International Journal of Pharmaceutics, 92 (1993) 157–166

IJP 03129

Temperature changes during tabletting measured using infrared thermoviewer

Jarkko Ketolainen, Jukka Ilkka and Petteri Paronen

Department of Pharmaceutical Technology, University of Kuopio, POB 1627, SF-70211 Kuopio (Finland)

(Received 4 September 1992)
(Accepted 18 November 1992)

Key words: Tabletting; Powder compression; Friction; Temperature rise; Thermal energy; Infrared thermoviewer; Thermography

Summary

To observe the rise in temperature during tabletting an infrared thermoviewer was used together with an instrumented eccentric tablet press. To evaluate the tabletting process, temperatures measured from surfaces of recently ejected tablets were used together with energy parameters. Two direct compression excipients, plastically deforming microcrystalline cellulose and fragmenting dicalcium phosphate dihydrate, were tabletted. Due to differences in specific heat values, the temperature rise of the tablets was higher with microcrystalline cellulose than with dicalcium phosphate. Also more non-homogeneous particle shape and plastic deformation instead of fragmentation may have led to higher temperatures of microcrystalline cellulose tablets. For both test materials the temperature of the tablets rose with the compressional force whereas lubrication diminished the rise in temperature. Due to the non-homogeneous densification the highest temperature values were obtained at the centre of the upper surfaces of the tablets. After a short initial stabilization phase, the rise in tablet temperature became greatly dependent on the temperature increase of the powder in the hopper. From the energy parameter values, derived either from force and displacement data (mechanical energy), or from specific heat, temperature increase and tablet weight values (thermal energy), it was noted that the mechanical energy was very extensively converted to thermal energy. Thus, a permanent increase in energy content of powders by compression seemed to be small. The infrared thermoviewer was found to be an accurate and informative method for evaluating changes in the temperature and energy content of compressed powders during a dynamic tabletting process.

Introduction

The temperature of tablets immediately after ejection from the die is higher than the ambient room temperature or the temperature of the powder just before compression. This rise in temperature is dependent on the formulation of the tablet as well as on the procedure of the whole tablet manufacturing process (Wurster and Creekmore, 1986). Increase in temperature may induce changes in tablet structure (York and Pilpel, 1972). The temperature rise is caused by several exothermic processes which take place during the compression and ejection phases (Hölzer and Sjögren, 1981; Coffin-Beach and

Correspondence to: J. Ketolainen, Department of Pharmaceutical Technology, University of Kuopio, POB 1627, SF-70211 Kuopio, Finland.

Hollenbeck, 1983). Exothermic temperature changes are caused mainly by die wall and interparticulate friction, but elastic and plastic deformation as well as bonding can also release heat during the tabletting process. Due to non-homogeneous densification of the powder in the die the temperature changes could be different in different parts of the powder column.

Several studies have dealt with measurements of tablet temperatures, but only a few of these present temperatures measured from different parts of tablets. In the first reliable studies of tablet temperature, thermocouples and thermochromic indicators were used (Hanus and King, 1968; Juslin, 1969; Travers and Merriman, 1970). In recent studies the thermal energy of tablet compression has been evaluated with different kinds of sophisticated calorimetric methods and embedded temperature probes (Coffin-Beach and Hollenbeck, 1983; Wurster and Creekmore, 1986). The main disadvantage of these methods is that neither compact strength nor any other physicomechanical property can be measured. Bardon et al. (1985a,b) and Diaz Esquivel et al. (1991a,b) have evaluated tablet temperatures by utilizing a microcalorimetric method in which compacts remain undamaged and can be used for further studies. The main disadvantage of this method is that only the overall increase in tablet temperature can be measured.

Detection of infrared radiation is not widely used in evaluation of tabletting processes, even though it is much utilized in other fields of science. Nurnberg and Hopp (1981) employed a portable device for infrared detection to measure the surface temperatures of tablets as they were ejected from the dies of rotary and eccentric tablet presses. They pointed out that an increase in the speed of compression has a greater effect on tablet temperatures than an increase in compressional force has. More recently Bechard and Down (1992) utilized a high-resolution infrared camera for investigating the heat release during compaction and they pointed out that the heat release increased with compaction force. They also pointed out that the compression rate did not have any significant effect on heat release which is in contradiction with the study of Nurnberg and Hopp (1981). Different results may be due to differences in behaviour of compressed powders as well as in tabletting processes.

Infrared detection seemed to be a useful method in evaluation of tabletting, because it does not have any physical contact with the target to be measured and therefore can in no way influence the proceeding of the tabletting process (Nurnberg and Hopp, 1981; Bechard and Down, 1992). Infrared detector measures the levels of infrared radiation emitted by an object. Optical filters eliminate interference from solar radiation or incidental sources. With a modern infrared thermoviewer, in which a two-dimensional scan builds up a picture of the field of view, the temperature variations are seen as changes of brightness or colour (Ridgway Watt, 1988). With this method, it is possible to obtain simultaneously a real-time temperature distribution from the whole tablet surface on a multi-colour scale and accurate temperature values from a certain point on the tablet surface even during the tabletting process. Furthermore, compacts remain undamaged and can be used for further studies.

The aim of this study was to evaluate the applicability of an infrared thermoviewer, in combination with an instrumented eccentric tablet machine, for investigating the conversion of mechanical energy to thermal energy during the tabletting process.

Materials and Methods

The compressed excipients used were microcrystalline cellulose (Avicel PH 102, FMC Corp., U.S.A.) and dicalcium phosphate dihydrate (Emcompress, E. Mendell Corp., U.S.A.). The specific heat (C_s) of the compressed materials was determined using a differential scanning calorimeter (Perkin-Elmer DSC 7, Perkin-Elmer Corp., U.S.A.). The C_s values for microcrystalline cellulose and dicalcium phosphate dihydrate were 5.0 and 2.5 J/g per °C, respectively. The material densities of the compressed powders were determined with a pycnometric method (Multipycnometer MVP-1, Quanta Chrome, U.S.A.) using helium as an inert gas. The material density values

for Avicel and Emcompress were 1.602 and 2.385 g/cm^3, respectively.

Both studied excipients were compressed with and without lubricant using compressional forces of 5 and 20 kN. The relative standard deviation of compressional force was in every five-tablet series less than 1.6% for both pressure levels. The mean values of forces varied between test runs less than 0.6 kN. Magnesium stearate 0.5% w/w, when used as a lubricant, was mixed with the excipients for 5 min (20 rpm) in Turbula T2C-mixer (WA Bachofen, Switzerland). The filling level for mixing was two thirds of the volume of a 2 l glass vessel. Tablets were compressed with an instrumented Korsch EK-0 DMS (Korsch Maschinenfabrik, Germany) eccentric tablet press using flat-faced punches, 10 mm in diameter. The lower punch was adjusted so that the die was filled with an appropriate amount of powder to produce tablets having a height of 2.00 mm at theoretical zero porosity. The mean tablet weights were for Avicel and Emcompress tablets 251 and 376 mg, respectively. The relative standard deviation of the tablet weights was less than 0.5% in every case. The running speed of the tablet press was 25 rpm. Tablets were made at normal room temperature (20–21°C) and low relative humidity (9–10%). The measured force and displacement values from five tablets were recorded 1, 5, 25, 45, and 65 min after the tabletting started.

The tabletting data was acquired by a PC computer with a suitable software (PuuMan Corp., Finland). The compressional work and ejection work were calculated from the upper and lower punch forces and displacements measured during the compression and ejection phases of tabletting. The apparent net input energy was calculated by subtracting the friction and expansion work values from the compressional work values. For every five-tablet series the relative standard deviation of the mean mechanical energy values was less than 3.5%. Only for the mean values of expansion work the relative standard deviation was higher, in cases with very small expansion of tablet even 50%.

The infrared thermoviewer Inframetrics 600 IR Imagine Radiometer (Inframetrics Corp., U.S.A.) consists of an infrared scanner, a control unit, a colour monitor, and a video cassette recorder for later playback and analysis. The infrared scanner was placed just in front of the die plate, at a distance of 15 cm from the punches. To obtain an inert background, the metal parts of the hopper and the die plate, which were in the field of view of the scanner, were covered with polystyrene plates. Temperature gradients were scanned from the side, upper, and lower surfaces of the tablets and the local maximum temperatures on the surfaces were measured with an accuracy of 0.1°C, 1–2 s after the ejection from the die. The temperature of the powder in the hopper during the tabletting was measured with an electrical contact thermometer. Temperatures were evaluated 1, 5, 25, 45, and 65 min after tabletting began. At every moment of measurement for both materials and forces five measurements were made from the centre point of all three tablet surfaces. The relative standard deviation of the mean values of maximum temperatures was in every case less than 0.9%. The tablet press was allowed to cool for 4 h after each test run.

The rise in temperature (T) was calculated using the following equation

$$T = T_1 - T_2 \tag{1}$$

where T_1 is the temperature of the tablet surface immediately after ejection or the temperature of the powder in the hopper at the same moment and T_2 denotes the temperature of the powder inside the hopper just before the tabletting began. Temperature difference, T, was registered as a function of the run time of tabletting process.

The thermal energy (ΔQ_T) emitted as infrared radiation from recently compressed tablets was calculated using the equation (Hanus and King, 1968; Nurnberg and Hopp, 1981)

$$\Delta Q_T = T_d \cdot C_s \cdot m \tag{2}$$

where T_d is the difference between the temperature of the side surface of the ejected tablet and the temperature of the tablet mass in the hopper at the same moment after starting the tabletting. C_s is the specific heat of the compressed material

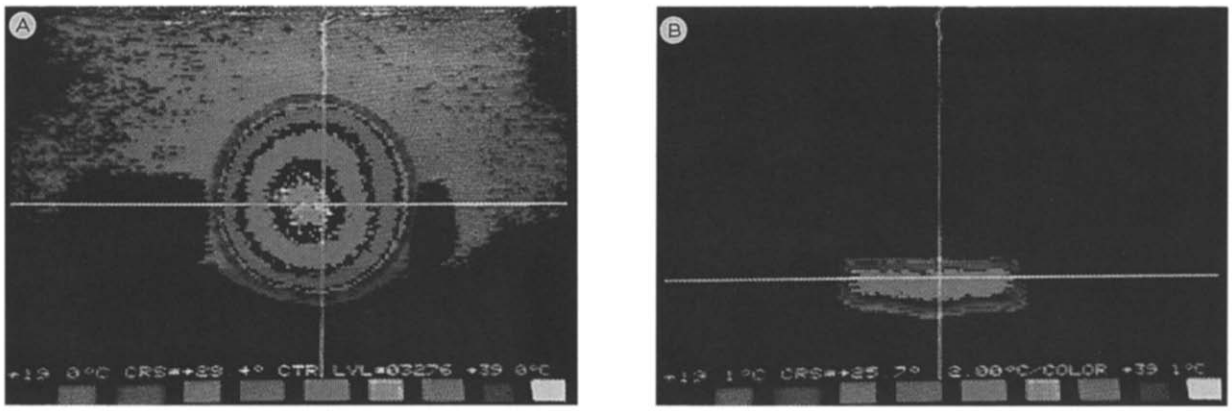

Fig. 1. Thermograms of upper (A) and side (B) surfaces of lubricated Avicel PH 102 tablets compressed at 20 kN. Temperature scale is from 19 to 39°C.

and m is the weight of the tablet. Consequently, to achieve an increase of 1°C in the temperature of Avicel or Emcompress tablets compressed in this study the energy needed is approx. 1.25 or 0.94 J, respectively.

The breaking strength of the tablets was determined 24 h after tabletting as a mean of 10 tablets with a Schleuniger 2E-apparatus (Dr Schleuniger, Switzerland).

Results and Discussion

Temperature rise on tablet surfaces

With both Avicel and Emcompress powders, the temperature of the ejected tablets was clearly higher than the ambient room temperature. Typical thermograms of the upper and the side surfaces of the tablets are presented in Fig. 1A and B. With all tablets the highest temperature values

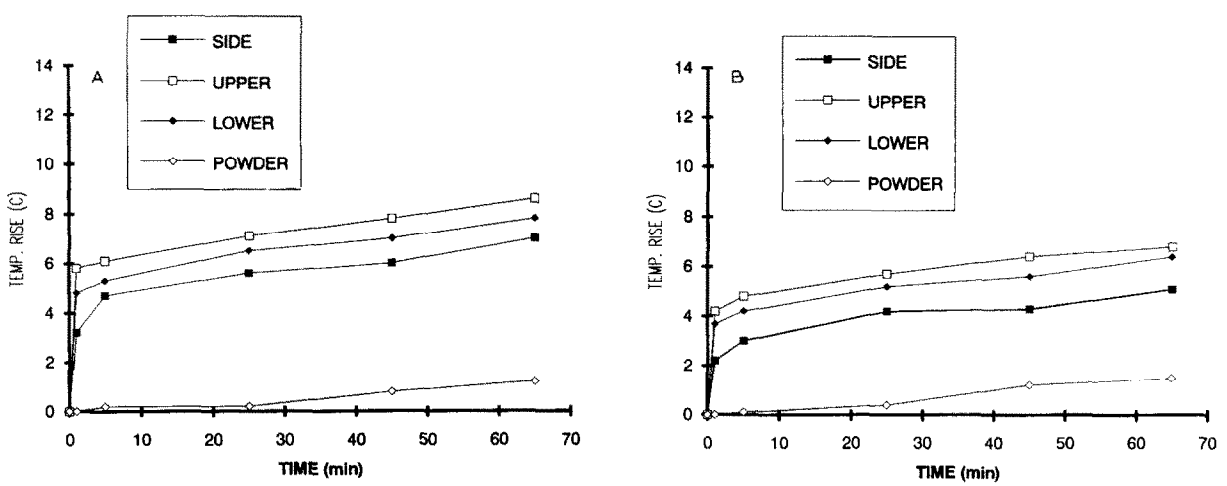

Fig. 2. Temperature rise (°C) of lubricant-free (A) and lubricated (B) Avicel PH 102 tablets compressed at 5 kN and powder in the hopper as a function of the run time of tabletting (min).

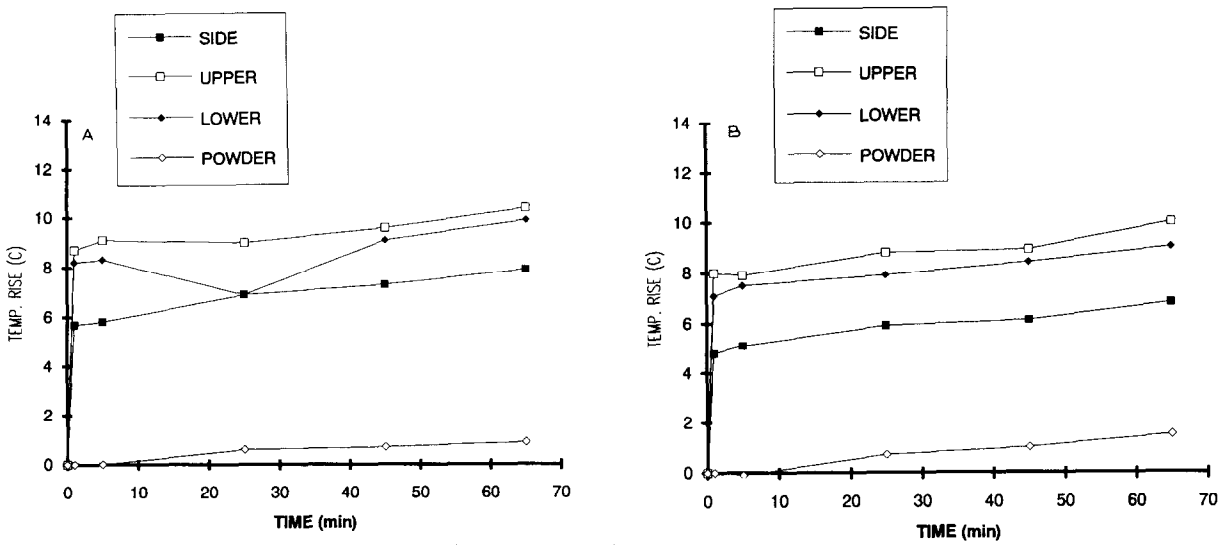

Fig. 3. Temperature rise (°C) of lubricant-free (A) and lubricated (B) Avicel PH 102 tablets compressed at 20 kN and powder in the hopper as a function of the run of tabletting (min).

were obtained from the upper surfaces of the tablets and the lowest temperature values from the side surfaces (Figs 2–4). The lowest temperatures on the side surfaces of the tablets may be due to the more effective conduction of heat from the die wall than from the moving punches. This was supported by the result that the temperature distribution on the side surfaces of tablets was clearly more homogeneous than those of the upper and lower surfaces (Fig. 1A and B).

The warmest spots were located on the centre parts of the lower, and especially of the upper tablet surfaces (Fig. 1A). This can be explained by differences in the particle movement and densification within the powder column during tabletting. The particles lying on the upper part of the powder column were forced to move more extensively and so high interparticulate friction as well as die wall friction were formed, especially in the upper part of the compact during eccentric compression. During compression the densest parts have been reported to form in the centre of the upper and lower parts of the powder column (Macleod and Marshall, 1977). The warming of tablets also seemed to be very intensive in these high density areas, which was reflected in the temperature distributions of the upper and lower tablet surfaces. Although the lower punch did not move during the compression phase, it moved upwards during ejection and often brought a lot of mechanical energy into the compact, thus warming up the tablet. Differences in temperatures between different parts of the tablets remained virtually unchanged during the whole 65 min tabletting process (Figs 2–4). This indicates that the whole tabletting process proceeded without extensive thermodynamic changes between different parts of the compacts.

Warming of tablets as a function of run time of tabletting

The increase in tablet temperatures, caused by the compression and ejection phases, reached a steady warming phase rather rapidly after the tabletting process began (Figs 2–4). For all the materials except lubricant-free Emcompress the initial warming period lasted less than 5 min. During this period the surface properties of the die wall and punches were apparently greatly influenced by the moving powder surfaces. The changes in the surface properties of the steel parts depended to a great extent on the properties of the tablet formulation. When lubricated excipients are tabletted, even an actual surface layer can be formed on the die wall and punches. This hydrodynamic lubrication layer seemed to

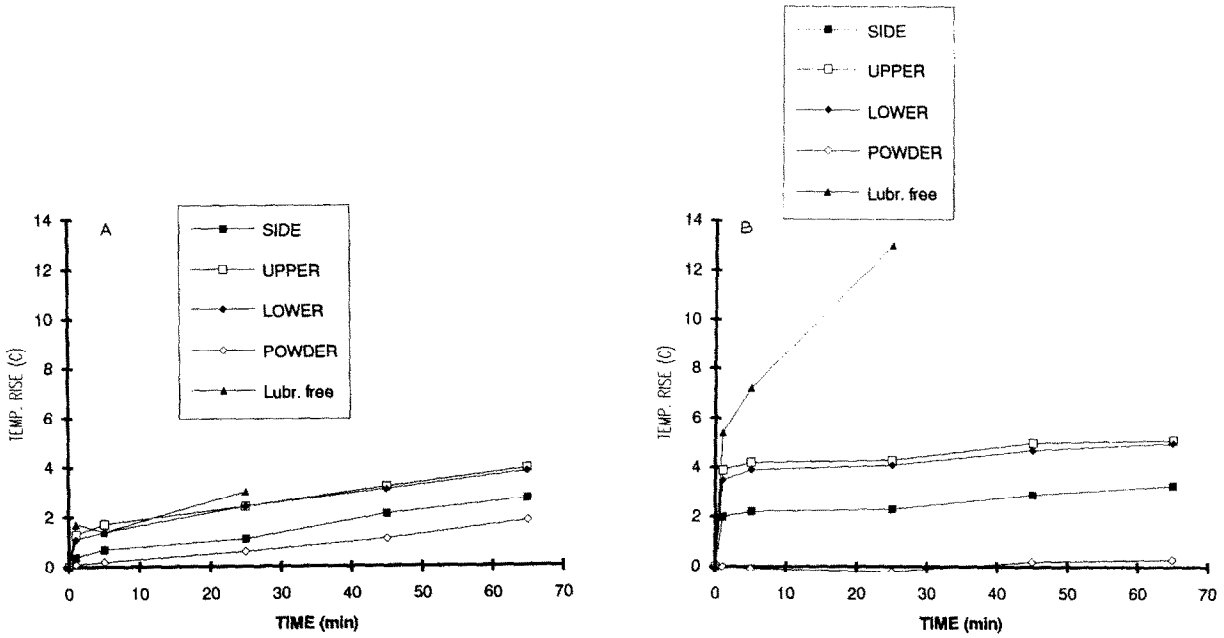

Fig. 4. Temperature rise (°C) of lubricant-free (side temp.) and lubricated Emcompress tablets compressed at 5 kN (A) and 20 kN (B) and powder in the hopper as a function of the run time of tabletting (min).

form quickly and thus the initial warming period lasted only about 1 min for both lubricated excipients. For lubricant-free Avicel, consisted of plastically deforming particles, the initial warming period lasted longer than for lubricated Avicel. Because the Avicel particles did not stick exten-

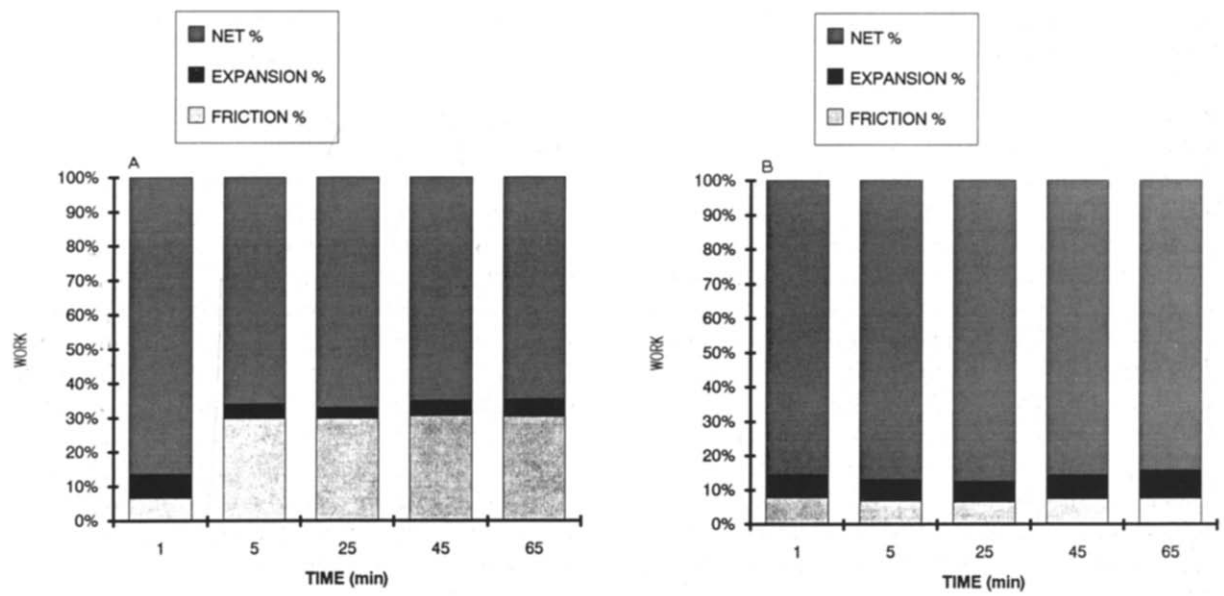

Fig. 5. Proportions (%) of friction work, expansion work and apparent net work from compressional work as a function of the run time of tabletting (min) for lubricant-free (A) and lubricated (B) Avicel PH 102 tablets compressed at 20 kN.

sively on the surfaces of the steel parts, the contamination phase was, however, relatively short.

With lubricated excipients as well as with lubricant-free Avicel, the surfaces of the die wall and punches seemed to remain virtually unchanged after this initial period of stabilization; and therefore the increase in tablet temperature was caused mainly by the temperature rise of the powder in the hopper (Figs 2–4). With fragmenting and effectively sticking lubricant-free Emcompress, the surfaces of the die wall and punches became more and more contaminated and temperature rose continuously until the tabletting was impossible due to breakage of compacts (Fig. 4A and B).

The energy parameters measured at different moments of the 65 min tabletting process showed similar trends to the temperature values. The compressional work, friction work, expansion work, as well as the values for ejection work varied considerably between formulations (Table 1), but for each formulation they remained virtually unchanged after the initial phase of stabilization. The effect of changes in surface properties of the die wall on the energy values was clearly seen with Avicel powders (Fig. 5A and B). Thus the proportion of friction work from the compressional work increased extensively after the early moments of the tabletting process for lubricant-free Avicel. For lubricated Avicel the stabilization of die wall occurred evidently so quickly that no marked difference as a function of run time of tabletting was noted.

In general, the lubrication significantly decreased the temperature rise with both powders (Figs 2–4). This tendency was seen clearly with Emcompress at the high pressure, even though, due to breakage, only the side temperatures of the lubricant-free Emcompress tablets could be measured (Fig. 4A and B). The diminished temperature rise is due mainly to the decrease in friction, which was clearly seen from the friction work and ejection work values of the both materials studied (Table 1). On the other hand, in addition to the decreased friction, prevention of breakage was important for fragmenting Emcompress. There were no marked differences in the diminution of the temperature rise between the different parts of the lubricant-free and lubricated tablets, which indicates that the lubricant affected the different parts of the tablets equally (Figs 2–4).

Comparison of energy parameters

The thermal energy values, calculated from the values for temperature difference, specific heat, and tablet weight, are presented in Table 1 with the work values derived from force and displacement measurements. By comparing these values it is possible to evaluate the conversion of mechanical energy to emitted thermal energy. The mechanical energy is brought into the powder column by the moving punches. At least theoretically the difference between produced mechanical energy and emitted thermal energy describes the permanent increase in the energy content of the powder caused by compression. The values in Table 1 must, however, be considered cautiously. For example, the temperature values used in our calculations were measured on the side surfaces of the tablets. According to our measurements, in most cases the rather homogeneous side surface temperatures represented the mean temperature of the whole compact better than the measured local maximum temperatures on the centre of the upper or lower surfaces. Although the actual heat content of the tablets is not known, the methods used are suitable for obtaining a general impression of the energy consumption during tabletting.

Compressional work (i.e., upper punch work) describes the total amount of mechanical energy needed to compress a loose powder column into a dense tablet. During compression a part of this energy is consumed to overcome the friction between the powder column and the die wall (friction work in Table 1). The second measurable part of energy consumption is caused by expansion of the tablet after maximum compression (expansion work in Table 1). The expansion work measured from the force-displacement data describes only partly the work done by the recovery of the tablet. Thus the widely used apparent net input parameter calculated by subtracting the friction work and expansion work from the com-

TABLE 1

Tabletting and thermal energy parameters of compressed materials 5 min. after the tabletting process began. All values are means of five tablets

Material	Compression force (kN)	Tablet weight (g)	Compression work (J)	Friction work (J)	Expansion work (J)	Net input (J)	Ejection work (J)	Temperature rise (°C)	Thermal energy (J)	$\frac{\text{Thermal energy}}{\text{compression work} + \text{ejection work}}$ (%)
Avicel PH 102	5	0.245	5.99	1.98	0.10	3.91	0.74	4.7	5.76	85.6
	20	0.250	11.90	3.55	0.50	7.84	1.15	5.8	7.25	55.6
Avicel PH 102 +	5	0.253	5.22	0.36	0.07	4.79	0.38	2.9	3.67	65.5
Mg. stearate 0.5%	20	0.255	10.66	0.74	0.65	9.26	0.16	5.1	6.50	60.1
Emcompress	5	0.376	2.91	1.27	0.15	1.49	3.12	1.2	1.13	18.7
	20	0.376	6.81	3.46	0.37	2.98	7.83	7.2	6.77	46.2
Emcompress +	5	0.374	2.36	0.16	0.06	2.16	0.14	0.5	0.47	18.8
Mg. stearate 0.5%	20	0.377	6.93	0.65	0.59	5.69	0.41	2.3	2.17	29.6

pressional work overestimates the possible energy available for bonding and formation of firm compacts. The main disadvantage of the above-mentioned parameters is that the effect of interparticulate friction cannot be determined. It seems reasonable to assume that a large proportion of compressional work is done especially to overcome friction between particles, thus warming up the tablet.

The above-mentioned assumption is supported by the thermal energy values (Table 1). When the proportion of thermal energy from different work values is examined, it is clear that a lot of mechanical energy is used to form heat. In some cases the thermal energy values were practically same as the compressional work values and higher than the apparent net input, owing to the ejection phase of tabletting, in which the work is done to remove the powder column from the die. This seems to be a very important heat-generating phase, especially for lubricant-free Emcompress powders.

In general, the thermal energy values are higher for Avicel powders than for Emcompress powders, which might besides the difference in specific heat values be due to the more irregular particle shape of microcystalline cellulose particles. Thus densification of the powder column by a rearrangement of particles might consume relatively large amounts of energy, especially to overcome the interparticulate friction, i.e., warming the powder compact. On the other hand, the brittle fracturing of dicalcium phosphate particles might have permanently consumed a greater part of the input energy than the plastic flow of cellulose particles did. Also, due to the stress relaxation, more energy is released as heat from Avicel tablets than from Emcompress tablets.

Breaking strengths of the tablets

The breaking strengths of the tablets changed only slightly during the tabletting process (Fig. 6). Also, the mechanical energy values did not vary significantly during the process. Thus the rise in temperature having a magnitude like in our case did not affect the mechanical strength of tablets or the compressional behaviour of the two compressed materials unlike in the studies of Diaz

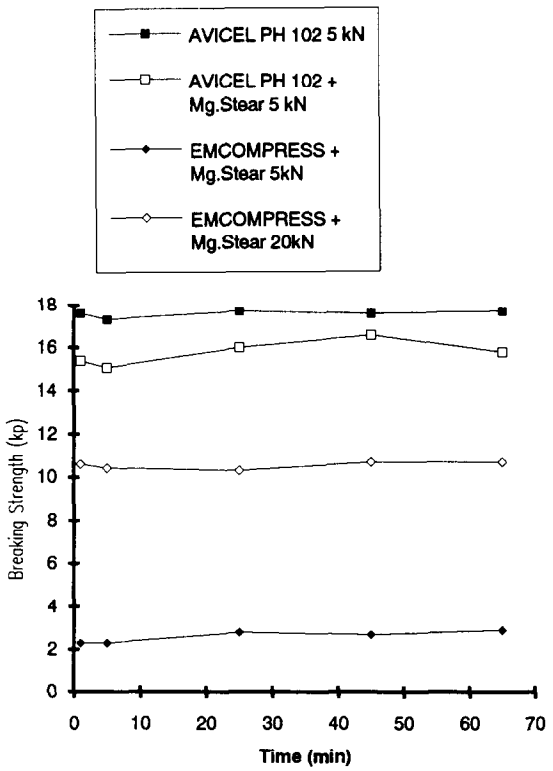

Fig. 6. Breaking strength of tablets (kp) as a function of the run time of tabletting (min). Breaking strength values of Avicel PH 102 tablets compressed at 20 kN (> 20.0 kp) and lubricant-free Emcompress tablets compressed at 5 kN and 20 kN (< 1.0 kp) could not be measured.

Esquivel et al. (1991a,b). Powders having higher compressional work values seemed to be more prone to form mechanically firm tablets. This kind of relationship did not exist between the ejection work and the breaking strength. Although the ejection phase is important for warming up the tablets and the ejection work values can be very high, this phase seemed to be less important for the formation of firm tablets. Only with very poorly compressable lubricant-free Emcompress, when the ejection phase has very high work values, there may be an extra fracturing effect on tablets.

Conclusions

In this study we evaluated the applicability of an infrared thermoviewer for investigating the tabletting process. The main advantages of this method are that infrared detection does not disturb the tabletting physically and that it is rapid to proceed. Furthermore, the temperature information can be observed and applied during detection. The results of this study indicated that, despite the fact that surface temperatures measured by an infrared detector may not be truly indicative of the interior temperatures of tablets, a modern infrared thermoviewer, when used together with other kinds of tablet machine instrumentations, is a useful tool for characterizing the conversion of mechanical energy to thermal energy during the tabletting process.

Acknowledgements

The authors wish to thank Dr E. Marttila, Lappeenranta University of Technology, Finland, for making it possible for us to use the detection device, and Ms M. Laakkonen for skilful technical assistance. Financial support from the Technology Development Centre of Finland (Tekes) is gratefully acknowledged.

References

Bardon, J., Sébert, P., Chaumat, C., Robelin, N. and Rollet, M., Elévation de température subie par les mélanges de poudres ou de granulés lors de leur transformation en comprimés. I: Influence de facteurs liés au matériel de compression. *STP Pharm.*, 1 (1985a) 706–710.

Bardon, J., Sébert, P., Chaumat, C., Robelin, N. and Rollet, M., Elévation de température subie par les mélanges de poudres ou de granulés lors de leur transformation en comprimés. II: Influence de la nature du taux de lubrifiant. *STP Pharm.*, 1 (1985b) 948–955.

Bechard, S.R. and Down, G.R.B., Infrared imaging of pharmaceutical materials undergoing compaction. *Pharm. Res.*, 9 (1992) 521–529.

Coffin-Beach, D.P. and Hollenbeck, R.G., Determination of the energy of tablet formation during compression of selected pharmaceutical powders. *Int. J. Pharm.*, 17 (1983) 313–324.

Diaz Esquivel, J.J., Bardon, J., Rollet, M. and Ozil, P., Phénomènes thermiques lors de la compression des poudres pharmaceutiques. I: Influence du taux d'un lubrifiant: béhénate de glycérol. *Pharm. Acta Helv.*, 66 (1991a) 109–119.

Diaz Esquivel, J.J., Bardon, J. and Rollet, M., Phénomènes thermiques lors de la compression des poudres pharmaceutiques. II: Influence du matériel de compression. *Pharm. Acta Helv.*, 66 (1991b) 141–150.

Hanus, E.J. and King, L.D., Thermodynamic effects in the compression of solids. *J. Pharm. Sci.*, 57 (1968) 677–684.

Hölzer, A.W. and Sjögren, J., Friction coefficients of tablet masses. *Int. J. Pharm.*, 7 (1981) 269–277.

Juslin, M.J., A study on the rise of temperature on the upper and lateral surfaces of tablets during compression. *Farm. Aikak.*, 78 (1969) 201–210.

Macleod, H.M. and Marshall, K., The determination of density distributions in ceramic compacts using autoradiography. *Powder Technol.*, 16 (1977) 107–122.

Nurnberg, E. and Hopp, A., Temperature measurement during tableting. *Pharm. Technol.*, 168 (1981) 81–101.

Ridgway Watt, P., *Tablet Machine Instrumentation in Pharmaceutics*, Ellis Horwood, Southampton, 1988, p. 191.

Travers, D.N. and Merriman, M.P.H., Temperature changes occurring during the compression and recompression of solids. *J. Pharm. Pharmacol.*, 22 (Suppl.) (1970) 11S-16S.

Wurster, D.E. and Creekmore, J.R., Measurement of the thermal energy evolved upon tablet compression. *Drug. Dev. Ind. Pharm.*, 12 (1986) 1511–1528.

York, P. and Pilpel, N., The effect of temperature on the frictional, cohesive and electrical conducting properties of powders. *Mater. Sci. Eng.*, 9 (1972) 281–291.