J. Pharm. Pharmac., 1974, **26**, 289–297 *Received November* 8, 1973

Compression characteristics of powders: radial die wall pressure transmission and density changes

J. E. CARLESS AND S. LEIGH

Department of Pharmacy, Chelsea College (University of London), Manresa Road, London, SW3 6LX U.K.

An instrumented single punch tablet machine was used to study the relations between axial pressure, radial die wall pressure and density changes in beds of powder undergoing compression. Aspirin, sodium chloride, paracetamol, paracetamol-polyvinylpyrrolidone and sucrose crystals were examined. The development and transmission of radial pressure during loading and unloading of the bed is discussed in relation to yield and elastic behaviour of the powder. The relation between mean stress and relative density of the compact is explained on the basis of plastic flow and/or crushing of the particles. Differences in stress *vs* density relations due to difference in particle size of sucrose were due to initial packing conditions in the die and not to differences in pressure transmission.

This paper is an extension of previous work (Leigh, Carless & Burt, 1967) where the behaviour of the powder bed in the die during loading and unloading stages was studied and its significance to the tableting process was discussed. The "compression cycles" so obtained enabled a distinction to be made between an elastic material, a constant stress material and a Mohr body for which the yield stress in shear is a function of the applied normal stress. Ridgway, Glasby & Rosser (1969a) confirmed the general shape of these compression cycles and related the radial die wall pressure to the degree of hardness of the material.

The relation between the force transmitted to the die wall can be expressed as a "stress ratio", i.e. ratio of radial to axial stress. This ratio has been incorporated in basic equations relating to die wall friction and pressure transmission (Shaxby & Evans, 1923; Unckel, 1945; Shank & Wulff, 1949). Nelson, Busse & Higuchi (1955) and Windheuser, Misra & others (1963) have reported on the magnitude of radial stress developed during compaction in relation to the physical properties of the powdered material and final compact. Higuchi, Shimamoto & others (1965) have observed the rate of decay of die wall pressure as the punch pressure was relaxed.

MATERIALS AND METHODS

Aspirin B.P. −20 +40 mesh crystals. Density 1·39g cm^{-3}. Moisture content 0·25%.

Paracetamol granules −20 +40 mesh. Prepared by moist granulation (acetone; No. 20 mesh sieve) of paracetamol B.P. Density 1·3g cm^{-3}. Moisture content 0·2%.

Paracetamol—PVP granules −20 +40 mesh. Prepared by granulating paracetamol (97 parts) and polyvinylpyrrolidone (PVP) (Luviskol VA64) with acetone. Density 1·3g cm^{-3}. Moisture content 0·2%.

Sodium chloride B.P. −20 +40 mesh. Density 2·17g cm^{-3}. Moisture content 0·1%.

Sucrose B.P. Crystals were graded into fractions:— −22 +45; −30 +36; −44 +52; −60 +72; −85 +100 mesh sizes. Density 1·59g cm^{-3}. Moisture content 0·15—0·18%.

Densities were determined using specific gravity measurements with xylene as displacement fluid.

All materials were dried over silica gel in a vacuum oven at 65° overnight and stored in Kilner jars.

The moisture content for aspirin was determined by loss of weight over P_2O_5 in vacuum oven at 65°. Moisture content of the other materials was determined from loss of weight at 95° under an infrared lamp (Denward Moisture Determination Apparatus).

Apparatus and procedure

A modified Manesty E2 single punch machine was essentially that described by Leigh & others (1967). Flat ½″ punches were used. Additional modifications included a linear displacement transducer to monitor the position of the top punch and an improved strain gauge circuit giving increased sensitivity (Leigh, 1969).

The die wall was lubricated with 5% stearic acid in chloroform before each compression. The material for each tablet was weighed ±1 mg and introduced into the die to give tablet 4 mm ± 0·02 mm thickness at 237 MN m^{-2}. The bottom punch was raised and lowered a few times to spread the material uniformly before compression.

The compression cycles were determined by applying a punch pressure in increments of 21·6 MN m^{-2} every 0·5 s to the maximum of 237 MN m^{-2}. The punch pressure was then reduced in the same step wise manner. The cycle was completed in 11 s. A typical recording of the forces developed during a compression cycle (machine speed 50 tablets min^{-1}) is shown in Fig. 1. For the incremental method 5 readings were taken and the mean value used to plot each point in the compression cycle (e.g. Fig. 2).

The height of the compact during compression was determined from the position of the top punch. A correction for punch deformation was made by compressing a specially hardened steel disc (4 mm × 12 mm) up to 3000 kg load and the elastic deformation was followed from the linear displacement transducer readings. The relative densities of the compacts were calculated from the density of the solid and its volume within the die.

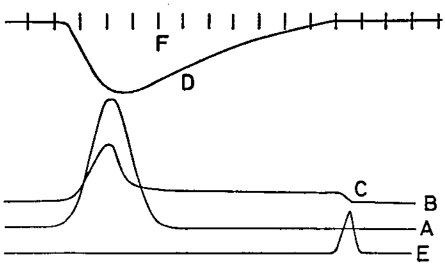

FIG. 1. Typical recording of forces developed during a compression cycle. A = axial force; B = radial force; C = residual die-wall force; D = top punch displacement; E = ejection force; F = timing marks (0·1 s).

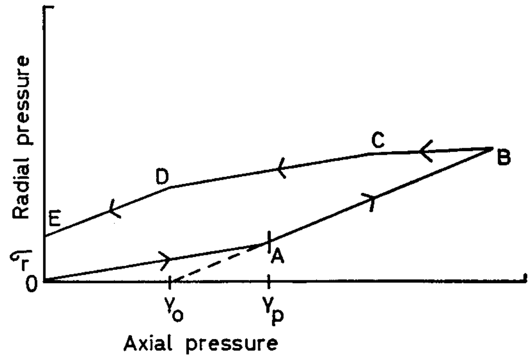

FIG. 2. Relation between axial and radial pressures during compaction of powder in die.

Radial and axial stresses (Mn m^{-2}) were calculated from the total force exerted at the die wall and upper punch respectively, divided by the area over which the force was applied. The die wall area was calculated from the tablet height at each compression.

RESULTS

Compression cycles

Fig. 2 shows diagrammatically the compression cycle of axial *vs* radial stress. The stress ratio defined by radial stress/axial stress for the different regions of the cycle is given in Table 1. During the compression stage O-A, a break at A occurs at a yield pressure Yp. Yo is an arbitrary yield value obtained by extrapolating BA to zero axial pressure. Maximum pressure is exerted at B and the upper punch is then released gradually from B to E. On complete removal of axial pressure, a residual die wall pressure σ_r was exerted by all the materials studied but was much higher for sodium chloride than for aspirin and paracetamol (Table 1).

To evaluate the effect of particle size on the compression cycles, five narrow sized fractions of sucrose crystals were compressed. The results appear in Fig. 3 and it can be seen that the yield point A occurs at higher axial pressures as the particle size is reduced. The residual die wall pressure also increases with decreasing particle size.

Table 1. *Stress ratios of materials for complete compression cycle, i.e. loading and unloading.*

Material	Size (mesh)	Stress ratios					σ_r MN m^{-2}
		OA	AB	BC	CD	DE	
Aspirin crystals	20/40	0·44	0·47	0·30	0·50	0·67	9·8
Sodium chloride crystals	20/40	0·36	0·49	0·07	0·34	1·63	29·4
Paracetamol granules	20/40	0·27	0·34	0·18	0·29	3·47	5·9
Paracetamol-PVP	20/40	0·29	0·39	0·20	0·27	0·50	14.7
Sucrose crystals	85/100	0·31	0·37	0·12	0·24	0·32	31·4
	60/72	0·30	0·36	0·13	0·25	0·31	29·4
	44/52	0·29	0·36	0·13	0·24	0·33	26·4
	30/36	0·28	0·35	0·14	0·24	0·34	26·4
	22/25	0·23	0·32	0·14	0·25	0·34	22·1

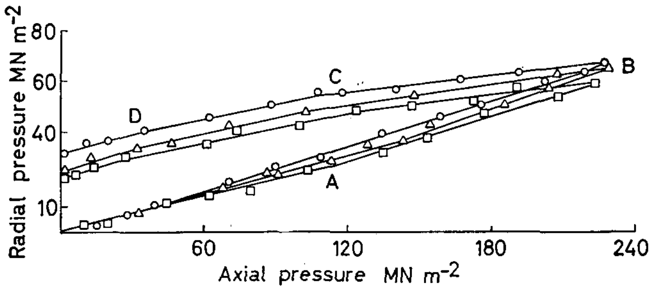

FIG. 3. Relation between axial and radial pressure; sucrose crystals. □ 22/25 mesh; △ 30/36 mesh; ○ 85/100 mesh.

Relative density changes

The change of relative density of the powder bed with mean stress is shown diagrammatically in Fig. 4. Mean stress, which is the mean of the axial and radial stress can be regarded as the effective stress acting on the bed (Leigh, 1969). Plastic deformation and/or crushing of the particles occurs between the mean stress values $\bar{\sigma}_1$ to $^{-}{}_2$, corresponding to relative densities $\bar{\sigma}_1$ and $\bar{\sigma}_2$ respectively. The values are determined from the plot of log mean stress against relative density and noting where the line departs from linearity at high and low stress values. Relative density *vs* log mean stress (limits $\bar{\sigma}_1$ to $\bar{\sigma}_2$) plots for aspirin, paracetamol and sodium chloride are shown in Fig. 5A.

The effect of particle size on densification during compression was investigated for sucrose crystals (Fig. 5B). It can be seen that the relative density of the compact decreased slightly with decrease in particle size.

Elastic recovery

Each material was subjected to a maximum axial pressure of 39, 98, 157, 206 and 236 MN m^{-2} and the thickness of the compact measured with the linear displacement transducer at each of these pressures. The axial pressure was gradually reduced until negligible pressure was detected by the punch and the thickness of the tablet again measured. The percentage recovery (k) of the tablet within the die was plotted against the mean stress applied and some values are given in Table 2 together with the percentage total recovery of the ejected tablet.

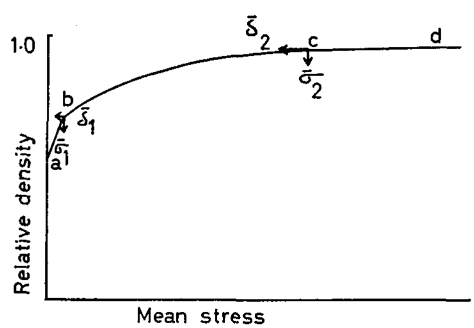

FIG. 4. Relation between mean stress and relative density of compacts.

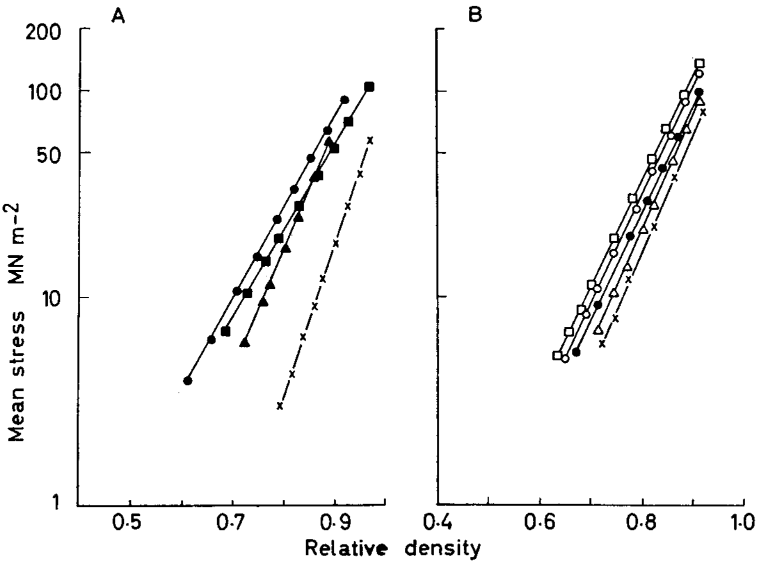

FIG. 5A. Relation between log mean stress and relative density during plastic flow/crushing in compressed bed. ■ sodium chloride; × aspirin; ▲ paracetamol; ● paracetamol-PVP.

B. Influence of particle size of sucrose crystals during plastic flow/crushing in compressed bed. × 22/25 mesh; △ 30/36 mesh; ● 44/52 mesh; ○ 60/72 mesh; □ 85/100 mesh.

DISCUSSION

Application of punch pressure

When the powder is filled into the die, the particles are randomly packed and the density approximates to the tap density. The application of axial pressure results in the reorganization and rearrangement of particles (Cooper & Eaton, 1962) but, in addition, a proportion of the axial pressure is transmitted through point and line contacts between particles to the die wall resulting in the stress ratio given by OA (Fig. 2). At point A in Fig. 2, maximum point and line contacts between closely packed particles have been established and further increases in axial pressure result in substantial deformation and/or fracture of the particles to fill in the voids.

This results in a bulk structure sufficiently strong to support the imposed load and the pressure transmission is now seen to increase as the powder bed assumes the behaviour of a solid body. It may be postulated that, although the development

Table 2. *Percentage recovery of compact after removal of upper punch.*

Material	Mean stress MN m⁻² 29		88		147	
	a	b	a	b	a	b
Aspirin	1·0	1·0	1·8	1·8	2·5	2·9
Sodium chloride	0·0	0·1	0·0	0·6	0·6	2·1
Paracetamol	0·5	2·9	3·8	7·0	7·1	cap.
Paracetamol—PVP	0·5	0·9	0·7	1·1	2·0	2·8

a = % recovery in die.
b = % total recovery of ejected tablet.
cap = capping.

of radial pressure up to A is due to pressure exerted on the die wall as a result of powder being forced into voids, at A a structure strong enough to support the imposed load is formed, and as axial pressure is increased the structure expands laterally to give a stress ratio AB. AB is thus a measure of the elastic compressibility of the bulk material.

From the volume changes of the powder on compression (Fig. 4) it was found that the increase of relative density was a logarithmic function of the mean stress between the limits $\bar{\sigma}_1$ and $\bar{\sigma}_2$. The axial pressure $\bar{\sigma}_2$ corresponds to a pressure slightly above yield point A in Fig. 2 so that stresses greater than $\bar{\sigma}_2$ will result in elastic compression of the bulk material, i.e. region C to D. At stresses below σ_1, the linear relation between density and mean stress region A to B is likely to be due to rearrangement of the particles to give closer packing. The following equation expresses the relation between mean stress and relative density:

$$\omega = \log \frac{\bar{\sigma}_2}{\sigma_1} / \phi \qquad \ldots \quad \ldots \quad \ldots \quad \ldots \quad \ldots \quad \ldots \qquad (1)$$

where ω = proportionality constant for plastic flow/crushing between limits set by $\bar{\sigma}_1$ and $\bar{\sigma}_2$; ϕ = increase in relative density from $\bar{\delta}_1$ to $\bar{\delta}_2$; $\bar{\sigma}_1$ = mean stress corresponding to relative density $\bar{\delta}_1$; $\bar{\sigma}_2$ = mean stress corresponding to relative density $\bar{\delta}_2$.

Although the term "yield" is used to describe the event at "A" it is unlikely that this is due to a "plastic gliding process of total internal structural elements" as defined by Houwink (1958) and is probably similar to the "pressure of fluidity" of O'Neil & Greenwood (1932). This seems a reasonable assumption because the radial transmission after "yield" remains linear and each compact shows considerable elastic recovery when pressure is released. Therefore, it seems likely that, although some form of plastic flow or crushing has taken place at "A", this is confined mainly to the interparticulate regions and total plastic flow of the bulk material has not occurred. Photomicrographs of the crushed compacts formed at pressure levels corresponding to points "A" and "B" of Fig. 2 confirm this view.

The compression behaviour of different materials can be characterized in terms of equation 1. Values of ω, $\bar{\sigma}_1$, $\bar{\delta}_1$ and $\bar{\delta}_2$ are tabulated in Table 3. Sodium chloride has smallest value for ω indicating that considerable interparticle yielding accompanies each increment of pressure. Aspirin shows quite a different effect and considerable deformation/crushing occurs at low pressures. It can be seen that paracetamol—PVP granules undergo more plastic flow/crushing than paracetamol alone. The low value of $\bar{\sigma}_2$ paracetamol is evidence that plastic flow/crushing is virtually complete at a mean stress of 59 MN m^{-2}, whilst paracetamol-PVP granules continued to yield up to 96 MN m^{-2}. These values were close to the yield pressures (Yp) given by "A" in the compression cycles (Fig. 2) and thus support the proposed sequence of changes in the compact during compression.

We have not measured the hardness of the crystals or granules used in this work but the data of Ridgway & others (1969b) is given in Table 4 for aspirin, sodium chloride and sucrose.

The harder the material the higher the yield value but the proportion of force transmitted to the die wall (stress ratio) is less. Ridgway & others (1969a) found similar results for radial die wall transmission but yield values were not reported.

Table 3. *Parameters relating to plastic flow during compression.*

Material	ω	$\emptyset$	$\bar{\sigma}_1$ MN m^{-2}	$\bar{\sigma}_2$ MN m^{-2}
Sodium chloride	4·2	0·29	6·9	113
Aspirin	7·3	0·18	2·9	62
Paracetamol	5·9	0·17	5·9	58·9
Paracetamol-PV	4·5	0·31	3·9	96
Sucrose 85/100	5·2	0·28	4·9	144
60/72	5·4	0·26	5·1	128
44/52	5·3	0·24	5·4	103
30/36	5·7	0·24	6·9	95
22/25	5·7	0·20	7·9	82·4

ω = proportionality constant for plastic flow/crushing between limits set by $\bar{\sigma}_1$ and $\bar{\sigma}_2$.
$\emptyset$ = increase in relative density from $\bar{\delta}_1$ to $\bar{\delta}_2$.
$\bar{\sigma}_1$ = mean stress corresponding to relative density.
$\bar{\sigma}_2$ = mean stress corresponding to relative density.

Relaxation of punch pressure

Owing to the reorganization that has taken place in a powder bed under compression the relation between axial stress and radial stress will not be the same for compression as for relaxation. During compression both elastic potential energy and permanent strains are induced in the compact and these should be detected during the relaxation stage. The relaxation halves of the compression cycle in Fig. 2 indicate a plastic and elastic component. The elastic component is relieved as pressure is removed whilst the plastic component remains as a residual die wall pressure.

It is apparent that the radial pressure dissipates in three definite stages during release of axial pressure. Each stage in the relaxation is described by a different stress ratio (Table 2). The three different stress ratios during relaxation as shown by each material suggest that radial pressure dissipation is non-uniform and each break could be ascribed to the fact that the axial pressure at that point is not sufficient to overcome the radial pressure exerted by the die wall. Alternatively, it may be postulated that the frictional forces at the compact/die wall interface decrease as the axial loading is reduced, and this allows for greater relaxation. The values of the ratios listed in Table 1 reveal that in each case, the ratio (CD) corresponds quite well to the stress ratio (AB) of the first half of the compression cycle, indicating that the processes of pressure transmission and relaxation follow essentially similar elastic behaviour.

The high stress ratio DE shown by paracetamol (Table 1) corresponds to a considerable elastic recovery when the axial pressure is withdrawn (Table 2). The high elastic recovery could be responsible for the breakage of interparticulate bonds

Table 4. *Comparison of yield value, hardness and stress ratio of compacts.*

Material	Yield value Y_p MN m^{-2}	*Hardness of crystal MN m^{-2}	Stress ratio of compacts
Aspirin	88·3	85	0·44
Sodium chloride	118	208	0·36
Sucrose 85/100	196	625	0·31

* Values from Ridgway, Shotton & Glasby (1969).

leading to capping as the tablet is ejected. This contrasts with the behaviour of paracetamol—PVP, which shows a low stress ratio, a reduced elastic recovery and absence of capping.

The magnitude of the residual stress in the compact can be assessed to some extent by the residual die wall pressure, i.e. pressure exerted after removal of upper punch. Materials that undergo irreversible deformation, e.g. plastic flow, will show a high residual die wall pressure, whilst elastic materials will have lower values provided that die wall friction does not inhibit the relaxation of the compact. Sodium chloride exhibits a high residual stress whilst aspirin and paracetamol have a considerably lower value (Table 1). These results are compatible with the elastic recovery of the compacts shown in Table 2.

Influence of particle size

The pressure cycles for sucrose in Fig. 3 show that the yield pressure indicated by A increases significantly with the decreasing particle size. In the case of the finest fraction (85/100) it appears that the yield pressure was just reached before relaxation commenced. It may also be noted that the apparent yield value (Yo) also increases for the finer crystals. The increased transmission of smaller particles may be due to (*a*) more numerous point contacts which increase the total contact areas, facilitating transmission of applied forces, (*b*) increased shear stresses at the die wall.

It may also be seen that the 85/100 mesh crystals exert a higher residual die wall pressure than the coarser 22/25 mesh crystals. A possible explanation is the increased number of junctions in the fine powder which results in a more cohesive structure that would relax to a lesser extent than that formed from coarser particles. In addition, the increased transmission of the finer particles results in higher frictional forces at the die wall which could restrain recovery of the compact in the die.

Referring to the relative density changes for the different size fractions, it appears that $\bar{\sigma}_2$ increases markedly and the values for the five different sizes are recorded in Table 3. This is in accord with the higher yield pressures denoted by point A in the pressure cycles (Fig. 2), as particle size decreases. Table 3 also reveals that whilst the exponential constant ω decreases only slightly with particle size, the parameter Ø increases considerably. Both these constants indicate the relative tendency for plastic flow/crushing in a compressed bed. Since higher values for Ø are associated with greater densification, this term could be a more useful parameter for assessing the influence of particle size on the compaction process. It seems reasonable to assume that the constant ω should be essentially a property of the bulk material although the degree of densification could vary with the particle size, distribution and packing characteristics. Fig. 5B also shows that the relative density at a given pressure is higher for the coarser powders and this can be due to the coarser bed being more closely packed at the commencement of radial transmission. If correction is made for the difference in densities at the start of compression, then the data from Fig. 5B can be fitted to a common straight line. This suggests that the differences due to particle size in pressure/density relations are only functions of the packing conditions and not due to any significant differences in pressure transmission.

CONCLUSIONS

The development and transmission of radial pressure during compaction is due to the mechanisms of rearrangement, plastic flow/crushing of the particles followed by elastic compression of the bulk structure formed. The pattern and magnitude of the radial transmission reflect the influence of the physical properties of the material on these mechanisms, whilst pressure-density observations reveal the nature and sequence of these stages. The recovery pattern of the compact, moreover, indicates the elastic properties of the material and also the extent of plastic flow/crushing that has taken place. Therefore, a study of the complete pressure cycle in association with the density changes during both the application and the withdrawal of compacting pressures would yield useful information regarding the compression behaviour of materials and the data derived could be used to characterize these materials. It has been demonstrated that the stress ratio for a particular system at low pressure depends on a number of factors such as particle size, elastic/plastic properties and packing conditions of the bed. It is therefore suggested that this term could be used to exercise control over these properties of a powdered material. The yield pressure indicates the stresses at which the interparticle contacts yield to form a bulk structure. The stress ratio after yield therefore reflects the compressibility of the bulk material. The radial pressure relaxation behaviour of the compact as axial pressure is released reveals the elastic recovery of the material. High residual die wall pressures may be attributed to considerable plastic flow/crushing of the particles.

There is a growing awareness of and emphasis on the dissolution profiles of tablets. Dissolution is governed by the degree of fusion/adhesion between particles in a compact so that standardization of the materials in terms of their compression characteristics may prove useful in this area.

REFERENCES

Cooper, A. R. & Eaton, L. E. (1962). *J. Am. Ceram. Soc.*, **45**, 97–101.

Higuchi, T., Shimamoto, T., Ericksen, S. P. & Yashiki, T. (1965). *J. pharm. Sci.*, **54**, 111–118.

Houwink, R. (1958). *Elasticity, plasticity and structure of matter.* 2nd Edn. New York: Dover Publications.

Leigh, S. (1969). Ph.D. Thesis. London University.

Leigh, S., Carless, J. E. & Burt, B. W. (1967). *J. pharm. Sci.*, **56**, 888–892.

Nelson, E., Busse, L. W. & Higuchi, T. (1955). *J. Am. Pharm. Ass.* (*Sci. Edn.*), **44**, 223–225.

O'Neil, H. & Greenwood, H. J. (1932). *J. Inst. Metals*, **48**, 47–67.

Ridgway, K., Glasby, J. & Rosser, R. H. (1969a). *J. Pharm. Pharmac.*, **21**, *Suppl.*, 24S–29S.

Ridgway, K. Shotton, E. & Glasby, J. (1969b). *Ibid.*, **21**, *Suppl.* 19S–23S.

Shank, M. E. & Wulff, J. (1949). *J. Trans. Amer. Inst. Min.* (*metall*)*Engrs*, **185**, 561–570.

Shaxby, J. H. & Evans, J. C. (1923). *Trans. Faraday. Soc.*, **19**, 60–89.

Unckel, H. (1945). *Arch. Eisenhutt.*, **18**, 161–167.

Windheuser, J. J., Misra, L., Ericksen, P. S. & Higuchi, T. (1963). *J. pharm. Sci.*, **52**, 767–772.