D_{MAX}), P_Y, relative density at the end of the compression cycle (D_{FIN}) and relative density of the ejected tablet (D_{EJC}) are presented for the three materials in both machines.

As expected, for each material, D'_0 values are very similar in both machines but D_A values are a little higher in the rotary machine, and consequently, D'_B values are higher too. This means fragmentation phenomena seem more relevant, although the previously described considerations must be done.

Another very important difference between eccentric and rotary machine is represented by the P_Y values. Avicel PH 102 which is a deforming material, shows similar values in both machines but lactose (a less deforming material) shows a considerable difference and the difference is even more remarkable for Emcompress, which is the least ductile material.

So, unless well deforming materials are used, Heckel parameters recovered using a rotary machine may better describe the behaviour of a certain material when a fast increasing stress is applied, such as during tablets production on an industrial scale, provided that punch tilting effect is minimised or monitored.

Yet, the maximum value of relative density is quite similar between the two machines for all the three materials, when a maximum pressure of 100 MPa is applied. On the contrary, D_{MAX} becomes a little higher
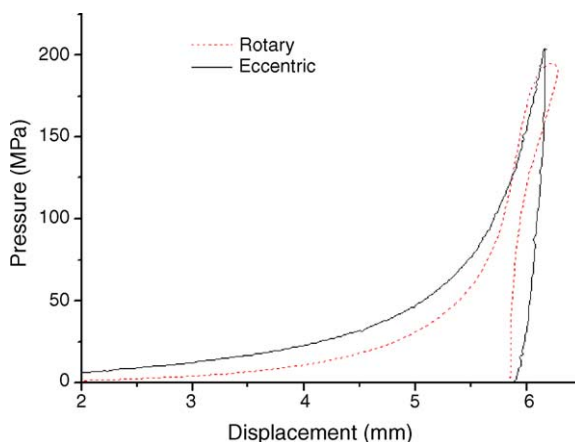

Fig. 10. Pressure/displacement plots of Avicel PH 102 in eccentric and rotary machines.

in the rotary machine at the maximum pressure used of 150 MPa and this difference is even more remarkable at 200 MPa, particularly when a more ductile material is used (Avicel PH 102). Of course, the effect of the dwell time gets gradually more important as the maximum pressure increases.

Fig. 10 shows the pressure/displacement curves of Avicel PH 102 obtained at 200 MPa as an example of the different trends between eccentric and rotary machines. The displacement in the curve from the rotary machine is given by the sum of both lower and upper punch displacement in order to allow a direct comparison, since in a rotary machine the penetration inside the die is divided between upper and lower punches. The zero point in the X axis (not shown) corresponds to the penetration level at the first detected pressure. In the rotary machine, pressure remains at a low level for a longer time and pressure increase rate is higher in the eccentric machine where maximum pressure corresponds to maximum punch penetration, giving a sharp peak. At a certain punch penetration level, this trend is inverted and pressure increases faster in the rotary machine. As a result, the plot obtained in the rotary machine is narrower and maximum pressure does not correspond to maximum punch penetration. As explained previously, maximum pressure occurs almost at the beginning of the dwell time and maximum punch penetration occurs at the end of the dwell time. With relation to the decompression curve, the decrease of punch penetration has a different trend

between rotary and eccentric machine. In this last one, there is a fast pressure reduction as the upper punch goes up after having reached the maximum penetration. In the rotary machine, pressure remains at considerable values despite the immediate punch penetration reduction that occurs after the maximum. As already explained, this difference may in part depend on punch tilting occurring in the rotary machine. Besides, what is more important, contact between tablet and punches (and rollers for the rotary press) is maintained for a longer time: due to punch penetration reduction, contact is lost at 0.29 mm after the maximum penetration in the eccentric machine and 0.43 mm after the end of the dwell time in the rotary machine. This longer contact between tablet, punches and rolls, occurring in the rotary machine may depend, as already said, on the machine relaxation causing a consequent small displacement of the upper and lower rollers (previously deflected) downwards and upwards, respectively.

As a consequence of all these phenomena, the energy truly involved in the formation of the tablet (area between compression and decompression curves) is lower in the rotary machine.

Concerning the example reported in Fig. 10, the calculated energies are 15.27 ± 0.14 J in the eccentric machine and 9.85 ± 0.19 J in the rotary machine.

4. Conclusion

Stress/strain data can be easily recovered using a rotary machine equipped with a fixed device. Heckel plots, derived from these stress/strain data and compared with those obtained in an eccentric machine, pointed out some differences. Curves obtained in the rotary machine possess a narrower zone of linearity for the calculation of P_Y and D_A since it is lost when penetration speed of both punches is very low because the dwell time is approaching. At the same time, the initial non-linear portion of the curve is a fraction more extended. The effect of the different compression mechanism of the rotary machine on the shape of the Heckel plot is more remarkable in a non-deforming material. The effect of the longer dwell time of the rotary machine on the porosity reduction occurring after the maximum pressure has been reached, is more remarkable in a ductile material. As a consequence of that Heckel parameters obtained in the

rotary press are also somewhat different from those recovered in the eccentric machine, according to the type of material. Starting from the more deforming (cellulose) and ending with the less ductile (dicalcium phosphate), the same rank of material characteristics are encountered between eccentric and rotary machine but in this last one, differences between materials are more marked. All these differences between the two machines depend on the long dwell time, the considerable machine deflection and punch tilting occurring in the rotary machine.

Finally, what could be more of interest, parameters recovered from the rotary machine may better describe the process of material densification into the die of a production scale rotary machine.

Concerning the rotary machine, the effect of machine speed and punch diameter on the Heckel plots and parameters will be discussed in another paper.

References

Altaf, S.A., Hoag, S.W., 1994. Deformation of the Stokes B2 rotary tablet press: quantitation and influence on tablet compaction. J. Pharm. Sci. 84, 337–343.

Celik, M., Aulton, M.E., 1996. The viscoelastic deformation of some tableting materials as assessed by indentation rheology. Drug Dev. Ind. Pharm. 22, 67–75.

Celik, M., Marshall, K., 1989. Use of a compaction simulator system in tableting research. Drug Dev. Ind. Pharm. 15, 759–800.

Charlton, B., Newton, J.M., 1984. Theoretical estimation of punch velocities and displacements of single-punch and rotary tablet machines. J. Pharm. Pharmacol. 36, 645–651.

David, S.T., Augsburger, L.L., 1977. Plastic flow during compression of directly compressible fillers and its effect on tablet strength. J. Pharm. Sci. 66 (2), 155–159.

Doelker, E., 1994. Assessment of powder compaction. In: Chulia, D., Deleuil, M., Pourcelot, Y. (Eds.), Powder Technology and Pharmaceutical Process. Elsevier, Amsterdam, pp. 403–471.

Dwivedi, S.K., Oates, R.J., Mitchell, A.G., 1991. Peak offset times as an Indication of stress relaxation during tableting on a rotary tablet press. J. Pharm. Pharmacol. 43, 673–678.

Gabaude, C.M.D., Guillot, M., Gautier, J.-C., Saudemon, P., Chulia, D., 1999. Effects of true density, compacted mass, compression speed and punch deformation on the mean yield pressure. J. Pharm. Sci. 88, 725–730.

Heckel, R.W., 1961a. Density–pressure relationships in powder compaction. Trans. Metall. Soc. A.I.M.E. 221, 671–675.

Heckel, R.W., 1961b. An analysis of powder compaction phenomena. Trans. Metall. Soc. A.I.M.E. 221, 1001–1008.

Hiestand, E.N., 1997. Mechanical properties of compacts and particles that control tableting success. J. Pharm. Sci. 86, 985–990.

Juslin, M.J., Paronen, T.P., 1980. On the accuracy of displacement measurements by instrumented single punch machine. J. Pharm. Pharmacol. 32, 796–798.

Matz, C., Bauer-Brandl, A., Rigassi, T., Schubert, R., Becker, D., 1999. On the accuracy of a new displacement instrumentation for rotary tablet presses. Drug Dev. Ind. Pharm. 25, 117–130.

Muller, F.X., Augsburger, L.L., 1994. The role of the displacement-time waveform in the determination of Heckel behaviour under dynamic conditions in a compaction simulator and a fully instrumented rotary tablet machine. J. Pharm. Pharmacol. 46, 468–475.

Nelson, E., Busse, L.W., Higuchi, T., 1955. The physics of tablet compression. VII. Determination of energy expenditure in the tablet compression process. J. Am. Pharm. Assoc. (Sci. Ed.) 44, 223–225.

Oates, R.J., Mitchell, A.G., 1989. Calculation of punch displacement and work of powder compaction on a rotary tablet press. J. Pharm. Pharmacol. 41, 517–523.

Oates, R.J., Mitchell, A.G., 1990. Comparison of calculated and experimentally determined punch displacement on a rotary tablet press using both Manesty and IPT Punches. J. Pharm. Pharmacol. 42, 388–396.

Radebaugh, G.W., Babu, S.R., Bondi, J.N., 1989. Characterization of the viscoelastic properties of compacted pharmaceutical powders by a novel non destructive technique. Int. J. Pharm. 57, 95–105.

Rees, J.E., Hersey, J.A., Cole, E.T., 1972. Simulation device for preliminary tablet compression studies. J. Pharm. Sci. 61, 1313–1315.

Ridgway Watt, P., 1988. Tablet Machine Instrumentation in Pharmaceutics. Ellis Horwood, Chichester, UK, pp. 305–310.

Rippie, E.G., Danielson, D.W., 1981. Viscoelastic stress/strain behaviour of pharmaceutical tablets: analysis during unloading and postcompression periods. J. Pharm. Sci. 70, 476–482.

Tsardaka, K.D., Rees, J.E., 1989. Plastic deformation and retarded elastic deformation of particulate solids using creep experiments. J. Pharm. Pharmacol. 41, 28–31.

Walter, J.T., Augsburger, L.L., 1986. Computerized force/displacement instrumentation system for a rotary press. Pharm. Technol. 10, 26–34.