J. Pharm. Pharmacol. 1986, 38: 254–258
Received September 26, 1985

Measurement of hardness and strength of tablets and their relation to compaction performance of powders

W. E. JETZER*

Pharmazeutisches Institut der Universität Basel, CH-4051 Basel, Switzerland

Materials with different deformation properties were compressed into tablets at a range of pressures. The compacts were characterized in terms of indentation hardness which quantifies resistance to deformation and tensile strength quantifying resistance to fracture. A unifying description of these two competing mechanical responses is made using the evaluated single compaction parameters obtained with the equation proposed by Leuenberger. The compaction performance parameter and the bonding index make it possible to characterize the deformation and bonding behaviour of a substance under pressure. Furthermore, useful information is obtained for predicting when capping problems occur. It is evident that more than one index or parameter is necessary to describe the variety and complexity of the tableting properties.

A knowledge of the mechanical properties of compressed tablets—tensile strength and indentation hardness—provides useful information for their formulation. Both measurements vary with porosity of the powder compact, but as separate entities they quantify two competing mechanical responses—fracture and deformation.

Leuenberger et al (1981) proposed equation (1) to correlate the indentation hardness, P, as a function of the applied compression pressure, σ_c, needed to make the compact, and the relative density ρ_r:

$$P = P_m [1 - \exp(-\gamma_P \sigma_c \rho_r)] \quad (1)$$

P_m is the magnitude of P when $\rho_r = 1$ (porosity = zero) quantifying the compactibility of the powdered material. The parameter γ_P, termed compression susceptibility, may reflect the compressibility. Also it is possible to apply the equation to characterize the development of compact strength σ_T determined by tensile strength measurements:

$$\sigma_T = \sigma_{Tm} [1 - \exp(-\gamma_T \sigma_c \rho_r)] \quad (2)$$

This is obtained by substituting σ_T values for P values. In analogy, σ_{Tm} is the maximum tensile strength.

Little effort has been made to combine such parameters into a common description. The importance of this factor has been recognized by Hiestand & Smith (1984) who proposed the ratio σ_T/P as a Bonding Index (BI). Jetzer & Leuenberger (1984) report a discontinuity of the ratio σ_T/P when fracture problems occur. A reduction or avoidance of fractures could be obtained with the same lot of materials using a triaxial decompression method during the fabrication process (Jetzer et al 1985).

Equations (1) for hardness and (2) for tensile strength may be connected and rearranged to obtain the following combined expression (Jetzer et al 1985). For σ_T/P one may obtain:

$$\frac{\sigma_T}{P} = \frac{\sigma_{Tm}}{P_m} \left[1 - \frac{1 - \exp[(\gamma_P - \gamma_T)\sigma_c \rho_r}{1 - \exp(\gamma_P \sigma_c \rho_r)}\right] \quad (3)$$

The purpose of the present report is to investigate the relation between the two mechanical properties —tensile strength and indentation hardness—to discuss the possibilities of using the compaction parameters to interpret, or predict, the tableting behaviour of a material. In this study the magnitude of the compaction performance parameter ($\gamma_P - \gamma_T$) of pure substances will be compared.

MATERIALS AND METHODS

The materials used were of Pharmacopoeial grade: aspirin 150 (aspirin powder 150, Bayer, FRG), aspirin FC (aspirin fine crystals 150/350, Bayer, FRG), ASS cryst, ASS powder (acetylsalicylic acid, crystalline and powder, Siegfried, Switzerland), microcrystalline cellulose (Avicel PH 102, FMC, USA), caffeine powder (caffeine anhydr. powder, Knoll, FRG), caffeine FGR (caffeine anhydr. free-flowing fine granulate, Boehringer, FRG), Emcompress (dicalcium phosphate dihydrate, Ed. Mendell, USA), lactose (lactose anhydr., Humko Sheffield,

* Present address: Kantonsspital Bruderholz, Pharmaceutical Department, CH-4101 Bruderholz BL, Switzerland.

USA), mannitol (BV Chemie, The Netherlands), dipyrone (metamizol, natrium novaminsulfonicum, Siegfried, Switzerland), paracetamol DC (Graesser Salicylates, UK).

All materials were previously characterized in terms of size, shape, poured and tapped bulk density, true density and moisture content (Jetzer 1982). In particular, to ensure that the effects of these variables had been eliminated, indentation hardness and tensile strength measurements were performed on the same lot of tablets and tested under the same experimental conditions. The tablets were fabricated, stored and tested at room temperature (22 °C) at 45 ± 10% r.h. Before testing the mechanical properties, the compacts were stored for at least 48 h.

Tablets (400 mg, round, flat-faced, with a diameter of 11 mm) were compressed on a Universal Testing Instrument (Model TT-DM, Instron Ltd, High Wycombe, UK) at a range of compaction pressures between 50 and 310 MPa.

Indentation hardness was evaluated as described earlier (Jetzer et al 1983) using an indentation load of 19·61 N and an indenter sphere of 1·761 mm diameter. Tensile strength was also determined using the Universal Testing Instrument (Jetzer & Leuenberger 1984). Details of experimental procedures, evaluation and statistics (non-linear regression analysis program No. 09835-15040 Hewlett-Packard and NLIN Procedure by SAS Institute Inc., Cary NC) of the compression parameters were reported by Jetzer et al (1983). For each measurement, six tablets were tested.

RESULTS AND DISCUSSION

The results and estimated values of compactibility parameters P_m, σ_{Tm}, and compressibility γ_P, γ_T, of the investigated substances determined by equations 1 and 2, respectively, using non-linear regression analysis, are given in Table 1.

The effect of compaction pressure on the development of indentation hardness is shown in Fig. 1a, b. The typical relation between the indentation hardness of tablet and the product of compression stress and relative density, according to equation (1), is not linear.

The form of the independent variable of this equation as the product of two variables ($\sigma_c\rho_r$) does not allow an exact prediction of the compression stress necessary to obtain a tablet of a specified mechanical quality. Here, in practice, it is necessary to know the exact relation existing between compression stress σ_c and relative density ρ_r. Nevertheless the magnitude of the parameters P_m and γ_P provide,

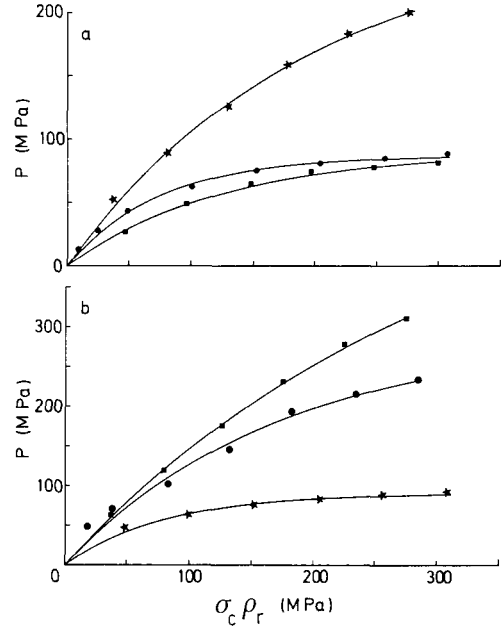

FIG. 1. Plot of indentation hardness P against the product of compression stress σ_c and relative density ρ_r according to eqn 1. (a) ● Aspirin FC. ■ Dipyrone. ★ Paracetamol DC. (b) ● Caffeine FGR. ■ Lactose. ★ Aspirin 150.

Table 1. Compaction parameters and indices of the materials studied.

Substance	P_m [MPa]	γ_P 10^2[MPa^{-1}]	σ_{Tm} [MPa]	γ_T 10^2[MPa^{-1}]	CPP 10^2[MPa^{-1}]	BI σ_{Tm}/P_m
Aspirin 150	91·37	1·23	3·13	0·70	0·53	0·034
Aspirin FC	87·17	1·34	2·96	0·70	0·64	0·034
ASS cryst.	88·29	1·26	2·43	0·88	0·38	0·028
ASS powder	92·65	1·39	3·57	0·74	0·65	0·039
Microcrystalline cellulose	168·26	1·33	8·50	1·49	−0·16	0·051
Caffeine FGR	288·12	0·58	2·26	2·19	−1·61	0·008
Caffeine powder	289·56	0·58	2·71	1·78	−1·20	0·009
Emcompress	752·25	0·17	14·12	0·11	0·06	0·019
Lactose	534·25	0·32	12·77	0·19	0·13	0·024
Mannitol	308·92	0·32	8·76	0·16	0·16	0·028
Dipyrone	90·82	0·79	2·38	0·37	0·42	0·026
Paracetamol DC	265·05	0·51	4·43	0·66	−0·15	0·017

in a quantitative way, useful basic information about the compaction of powders. P_m and γ_P are characteristic of the deformation behaviour of the material under compaction stress (Jetzer et al 1983). Materials that undergo predominantly plastic deformation, such as the investigated aspirin samples and dipyrone, attain a relative density close to unity at relatively low pressure (see Fig. 1a, b). Further increase in compression stress did not yield higher values of indentation hardness. An exception was observed with easily deformable materials in which deformation hardness of the compacts goes through a maximum. The decrease could be partly related to the presence of solvent traces, probably causing a change in lattice defects during compaction (Jetzer & Leuenberger 1985).

In the case of brittle materials (caffeine, Emcompress, lactose and mannitol), by contrast extremely high loads are necessary to produce non-porous tablets. The P_m value of these materials is only the best estimate obtained as an extrapolation of indentation hardness and compression stress data at lower relative densities (see Fig. 1a, b).

The graphical representation of tensile strength measurements of the compacts is also, as expected, not usually linear (see Fig. 2a, b). The observation that tensile strength values σ_T of caffeine tablets tend to reach a maximum value σ_{Tm} at porosity <0 must obviously be interpreted differently, as in the case of hardness measurements. This behaviour suggests a beginning of fracture tendency. Further increase in compressional force can produce capping or lamination, resulting in a dramatic strength decrease*. On the other hand, hardness is not very sensitive to capping phenomena. In fact, if a laminated compact is produced, the individual pieces are dense and strongly bonded.

Values of σ_T are principally used to quantify *resistance to fracture*. The exact determination of tensile strength depends upon the correct state of stress developing within the compact. Complex stress distribution, such as shear, tension and compression, can lead to various modes of fracture. It has been shown that to obtain reproducible results for strength, the compact must break in such a manner that tensile stress is the major stress (Fell & Newton 1970). A simple tension fracture can be identified easily as the specimen fails along the loaded diameter (ideal tensile failure) (Newton et al 1971).

For brittle materials the diametral compression test provides an experimentally simple method for measuring tensile strength. Compacts of predominantly plastic and/or viscoelastic materials usually exhibit complicated fracture patterns because they do not purely break in tension. In this case measurement provides only an approximate value of tensile strength, limiting the range of applicability and comparability of the test.

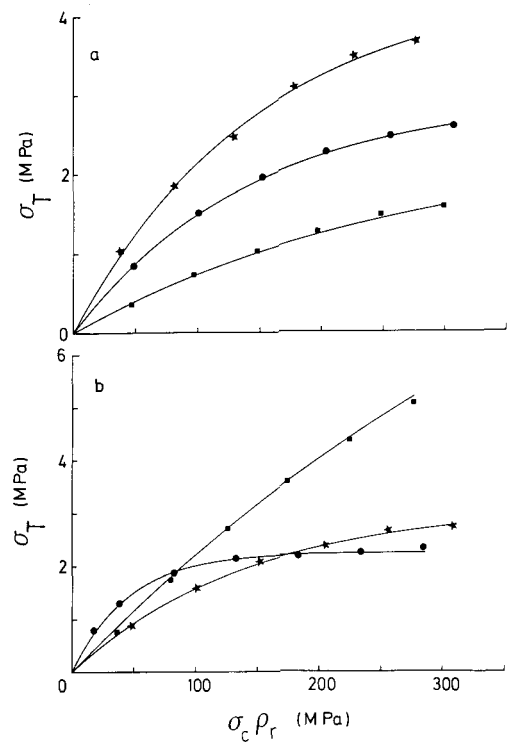

FIG. 2. Plot of tensile strength σ_T against the product of compression stress σ_c and relative density ρ_r according to eqn 2. (a) ● Aspirin FC. ■ Dipyrone. ★ Paracetamol DC. (b) ● Caffeine FGR. ■ Lactose. ★ Aspirin 150.

The scale of the irreversible local indentation is determined by P values, quantifying the *resistance to deformation*. In other words P is a measure of plasticity. For compacts, which are porous solids, inaccurate values are measured if the deformation-controlled indentation is influenced by crack propagation in the particles or between their bonded areas.

Both hardness and strength of a tablet are influenced by two competing mechanical responses—deformation and fracture—of the material towards pressure. In order to make comparisons it is obviously necessary to check the mode of failure and the indentation field.

For a study of a material's behaviour during the process of tablet formation, it therefore seems

* All tablets tested were visually examined before measuring the mechanical qualities; those showing laminar cracks were rejected.

reasonable to use a unifying description in terms of hardness and strength.

The difference $(\gamma_P - \gamma_T) = \Delta\gamma_{PT}$, called compaction performance parameter (CPP)†, obtained in equation (3) is an interesting parameter for this purpose. According to the material's performance, the values $(\gamma_P - \gamma_T)$ which are listed in Table 1 can assume a positive or negative magnitude. The two γ values specify the rate at which the compact hardness and tensile strength builds up with an increase in the applied compression stress and relative density. Efforts are under way to resolve the exact physical significance of the compression susceptibility parameter. The dimensions of γ are Pa^{-1} which are equal to those of volume/energy. One may therefore speculate that $1/\gamma_P$ can be interpreted as a volume or mass specific activation energy required to induce deformation or fragmentation in the material. Within experimental error the magnitude of γ_P was found to be equal to the K-proportionality constant of the Heckel equation, a parameter that is related to the yield pressure of the material (Heckel 1961; Leuenberger & Jetzer 1984).

During indentation hardness and tensile strength measurements γ_P and γ_T are influenced by different stress states. In the case of the hardness test the stress is compressive and is associated with a certain amount of plastic deformation which depends upon the work hardening characteristics of the material and test conditions. On the other hand, during radial tensile strength measurements, failure is associated with a specific amount of stress, i.e. shear stress and tension, at the maximum load.

Materials with predominantly plastic deformation properties, like the aspirin and dipyrone samples investigated, show a greater positive magnitude of $\Delta\gamma_{PT}$. In contrast, for materials known to exhibit predominantly a brittle behaviour (dicalcium phosphate, lactose and mannitol), the difference $\Delta\gamma_{PT}$ approaches zero, suggesting that phenomena related to plastic materials, such as work hardening, should not be relevant. This would also suggest that both hardness and strength measurements were made at the same point on a hypothetical stress–strain diagram. Here, both hardness and strength build up at the same rate as densification increases. A negative CPP was found in the case of caffeine, paracetamol DC and microcrystalline cellulose. At higher pressure levels both caffeine and paracetamol DC were found to exhibit a high degree of fracture tendency (Jetzer & Leuenberger 1984). During the radial tensile strength measurement, all the bonding points within the tablet are subjected to stresses. According to Griffith's theory, a defect which may be present will propagate if stress equals bonding energy, inducing a premature tensile failure. This behaviour is related to higher γ_T values causing the difference $\Delta\gamma_{PT}$ to assume negative values. An inhomogeneous structure in a tablet causes its tensile strength to be much lower than its hardness would indicate.

For the microcrystalline cellulose tested, the observed value $\Delta\gamma_{PT} = -0{\cdot}16$ is somewhat surprising. Here, other considerations have to be made as this material showed significant viscoelastic properties. Instead of ideal failure (Newton et al 1971), complicated fracture patterns were observed, suggesting that the elastic and plastic properties in some way influence the failure process (Rees & Rue 1978).

At the same time, a more suitable procedure for recording deformation of this type would be to measure indentation depth before and after elastic recovery has taken place (Aulton 1977) or to use a dynamic method which permits calculation of a strain index (Hiestand & Smith 1984). Here it is evident that one single parameter or index is not sufficient to describe the variety of compaction properties that may be incurred. For this purpose the bonding index σ_{Tm}/P_m gives additional information about the tableting behaviour. This index is believed to reflect the extent of survival during decompression and/or ejection of true contact areas that were established at maximum compression stress. Among the materials studied, microcrystalline cellulose exhibits the highest BI value of 0·051 (see Table 1) which is also consistent with the value given by Hiestand & Smith (1984), even using a different experimental procedure. In fact, in pharmaceutical practice this material is known to have excellent properties as a bonding agent and, furthermore, it is not brittle. A reason for the occurrence of a negative CPP value has therefore probably to be found in the viscoelastic properties of the material which may lead to inaccurate deformation and strength measurements with the technique of disposition.

Paracetamol DC and, especially, caffeine samples exhibit low BI values confirming the acute degree of fracture tendency when high pressures are applied. The other materials with essentially plastic or brittle deformation properties show larger BI values. In this case the magnitude of both CPP and BI indicates that fracture problems should not occur in the range of

† Obviously, instead of using the parameter $\Delta\gamma_{PT}$, it is also possible to formulate a dimensionless compaction performance index as the ratio γ_P/γ_T which gives the same information.

pressures used to test the compacts. The data suggest also that capping problems are often associated with brittleness but the contrary is not true.

CONCLUSION

Compression parameters and indices, obtained from hardness and tensile strength measurements such as the bonding index and the compaction performance parameter, make it possible to interpret the behaviour of a material during compaction and, furthermore, to predict when capping problems occur. It is evident that more than one index or parameter is necessary to describe the variety and complexity of the tableting properties.

Because of the previously discussed problems such as, e.g. fractures and non-ideal failure that may arise during indentation hardness and tensile strength measurements, caution is needed in comparisons of data.

The results reported are specific for the particular materials used in this test. Among other things, the fundamental mechanical properties depend on particle size and shape and moisture content of the material. It is obvious that indentation hardness and tensile strength measurements need to be performed under the same experimental conditions.

REFERENCES

Aulton, M. (1977) Manuf. Chem. Aerosol News 48: 28–36

Fell, J. T., Newton, J. M. (1970) J. Pharm. Sci. 59: 688–691

Heckel, R. W. (1961) Trans. Metall. Soc. AIME 221: 1001–1008

Hiestand, E. N., Smith, D. P. (1984) Powder Technology 38: 145–159

Jetzer, W. (1982) Ph.D. Thesis, University of Basel, Switzerland

Jetzer, W., Leuenberger, H. (1984) Pharm. Acta Helv. 59: 2–7

Jetzer, W., Leuenberger, H. (1985) Powder Technology 42: 137–144

Jetzer, W., Leuenberger, H., Sucker, H. (1983) Pharm. Technol. 7: 33–39

Jetzer, W., Johnson, B. W., Hiestand, E. N. (1985) Int. J. Pharm. 26: 329–337

Leuenberger, H., Jetzer, W. (1984) Powder Technology 37: 209–218

Leuenberger, H., Hietand, E., Sucker, H. (1981) Chem. Ingenieur Techn. 53: 45–47

Newton, J. M., Rowley, G., Fell, J. T., Peacock, D. G., Ridgway, K. (1971) J. Pharm. Pharmacol. 23, Suppl.: 195S–201S

Rees, J. E., Rue, P. J. (1978) Drug Dev. Ind. Pharm. 4: 131–156