

An infrared spectrophotometric study of the compaction mechanism of potassium bromide

K. RIDGWAY AND M. E. AULTON*

*Department of Pharmaceutics, The School of Pharmacy,
Brunswick Square, London, WC1N 1AX, U.K.*

Potassium bromide has been compacted in a vacuum die at pressures up to 1000 MN m^{-2} , and the infrared transmittance of the resulting flat discs measured. Four mean particle sizes were used: 71, 110, 388 and $550 \mu\text{m}$. Over the whole range of pressure, the $110 \mu\text{m}$ material had a higher transmittance. At any one particle size, the transmittance increased with increasing compaction pressure up to 400 MN m^{-2} , then fell slightly and was followed by a less pronounced increase in the region of 1000 MN m^{-2} . The following mechanical properties of the compacts were also investigated: tensile strength, relative density, surface hardness and dissolution time. The transmittance changes have been related to the probable compaction mechanism.

The potassium bromide disc technique is well-known as an infrared spectrophotometric analytical method. Since potassium bromide does not absorb in the near infrared region, any transmission losses in such a disc are due either to surface reflection or to scattering within the disc. The degree of scattering will be related to the occurrence of free surfaces within the disc, and of other interactions depending on the ratio of the radiation wavelength to the particle size of the material forming the disc. Thus, the possibility exists of following the stages of the compaction process by observing changes in the transmittance of the compact. Potassium bromide in a range of particle sizes and powder weights was compacted in a vacuum die at various applied pressures to produce flat discs of a range of thicknesses. The infrared transmittance of the discs was measured. A number of mechanical properties of the discs was also investigated in the hope that transmittance changes occurring during compression could be explained and possibly related to the compaction mechanism. The properties investigated were: tensile strength, relative density (voidage), surface hardness and dissolution time.

MATERIALS AND METHODS

Materials

Potassium bromide (Analar grade, B.D.H. Ltd.) was sieved into $355\text{--}400 \mu\text{m}$ (mean $388 \mu\text{m}$) and $500\text{--}600 \mu\text{m}$ (mean $550 \mu\text{m}$) fractions; a proportion of this material was ball-milled for 2 h and $53\text{--}90 \mu\text{m}$ (mean $71 \mu\text{m}$) and $90\text{--}140 \mu\text{m}$ (mean $110 \mu\text{m}$) fractions separated.

Methods

Potassium bromide from all these fractions was compacted into flat discs or tablets 13 mm in diameter, in a vacuum die (Research and Industrial Instruments Co.),

* Now at School of Pharmacy, The Polytechnic, Leicester, U.K.

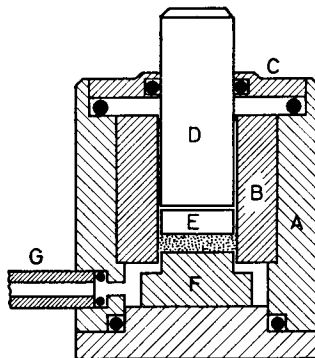


FIG. 1. The vacuum die used to prepare the potassium bromide discs. Key: A, outer casing; B, die-body proper; C, top cap to exclude air; D, upper punch; E, polished disc; F, lower punch, polished on its upper surface; G, vacuum offtake pipe.

based on the design of Ford & Wilkinson (1954). A vertical section of the die is shown in Fig. 1. The parts A, B and F were fitted together and a weighed quantity of potassium bromide, which had previously been dried at 110° for several hours, was introduced into the die-bore B. The powder was distributed evenly by lightly shaking until its surface was flat and level. The plunger D was then slowly rotated in the bore of B with no applied pressure, and removed. The top pellet, E, was inserted with its polished surface in contact with the powder, and the plunger D replaced. The assembly was completed with the rubber O-ring and the top cap C. The pipe G was attached to a Speedivac Pump Type 2 (W. Edwards & Co. Ltd.), which rapidly reduced the pressure to 6 mm Hg. The vacuum was maintained for 2 min, then compaction pressure was applied at a rate of $15 \text{ MN m}^{-2} \text{ s}^{-1}$ by means of a calibrated 20 ton hydraulic press (Tangye Ltd.). The required pressure was maintained for 2 min, then reduced at a similar rate; the vacuum was maintained for a further 1 min. The degree and duration of evacuation and compression were constant for all compacts. The interval between taking the dry powder from the container and closing the die was never greater than 2 min.

The disc was then removed by pushing it out with the plunger, D, in the hydraulic press. It was quickly transferred to a potassium bromide disc holder which was constructed for use in a Pye Unicam SP100 infrared spectrophotometer and the transmittance of the disc was measured working in the double-beam mode, between 650 cm^{-1} ($15.4 \mu\text{m}$) and 2510 cm^{-1} ($4.65 \mu\text{m}$). The reference cell contained an identical but empty disc-holder.

Discs were manufactured under the specified conditions from dried potassium bromide in 71, 110, 388 and $550 \mu\text{m}$ size fractions. At each size, discs weighing 0.25, 0.5, 0.75 and 1.0 g were prepared over the pressure range $100\text{--}1000 \text{ MN m}^{-2}$ resulting in disc thicknesses between 0.7 and 2.8 mm. A typical trace is shown in Fig. 2. Only one small adsorption band is present at about 1640 cm^{-1} ($6.1 \mu\text{m}$), corresponding to the O-H bending vibration of water. Other than in this wavelength region, the transmission loss is due entirely to reflection at the surface of the disc, scattering of radiation at interfaces such as cracks, dislocations and inclusions within the crystals, and at free surfaces caused by entrapped air. Surface reflection was shown to be negligible above 400 MN m^{-2} by making measurements with and without a second disc in the reference cell holder.

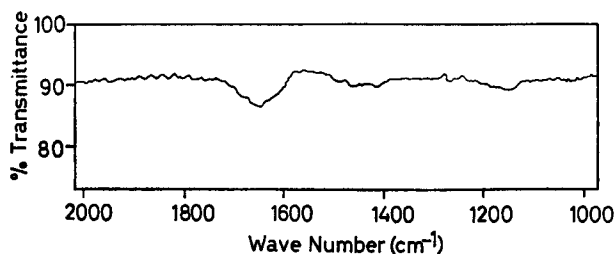


FIG. 2. The spectrogram of a potassium bromide disc, showing the weak absorption band at approximately 1640 cm^{-1} ($6.1\text{ }\mu\text{m}$), due to the O-H bending vibration of water.

A mean transmittance for each disc was determined by averaging the transmittance at 100 cm^{-1} intervals from 1000 to 2100 cm^{-1} , omitting the region of the adsorption peak, i.e. the 1600 and 1700 cm^{-1} values.

The tensile strength of the discs was measured by applying a diametral load to the disc, increasing at a constant rate until it fractured in tension along its diameter, using the apparatus of Shotton & Ganderton (1960), re-instrumented and re-calibrated by Leonard (1970). If the load at fracture is P , then the tensile strength of the compacted material is (Frocht, 1948):

$$\sigma = \frac{2P}{\pi Dt}$$

where D is the compact diameter and t is its thickness.

The relative density was obtained by measuring the diameter and thickness of a disc, together with its mass.

The surface hardness and elasticity of the discs were measured by the method of Ridgway, Aulton & Rosser (1970), the test material being indented with a sapphire sphere attached to a pneumatic micro-indentation apparatus (Research Equipment (London) Ltd.). The time interval between compaction and testing was kept as short as possible, and was never more than 2 h.

The dissolution time was measured in a B.P. apparatus modified by having five separate baskets so that five separate single-tablet determinations could be made simultaneously as a uniformity check. Water at 37° was the dissolution medium, and the oscillation rate was 30 min^{-1} .

RESULTS AND DISCUSSION

From the mean transmittance values for each disc, plots were made of $\log(\text{absorbance})$ against disc thickness (Fig. 3).

Three of the lines in Fig. 3 have equal slopes and yield an absorptivity of 0.275 mm^{-1} for the 71 and $110\text{ }\mu\text{m}$ particle size fractions at 400 MN m^{-2} , the pressure at which good compaction is achieved. This absorptivity value is thus characteristic of potassium bromide. At $388\text{ }\mu\text{m}$ particle size and above, the correlation is not so good, but at this size the particle diameter is comparable with the disc thickness.

The effect of particle size on absorbance can also easily be seen from Fig. 3; minimum absorbance (i.e. greatest transmittance) is shown by the $110\text{ }\mu\text{m}$ particle size; both 71 and $388\text{ }\mu\text{m}$ show increasingly greater absorbances than this. The $71\text{ }\mu\text{m}$ disc prepared at 600 MN m^{-2} compaction pressure can also be seen to have a greater transmittance than the one compacted at 400 MN m^{-2} . The pressure-

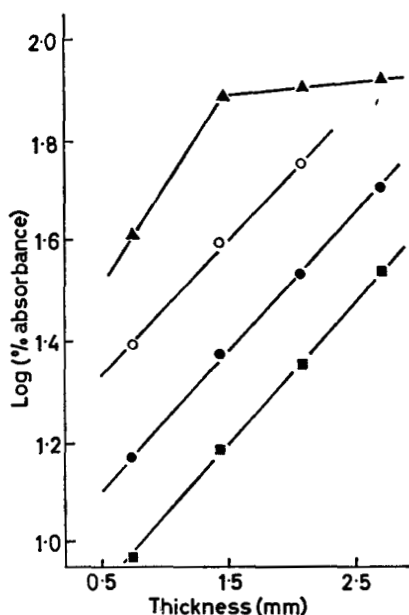


FIG. 3. The logarithm of absorbance ($\log I/I_0$) plotted against disc thickness. Key: \circ , 71 μm mean particle size, 400 MN m^{-2} compaction pressure; \bullet , 71 μm , 600 MN m^{-2} ; \blacksquare , 110 μm , 600 MN m^{-2} ; \blacktriangle , 388 μm , 600 MN m^{-2} . All the lines indicate an absorptivity of 0.275 mm^{-1} , though the larger material gives a poorer correlation.

transmittance relation was studied in greater detail by preparing discs from the same weight and particle size of powder at increasing compaction pressure in a range between 100 and 1000 MN m^{-2} .

A typical spectrogram for such a series is reproduced in Fig. 4. In the example shown, for discs prepared from 0.5 g of 110 μm particle size potassium bromide, the trace for 100 MN m^{-2} compaction pressure shows least transmittance. This was

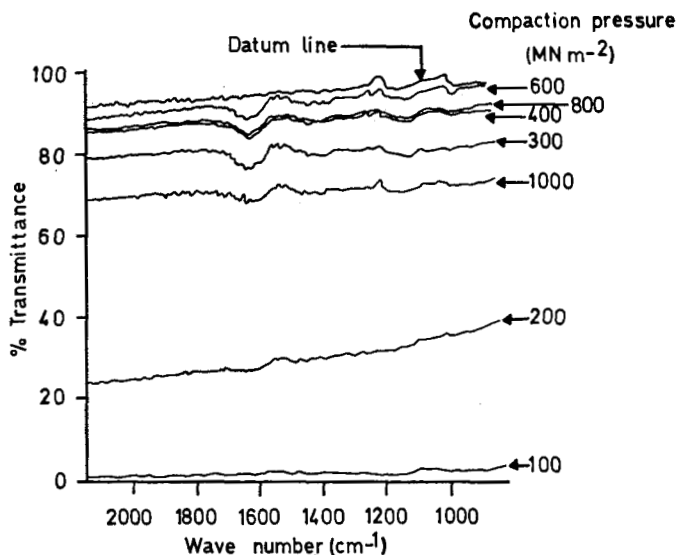


FIG. 4. Change in the potassium bromide spectrogram with increasing compaction pressure.

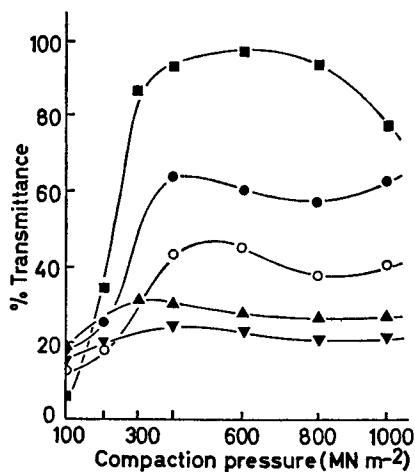


FIG. 5. Mean % transmittance as a function of compaction pressure. Key: ○, 71 μm mean particle size, 0.5 g compact weight; ●, 71 μm , 1.0 g; ■, 110 μm , 0.5 g; ▲, 388 μm , 0.5 g; ▼, 550 μm , 0.5 g.

a very opaque disc which also gave an appreciable amount of surface scatter in the visible region. Transmittance increased for the 200, 300 and 400 MN m^{-2} discs reaching a maximum at 600 MN m^{-2} . On further increase of compaction pressure the transmittance decreased, as shown by the 800 and 1000 MN m^{-2} traces.

The mean transmittances obtained from the spectrograms, corrected for the slight thickness differences, are plotted against compaction pressure in Fig. 5, together with the curves for the discs produced from 71 μm initial particle size (0.5 and 1.0 g) and the 388 and 550 μm powders (0.5 g).

Over practically the whole pressure range, the 110 μm material gave the higher transmittance, the 71, 388 and 550 μm particle sizes all scattering a greater proportion of the incident radiation. At any one particle size, the transmittances increased fairly rapidly with increasing compaction pressure, usually reaching a maximum at or about 400 MN m^{-2} . Thereafter, there was a fall, followed by a less pronounced rise in the region of 1000 MN m^{-2} . The change in transmittance with compaction pressure was most marked for the 110 μm powder. The shapes of the curves for the other powders were similar, although the changes in transmittance were less pronounced, becoming almost negligible for the 550 μm particle size disc.

The tensile strength of compacted potassium bromide as a function of disc thickness for various particle sizes at constant compaction pressure is shown in Fig. 6. In all cases there is a linear decrease in tensile strength with increasing thickness. The graph also indicates a dependence of the tensile strength of the resulting disc upon the initial particle size of the potassium bromide powder. Discs prepared from 71 μm potassium bromide showed the greatest strength, followed by those of 110 μm , the gradient of the line being the same as for 71 μm . The discs prepared from 388 μm and 550 μm KBr showed decreased tensile strengths together with a reduced slope.

The increase in tensile strength with increasing particle size indicates that tensile strength is a measure of some factor which is related to the surface area of the initial particles, suggesting that, at the compaction pressures applied, the major forces holding the disc together are those of welding together of the surfaces of the original

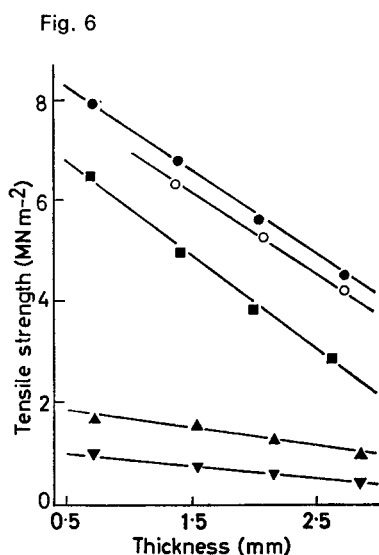


FIG. 6. The influence of thickness and initial particle size on the tensile strength of potassium bromide discs. Key: ○, 71 μm mean particle size, 400 MN m^{-2} compaction pressure; ●, 71 μm , 600 MN m^{-2} ; ■, 110 μm , 600 MN m^{-2} ; ▲, 388 μm , 600 MN m^{-2} ; ▼, 550 μm , 600 MN m^{-2} .

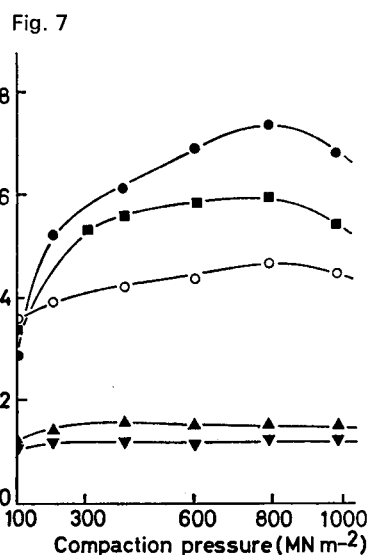


FIG. 7. Variation in tensile strength with compaction pressure. Key: ●, 71 μm mean particle size, 0.5 g disc weight; ○, 71 μm , 1.0 g; ■, 110 μm , 0.5 g; ▲, 388 μm , 0.5 g; ▼, 550 μm , 0.5 g.

particles. The welded bond is therefore the weakest part of the structure and has not attained the strength of the crystals themselves.

The decrease in the tensile strength of the compacts with increasing thickness can be explained by considering the distribution of forces within the compact. Forces within a thin disc will be approximately even and equal to the maximum applied (although buckling of the disc on removal from the die is sometimes observed, indicating that some uneven stresses remain in the compact). The forces within a thicker compact will decrease in a direction away from the moving punch (Train, 1957; Ridgway & others, 1970), hence the mean "welding force" for the compact as a whole will be lower, leading to a reduced tensile strength.

From Fig. 6 it can also be seen that the disc compressed at 600 MN m^{-2} has a greater tensile strength than the one prepared at 400 MN m^{-2} .

A plot of tensile strength against compaction pressure for constant thickness and particle size (Fig. 7) was of similar shape to that derived from the transmittance, i.e. the tensile strength increased to a maximum around 600 MN m^{-2} , after which a slight fall occurred.

The greatest tensile strength was obtained with discs prepared from 0.5 g to 71 μm KBr powder. The strength of the discs decreased with increasing particle size, the curves becoming progressively flatter until the weak 550 μm discs showed almost constant tensile strength with increasing compaction pressure. The curvature of the plot for the 1.0 g, 71 μm disc was not so pronounced as that of the 0.5 g, 71 μm . It showed a lower tensile strength, for reasons previously explained, but a maximum was again observed, this time at approximately 900 MN m^{-2} .

The relative density values plotted against compaction pressure followed the same trend as the transmittance and tensile strength curves. This is exemplified by the

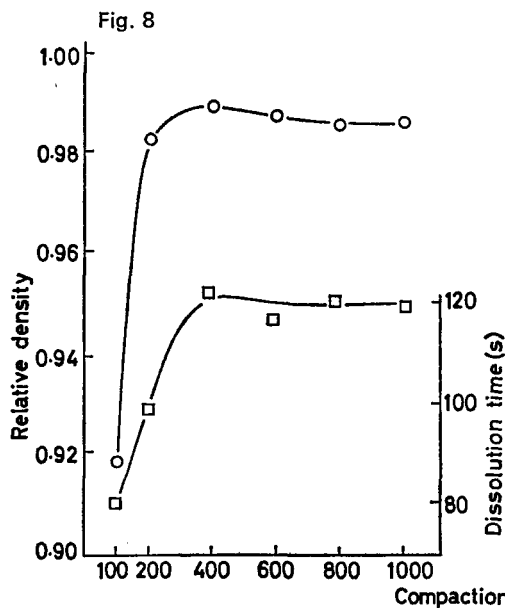


FIG. 8. The variation of relative density and dissolution time with compaction pressure for 0.5 g compacts made with 71 μm mean particle size powder.

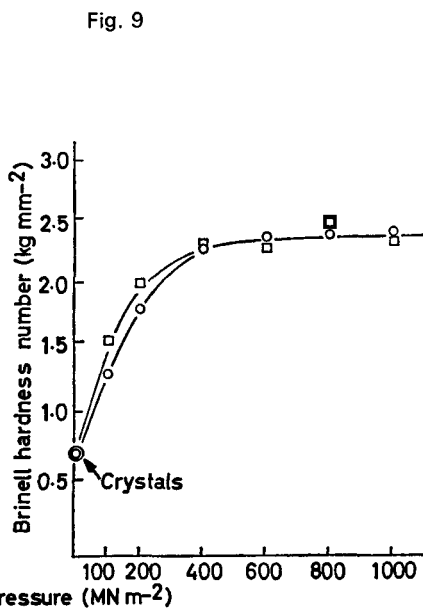


FIG. 9. The variation of Brinell hardness with compaction pressure for 0.5 g compacts made with 71 μm particle size powder. Key: □, face adjacent to moving punch; ○, face adjacent to stationary punch.

curve shown in Fig. 8, representing a 0.5 g compact of 71 μm powder. The relative density reaches a value of 0.99 at 400 MN m^{-2} and then decreases slightly, flattening out towards 1000 MN m^{-2} compaction pressure.

Dissolution times for tablets of the same weight and particle size are also shown in this figure. The dissolution time becomes constant at compaction pressures above 400 MN m^{-2} .

The relation between the mean Brinell hardness of each surface and the compaction pressure is shown in Fig. 9. The value for potassium bromide crystals is also included. The figure shows that initially the surface hardness increased with increasing compaction pressure, the upper surface being harder. However, at 400 MN m^{-2} the Brinell hardness reached a maximum, the upper and lower surfaces then having equal hardness. On increasing the compaction pressure, the hardness of both surfaces remained constant at approximately 2.3 kg mm^{-2} .

Estimates of the elastic moduli of the discs were made from the elastic recovery of the indentation. Results were less reproducible than the estimates of hardness. However, an increase in elasticity was observed, reaching a plateau at about 400 MN m^{-2} , the mean value of elasticity being approximately 120 kg mm^{-2} .

The increase in hardness and elasticity with increasing compaction pressure is thought not to be due to work-hardening but to consolidation of the compact, since the increase ceases once the consolidation is complete at 400 MN m^{-2} . This agrees with earlier conclusions about sodium and potassium chlorides (Ridgway, Glasby & Rosser, 1969).

The effect of increasing compaction pressure can possibly be explained as follows. The high proportion of entrapped air in the compacts prepared at the lowest pressure

(100 MN m⁻²) is shown by their low relative density, which would indicate that many internal surfaces, surrounding the air pockets, are present to scatter the infrared radiation. The transmittance is therefore low and the high voidage leads to a higher dissolution rate, and lower tensile strength and hardness.

Increasing the compaction pressure to around 400–600 MN m⁻² produces a steady increase in tensile strength, hardness, relative density and transmittance corresponding to the initial stage of compaction, where the crystals are being forced into contact and welded together. This would have the effect of reducing the number of surfaces for scattering and at the same time increase the strength of the compact as the binding surface area increases. Air would be extruded, an effect enhanced by the vacuum thus decreasing the voidage of the compact.

This is then followed, at higher compaction pressures, by a slight fall in these properties corresponding to the introduction of flaws into the crystals themselves. This effect would decrease the strength of the compact, increase the number of surfaces for light scattering and thus reduce the transmittance through the disc. The flaws would tend to increase the voidage of the compact. All these effects were observed.

At about 1000 MN m⁻² compaction pressure there was a slight rise in transmittance, tensile strength and relative density, corresponding to the healing of these flaws due to internal pressure welding.

A generally-applicable sequence of events which can occur sequentially during a tableting process has been shown, by following the infrared transmittance, to take place during the compaction of potassium bromide. Unfortunately, the method of following the compaction process reported here is specific to a small number of compounds, mainly the alkali and ammonium halides; some of these are used in pharmacy, e.g. the chlorides, bromides and iodides of ammonia, potassium and sodium, together with lithium chloride and sodium fluoride. It is probable, however, that the event-sequence occurs in the direct compression of many other crystalline materials.

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