RESEARCH PAPERS

A NEW TECHNIQUE FOR INVESTIGATING THE PROCESS OF TABLET COMPRESSION: A PRELIMINARY REPORT

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This preliminary report covers static tests involving the incorporation of an electrical conducting material in a tablet formula and subsequent recording during compression of the change in electrical resistance of the compact. An assessment of the experimental variables has indicated optimum conditions for making dynamic tests on an instrumented tablet machine and suggested more refined adaptations of the technique. Comparisons between this electrical property of the material and other properties such as relative volume, initial granule size, moisture content of the granules and crushing strength of the compact are presented. The resistance plots show evidence of change in the physical condition of the material under pressure; that is an initial packing stage followed by a brief period during which the granular structure is capable of supporting the imposed load without breakdown and finally gradual failure of the granules to give a compact mass.

The behaviour of powdered metals during compression has been expressed in terms of the changing physical condition of the material by Seelig and Wulff (1946) who divided the process into three stages. (i) Packing of the particles. (ii) Elastic and plastic deformation. (iii) Cold working with or without fragmentation. Train (1956) showed that this sequence of events occurred in other powdered materials and Seth (1956) suggested that the formation of pharmaceutical tablets from granules might follow as a result of similar changes.

Jones (1960) referred briefly to the use of electrical conductivity measurements as a means of following the sintering process of powdered metals and Duffield and Grootenhuis (1958–59) described the method in some detail during their studies on the effect of particle size on the sintering of copper powder.

Changes in the properties of compressed materials have also been followed by means of electrical conductivity measurements on the compacts at various pressure levels. Shapiro (1948) used this technique during his investigations on the ageing of silver bromide precipitates and Huffine (1956) made similar experiments with sodium chloride and copper sulphate. In both these instances the authors measured the ionic conductivities of the salts and Huffine concluded that the method was of limited value because of the sensitivity of ionic conductance to certain properties of the specimen. In addition, these investigations were made under essentially static conditions such as are not encountered in the manufacture of tablets.

An attempt has been made to develop a technique which might subsequently yield useful information under operating conditions as near as possible to those of the normal tabletting process. This report is an account of the initial work, carried out under static conditions, to determine the significance of certain experimental variables and the optimum conditions for translation to an instrumented tablet machine.

Experimental

Tablet granules were prepared incorporating a small amount of carbon in the form of synthetic graphite powder. The change in electrical resistance of the dried granular mass was determined at suitable intervals during its subsequent compression.

Preliminary experiments indicated the need for rigid control in the formulation and preparation of the granules, as well as in the methods of compression and recording of change in resistance. Standard techniques were therefore used for each of the above operations. Large variations were found in the conductivity of graphite obtained from different sources and the same batch of material was therefore used for all the experiments.

Formulation and Preparation of the Granules

Because of its known freedom from compression difficulties sodium chloride was chosen as the base material; this was mixed with the graphite in a small end-runner mill, both materials being in powder finer than 350 mesh. Half strength Mucilage of Acacia B.P.C. was then added as granulating fluid and binding agent and a further 5 min. allowed for mixing. The mass was then granulated by hand through a number 22 sieve and dried for 4 hr. at 60°.

The granules produced in this manner were separated into various size ranges by an Inclyno sieve shaker. Except where otherwise indicated the granules were stored over silica-gel in a desiccator for 48 hr. in order to achieve some uniformity of moisture content before tabletting.

A graphite content of 8 to 10 per cent was selected from tests which indicated that this concentration would give a range of resistance readings capable of application to a continuous recorder for use with the instrumented tablet machine. Lower concentrations of graphite gave very high resistance readings which were not reproducible.

Compression

A die fabricated from a non-conducting material was employed so that the electrical resistance of the compact could be measured during compression. From preliminary tests, Tufnol (a paper laminated synthetic resinoid material) was chosen as the most satisfactory material since it combined relative hardness with resilience and a surface which would take a reasonable polish. More recently polyacetal and polycarbonate resins have been tried and they may be equally suitable. A bakelite insert fitted within a metal die, as used by Huffine, was rejected in these experiments because of difficulties encountered when using such an assembly in a tablet machine. The punches used were standard $\frac{1}{4}$ in. flat faced punches from a Manesty hand tablet machine.

100 mg. of the granules was fed into the die and the punch and die assembly (Fig. 1) was placed between the platens of a hydraulic press. The electrical resistance between the faces of the two punches, at each pressure level, was determined using a Cambridge Decade Bridge. The experiments were carried out 10 times for each series of conditions and the experimental points recorded therefore represent the arithmetic mean of 10 readings. Most of the resistance readings, especially those at the high pressure end of the scale, showed little variation (less than 2 per cent). The variation however, was greater at low pressures. Some idea of this variation may be obtained from the standard deviation calculated from 30 measurements obtained at the lowest pressure; that is a pressure gauge reading of 20 p.s.i. The calculated value for the standard deviation was 2,725; the mean resistance reading at this pressure being 12,710 ohms. Despite this variation, the curves from individual sets of readings were always of the same general shape as those shown in the figures.

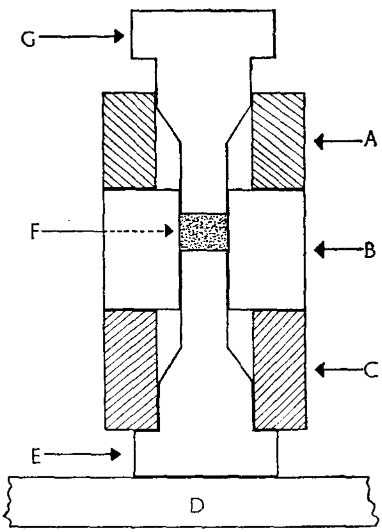


FIG. 1. Diagram of the punch and die assembly. A, Upper punch support. B, Tufnol die. C, Steel support for the die. D, Base plate (insulated from the press platform but carrying the connection to the decade bridge from the lower punch). E, Lower punch. F, Granules in the die. G, Upper punch (connected to the decade bridge).

A compact weight of 100 mg. was chosen as appropriate for a tablet diameter of $\frac{1}{4}$ in. and the applied pressures were selected to include the working range of the tablet machine. Resistance values at pressure gauge readings below 20 p.s.i. were not reproducible and are not therefore included; pressure gauge readings in excess of 600 p.s.i. frequently produced die failure. The pressure readings were obtained from a gauge connected to the main chamber of the hydraulic press and although this method does not indicate the absolute pressure transmitted to the compact it was thought sufficiently accurate for this preliminary study. It is anticipated that strain gauges will be the method employed for determining pressures on the instrumented tablet machine thereby eliminating this approximation.

In certain of the tests the relative displacement of the two punches at each pressure level was measured using a Pye cathetometer so that the volume of the compact might be calculated.

RESULTS

Fig. 2 shows the results of compressing 100 mg. of 25 on 30 mesh granules containing 8 per cent of graphite up to pressure gauge readings of 640 p.s.i. In the same figure the effect of variation in the graphite content is shown by the curves for granules containing 9 per cent and 10 per cent graphite.

Fig. 3 shows the effect of initial granule size on the resistance plot and Fig. 4 the effect of moisture content. The broken line portions of the

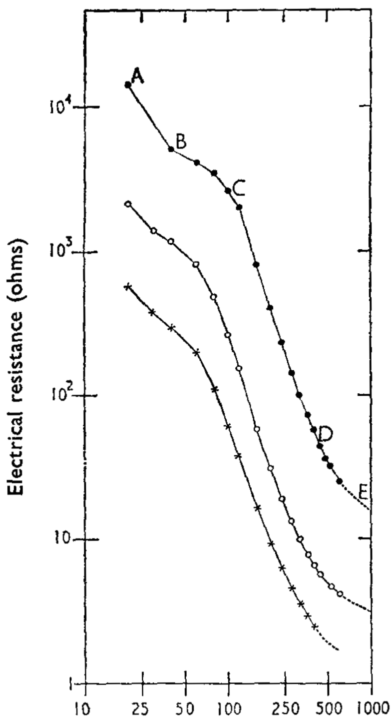


FIG. 2. The relation between compressional force and electrical resistance of a granular mass containing graphite.

8 per cent graphite —●—●—
9 per cent graphite —○—○—
10 per cent graphite —×—×—

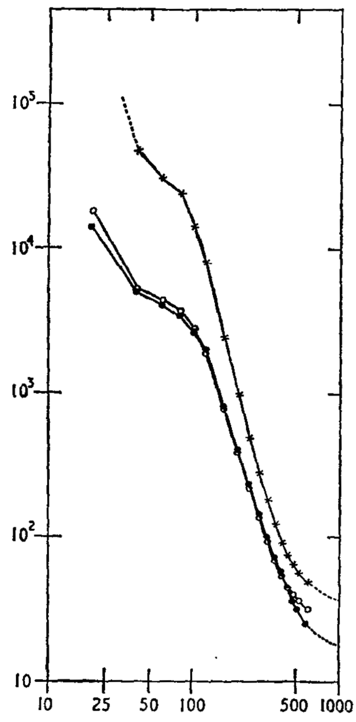


FIG. 3. The relation between compressional force and electrical resistance of granules containing 8 per cent graphite showing the effect of variation in granule size.

25 on 30 mesh granules —●—●—
52 on 60 mesh granules —○—○—
120 on 240 mesh granules —×—×—

curves represent isolated readings taken with compressional force readings up to 1,000 p.s.i. It was impossible to obtain a full set of results in this region due to die failure at high pressures.

The crushing strength of compacts compressed to various pressure gauge readings was determined by means of a Strong-Cobb hardness tester and the results obtained are illustrated in Fig. 6.

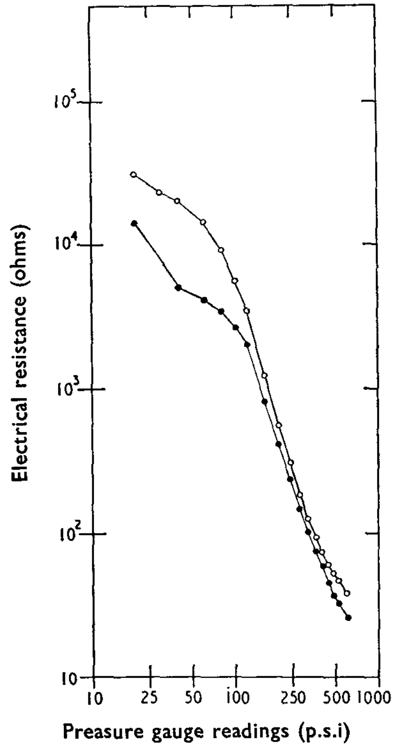


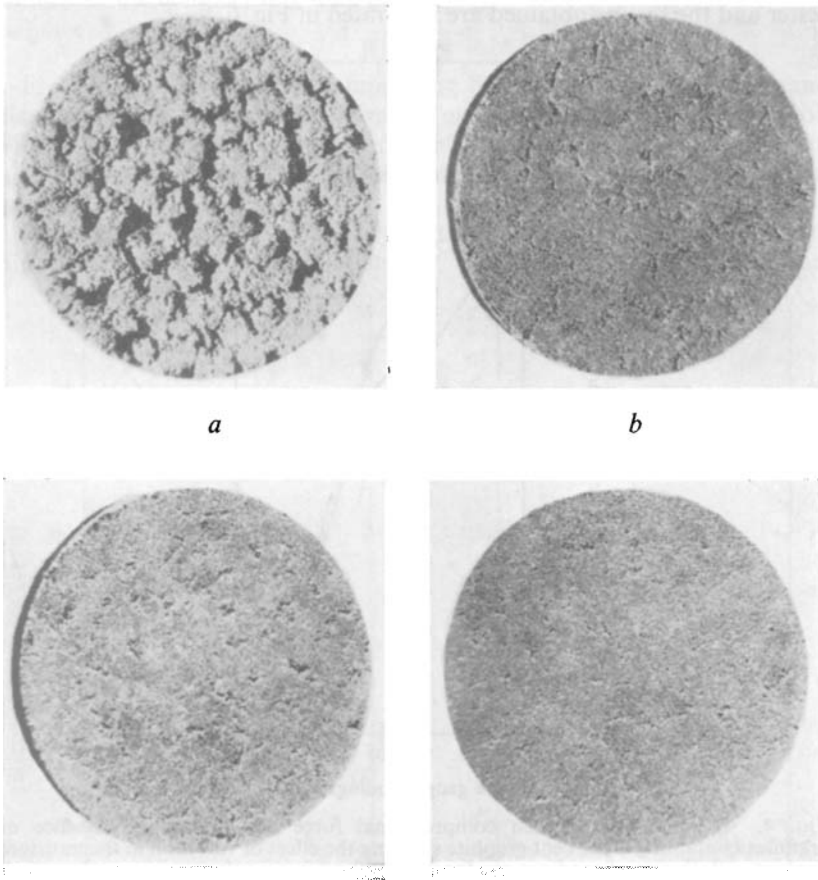
FIG. 4. The relation between compressional force and electrical resistance of granules containing 8 per cent graphite showing the effect of variation in the moisture content. Both sets of readings are for 25 on 30 mesh granules.

Air dry. —○—○—
Desiccated for 48 hr. —●—●—

DISCUSSION

The curves in Fig. 2 can be divided into sections. These, it is suggested, correspond to the stages in the change of the physical nature of a material under pressure referred to in the introduction. The findings of Shotton and Ganderton (1960) do not indicate a sharp demarcation between the various stages and this is in agreement with the results now reported.

It may be postulated that when pressure is first applied, the granules (which are loosely touching) move relative to one another to take up a more compact arrangement and thereby increase the number of inter-granular contact points. Due to the applied pressure, the contacts will be of a more effective nature, i.e. they will possess a lower electrical resistance. It may be the improved inter-granular contact which chiefly



a b c d

FIG. 5. The surface appearance of tablets after being subjected to the following compressional forces.

(*a*) 25 pounds per square inch.
(*b*) 100 ,, ,, ,, ,,
(*c*) 200 ,, ,, ,, ,,
(*d*) 400 ,, ,, ,, ,,

all at a magnification of $7\frac{1}{2}$ times.
Graphite content 8 per cent, granule size 25 on 30 mesh.

accounts for the marked fall in resistance (region A to B in Fig. 2). Train (1956) reported that at all stages of compression a skin was formed where the compacts had been in contact with the die walls. Though no evidence of this was seen at low pressures in the present investigation, it is possible that the fall in resistance may be due to such a skin layer giving a preferential conducting path.

From B to C the granules are probably incapable of tighter packing without deformation but are strong enough to support the imposed load. The electrical resistance will therefore undergo only a small change due to further lowering of the inter-granular contact resistances and the onset of the third stage in localised areas of the compact. During stage 3, the granular structure fails, contact between the individual particles within the granules will therefore be improved and the intra-granular contact resistances will be reduced. This effect would account for the steady fall in electrical resistance in the region C to D in Fig. 2.

Finally with complete breakdown of the granular structure the maximum conducting effect of the graphite content will be approached and from point D onwards increase in pressure will produce less and less effect on the resistivity of the compact.

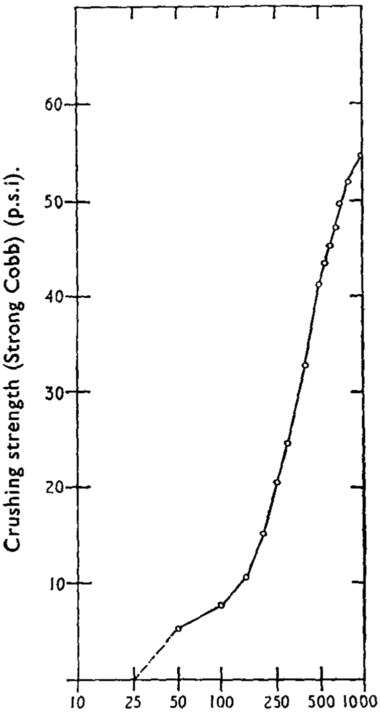
The above interpretation of the curves in Fig. 2 is supported by the changes in appearance of the tablet surface seen in Fig. 5 and by changes in the crushing strength of the compacts (Fig. 6). At pressure gauge readings up to about 25 p.s.i. the compacts possessed virtually no mechanical strength and the individual granules were clearly visible in a surface view of the tablet (Fig. 5a). Higuchi, Rao, Busse and Swintosky (1953) suggested an initial breakdown of granular structure, but it was found in the present work that granules retained their identity (i.e. could be recovered from the die apparently unchanged) up to pressure gauge readings of 20 p.s.i.

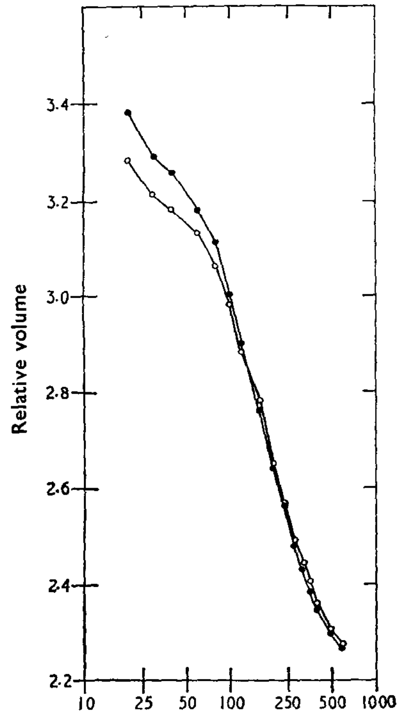
At pressure gauge readings between 100 and 500 p.s.i. the crushing strength of the tablet increased uniformly with increase in pressure and over this range the granular structure as shown by surface appearance (Fig. 5b, c and d) gradually disappeared. At pressure gauge readings above 500 p.s.i. the rate of increase in mechanical strength began to fall off and the surface of the tablet showed no trace of a granular structure.

The change in relative volume of the compacts with increasing pressure (Fig. 7) followed a similar curve to the resistance plot suggesting further evidence for the above argument. Relative volumes were calculated from the formula defined by Walker (1923) and used by Train (1956).

Although the results appear to favour this reasoning, further experimental work is desirable before the alternative explanation of preferential conduction via a skin layer can be excluded.

The curves showing the effect on the resistance of variation in the moisture content of the granules are confusing and several factors may be involved. The presence of moisture might be expected to affect the resistance readings irrespective of any action on the granule materials. Savage (1948) showed that while moisture is essential for graphite to act as an efficient lubricant, only a very low concentration was necessary. The lubrication effect is unlikely therefore to be a significant factor in the present work. It seems more feasible to suggest that, since the binding agent used was a gel, excessive desiccation might reduce its bonding properties to such an extent that the granular structure failed more readily under compression. This would explain the lower resistance readings obtained with desiccated material. A more detailed study of this aspect of the problem with more accurate control and assessment of the moisture content is necessary.





Pressure gauge reading (p.s.i.)

FIG. 6. The relationship between compressional force used and the crushing strength of tablets (Strong-Cobb). Graphite content 8 per cent, granule size 25 on 30 mesh.

FIG. 7. The relationship between compressional force and the relative volume of tablets containing 8 per cent graphite.
25 on 30 mesh granules —●—●—
120 on 240 mesh granules —○—○—

It might be argued that the presence of graphite in the formulation would seriously affect the behaviour of the granular mass since it is known to possess marked lubricant properties. Train and Hersey (1960) have described experiments from which they conclude that the mechanism of lubrication by graphite and talc is somewhat akin to a roller-bearing action; they further deduce that under high compression this lubricant action is seriously curtailed.

To investigate the significance of the graphite in the present work, granules were prepared in which it was substituted by wood charcoal. Unfortunately a resistance range similar to that of graphite could only be obtained with 100 per cent wood charcoal, that is, by omitting the sodium chloride completely. It was observed that these charcoal granules possessed very little mechanical strength and did not bond together to form a hard compact when compressed. In view of this it is not surprising that the resistance plot of the granules followed a curve similar to that obtained for 120 on 240 mesh granules of graphite (Fig. 2), though a strict comparison is hardly justifiable.

Granules containing powdered metals were also prepared but here again a higher concentration was found necessary before resistance readings within a useful range could be obtained. A further difficulty was chemical reaction at the metal surface which seriously impaired the conductivity. It is worth noting that, when powdered tin was used in a concentration of 40 per cent, the resistance readings remained above 1,000,000 ohms until the pressure readings reached a value of about 140 p.s.i.; at this point the resistance fell instantaneously to a value of 100 to 200 ohms and further increase in pressure soon reduced the value to less than one ohm. The most likely explanation for this phenomenon is that flow of the metal component occurred in addition to sudden breakdown of the granular structure. A similar result was recorded when compressing granules made from Mercury with Chalk B.P. 1948, though the sudden fall in resistance occurred at a lower pressure (approximately 50 p.s.i.). It would seem that graphite possesses a rather unique combination of properties which render it especially suitable for this type of investigation.

At this stage no attempt has been made to derive quantitative relationships between the variables involved in these experiments but it is anticipated that more refined techniques will offer an alternative method for obtaining information continuously during the normal operation of a tablet machine.

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