# THE STRENGTH OF COMPRESSED TABLETS

## III. The Relation of Particle Size, Bonding and Capping in Tablets of Sodium Chloride, Aspirin and Hexamine

By E. Shotton and D. Ganderton

*From the Department of Pharmaceutics, School of Pharmacy, University of London, 29–39 Brunswick Square, London, W.C.1*

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Examination of the fracture of tablets in a strength test has revealed two types of failure. Where the interparticulate bond is strong, fracture occurs across the grains and the strength of tablet is a simple function of the particle size. If the interparticulate bond is weak, fracture occurs around the grain and particle size has little effect. A thin coating of stearic acid was used to weaken the interparticulate bond when the effect of grain size on strength disappeared or was reversed. The relation of bonding and capping is discussed and it is shown that capping can be reduced by weakening the interparticulate bond.

Powdered materials can be aggregated by pressure so that the individual particles bond together. The strength of the resultant tablet depends upon a number of factors, one of which is the initial particle size of the material. This factor has been investigated by Nelson (1957), who used an empirical cleavage test on the tablet, and by Forlano and Chavkin (1960), who used disintegration and tumbling tests to assess their results.

To account for the great discrepancy between the practical and theoretical strengths of materials, Griffith (1920) postulated that stress concentrations occurred at flaws in the solid which were proportional to a function of the flaw length. This concept was developed by Orewan (1949) who suggested that the strength ($S_0$) of a brittle, polycrystalline material is inversely proportional to the square root of the mean grain size (L).

$$S_0 = KL^{-\frac{1}{2}}, \text{ where K is a constant} \quad .. \quad .. \quad (1)$$

It was assumed that (i) failure occurred at a flaw within the grain, (ii) that the grain boundary would resist the propagation of the failure to adjacent grains and that (iii) the stress required to overcome this resistance was inversely proportional to the square root of the grain size. With the introduction of the dislocation theory of the deformation of crystals (Taylor, 1934), this concept was modified by Petch (1953), who proposed the relation:

$$S_0 = S_1 + KL^{-\frac{1}{2}} \quad .. \quad .. \quad .. \quad (2)$$

where $S_1$ is an expression of the applied stress required to move a dislocation along a glide plane. These concepts were successfully applied to various systems; to iron by Petch and to certain ceramic materials by Knudsen (1959).

This paper describes investigations of the possible application of these hypotheses to pharmaceutical tabletting systems and aspirin, sodium chloride and hexamine were chosen because of their differing physical properties.

TABLE I

SUMMARY OF THE COMPRESSIVE FORCES IN THE PREPARATION OF TABLETS FROM DIFFERENT SIZE FRACTIONS OF SODIUM CHLORIDE, ASPIRIN AND HEXAMINE

| Mesh | Applied pressure (kg. cm.$^{-2}$) $P_a$ | Transmitted pressure (kg. cm.$^{-2}$) $P_b$ | Mean compaction pressure $P_m = \frac{P_a + P_b}{2}$ kg. cm.$^{-2}$ | $\frac{P_b}{P_a}$ | Ejection force $F_e$ (kg.) | Porosity per cent | Applied force—transmitted force $F_a$–$F_b$ (kg.) | Weight of tablet W (g.) |
|---|---|---|---|---|---|---|---|---|
| *Sodium chloride* | | | | | | | | |
| 30–40 | 809 | 683 | 746 | 0·844 | 69 | 15·9 | 162 | 1·0952 |
| 40–60 | 840 | 703 | 772 | 0·837 | 74 | 16·0 | 176 | 1·0944 |
| 60–80 | 855 | 712 | 784 | 0·833 | 76 | 16·5 | 183 | 1·0894 |
| –80 | 867 | 723 | 795 | 0·834 | 77 | 16·9 | 185 | 1·0856 |
| *Aspirin* | | | | | | | | |
| 20–30 | 946 | 666 | 806 | 0·704 | — | 4·5 | 361 | 0·7071 |
| 30–40 | 939 | 656 | 798 | 0·699 | — | 4·6 | 363 | 0·7062 |
| 40–60 | 963 | 661 | 812 | 0·686 | — | 4·9 | 388 | 0·7066 |
| 60–80 | 981 | 663 | 822 | 0·676 | — | 5·2 | 408 | 0·7065 |
| *Hexamine* | | | | | | | | |
| 20–30 | 810 | 622 | 689 | 0·768 | 98 | 4·8 | 241 | 0·6611 |
| 30–40 | 825 | 628 | 727 | 0·761 | 108 | 5·1 | 253 | 0·6611 |
| 40–60 | 838 | 625 | 732 | 0·746 | 113 | 5·1 | 273 | 0·6615 |
| 60–80 | 845 | 624 | 735 | 0·738 | 113 | 5·4 | 284 | 0·6592 |

All values are the mean of six results. For any material, the machine setting is constant.

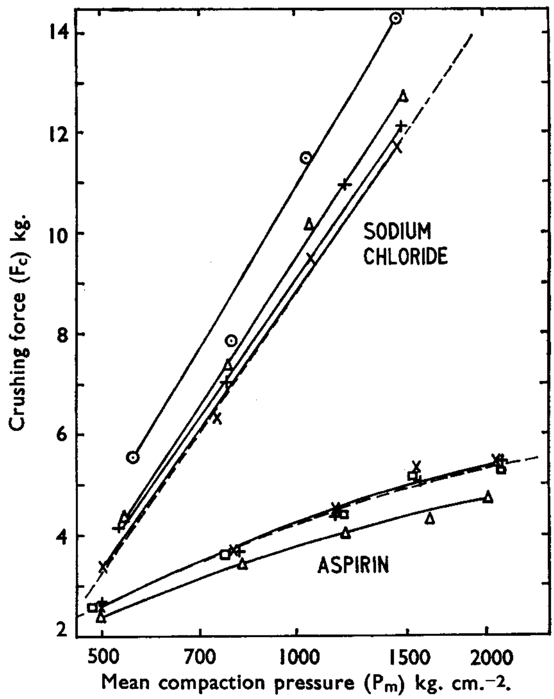


FIG. 1. Relation between mean compaction pressure ($P_m$) and crushing ($F_c$) for size fractions of sodium chloride and aspirin. Broken lines indicate results from a hopper-fed, mechanically driven series (30–40 mesh).

□ 20–30 mesh
× 30–40 mesh
+ 40–60 mesh
△ 60–80 mesh
○ –80 mesh

## EXPERIMENTAL AND RESULTS

Using British Standard sieves, sized fractions of sodium chloride, hexamine and coarse crystalline aspirin were separated and collected. The aspirin and sodium chloride were not further treated but the hexamine was dried at 55° and 2 cm. Hg absolute for 2 hr. before compression. This gave a strongly bonding, reproducible material.

The separate size fractions of each material were compressed on an instrumented tablet machine (Shotton and Ganderton, 1960a) over a pressure range of 500–2,000 kg. cm.$^{-2}$ in a $\frac{1}{2}$ in. diameter punch and die set. Mean values of applied pressure, transmitted pressure and ejection force were obtained for the compression of six tablets of each size fraction

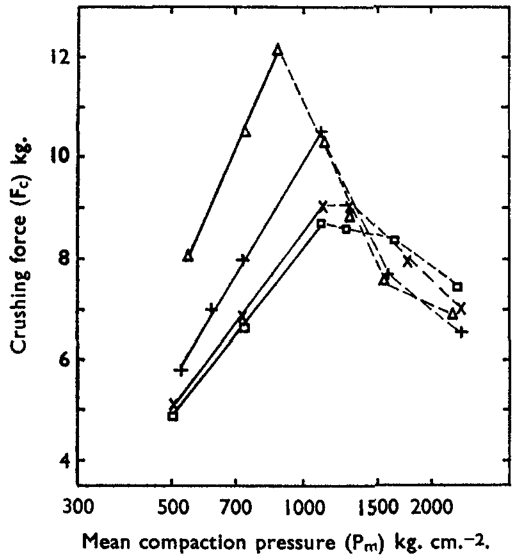


FIG. 2. Relation between mean compaction pressure ($P_m$) and crushing force ($F_c$) for size fractions of hexamine. The broken lines link results obtained from laminated compacts.

□ 20–30 mesh + 40–60 mesh
× 30–40 mesh △ 60–80 mesh

at each pressure level. The materials were fed to the die by hand and compressed by manually turning the machine. The weight of fill was calculated from the density to give a tablet of 0·4 cm. thickness at zero porosity. Each tablet was weighed, its dimensions measured, and finally the strength estimated using the crushing test previously described (Shotton and Ganderton, 1960a). A typical set of results is given in Table I and the relation of crushing strength and mean compaction pressure for the whole series is summarised in Figs. 1 and 2. The results of 30–40 mesh fractions of sodium chloride and aspirin compressed using hopper feed and mechanically driven conditions are superimposed as dotted lines on Fig. 1.

The mean particle size of each fraction was measured microscopically by taking the mean diameter of 800 particles. Some difficulty was encountered with hexamine because of the irregularity of shape and some aggregation. Fig. 3 gives the relation of particle size and strength.

In subsequent experiments, a dilute solution of stearic acid in light petroleum (100–120°) was progressively added to hexamine in a rotating

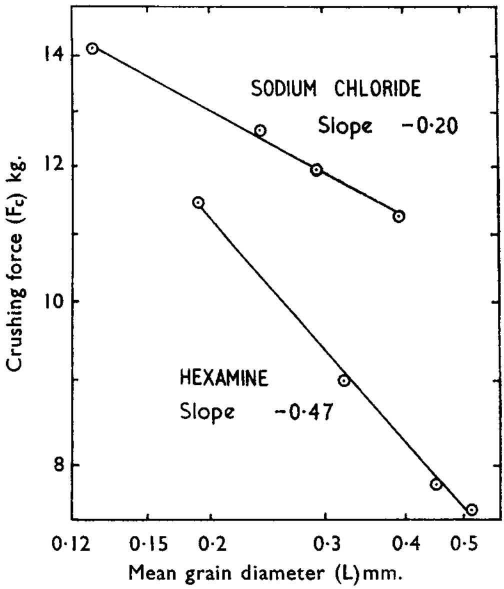


FIG. 3. The effect of grain size on the strength of tablets of sodium chloride and hexamine.

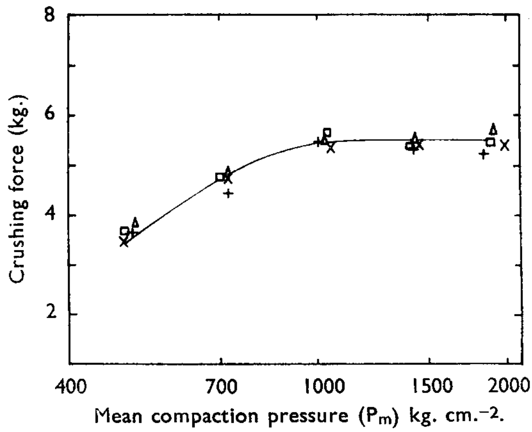


FIG. 4. The relation between the mean compaction pressure ($P_m$) and the crushing force for size fractions of hexamine coated with stearic acid.

| | | | |
|---|---|---|---|
| □ | 20–30 mesh | + | 40–60 mesh |
| × | 30–40 mesh | △ | 60–80 mesh |

coating pan until the dryweight percentage of stearic acid was 0·2. The particulate material was dried in an oven for 1 hr. at 60°, resieved and compressed using the same weight of fill as for the uncoated material. Sodium chloride was similarly coated and compressed. The results are presented in Figs. 4 and 5. Subsequently, a fraction of the hexamine was stripped of stearic acid using light petroleum, dried under vacuum as before, and recompressed. The results did not differ significantly from an untreated fraction of the same size range.

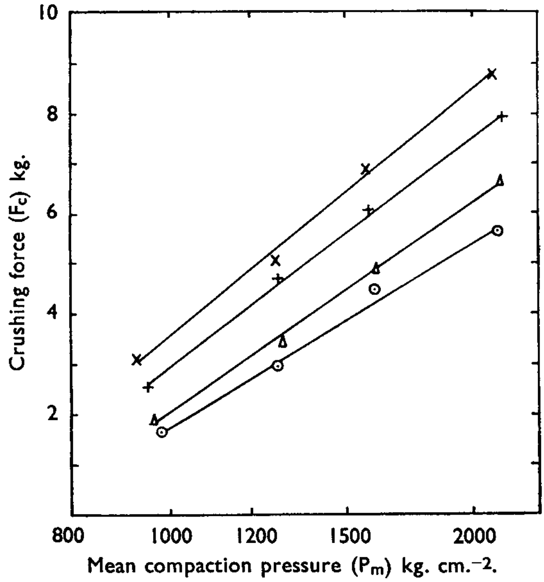


FIG. 5. The relation between the mean compaction pressure ($P_m$) and the crushing force ($F_c$) for size fractions of sodium chloride coated with stearic acid.

× 30–40 mesh    △ 60–80 mesh
+ 40–60 mesh    ○ −80 mesh

Hexamine, which in the first experimental series had shown marked capping and lamination, was now compressed in a die used for compressing discs under vacuum for spectrophotometric analysis. A 30–40 mesh size fraction was compressed under both normal and evacuated conditions using a hydraulic press. In the latter series a vacuum of 0·6 mm. Hg absolute was applied under standard conditions before compression and maintained during the application of the load. The dimensions of these compacts were measured and the crushing strength estimated. The results are presented in Fig. 6.

## DISCUSSION

The increase in ejection force and force lost to the die wall (the difference between applied and transmitted forces), with decrease in particle size is shown in Table I. Since these forces derive from the shear of the area in proximity to the die wall during compression or ejection, this increase must be due either to (*a*) an increase in total contact area with the die wall or (*b*) an increase in the effective shear strength. The former seems unlikely since, as Table I also shows, the voidage increases with decrease in particle size. However, the contact area will be composed of smaller areas intersected with pores and grain boundaries. The size of these individual areas will decrease with particle size and this will influence the shear strength in a manner indicated by Orewan's theory. The work of Train and Hersey (1960) and Long (1960) is a significant contribution to the theory of die-wall behaviour but insufficient is known about the actual

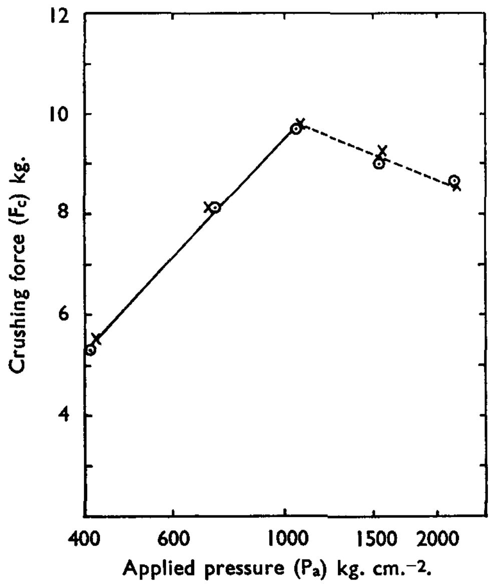


FIG. 6. The compression of hexamine under normal and evacuated conditions. The broken lines link results obtained from laminated compacts. (× normal, O evacuated.)

area in contact with the die wall and the change in the distribution of forces within the die with variation of particle size for the further elucidation of these effects.

The close agreement shown in Fig. 1 between the strength of tablets produced by a hand and a motor driven machine suggest that variation in the speed of compression inherent in the former method is not significant in the present work. In Figs. 1 and 2 the particle size of hexamine and sodium chloride affects the tablet strength in a manner indicated by Orewan's theory. With aspirin, however, the grain size has no effect except in the instance of the finest powder, where the strength of the tablet is lower. Examination of the tablet fracture under the microscope shows two types of behaviour. With sodium chloride and hexamine failure takes place to some extent across rather than round the grain, indicating a strong interparticulate bond. The extent of cross-grained failure increases as the applied pressure increases. A similar transition has previously been shown (Shotton and Ganderton, 1960b) in a sucrose granulation. Aspirin tablets break round the grain indicating a weak interparticulate bond which yields before failure occurs across the crystal. Under these conditions, grain boundary resistance will not be effective and the strength will be independent of it. The lower strength of the aspirin tablets produced from the finest fraction may be due to a weakening effect of entrapped air. Long and Alderton (1960) have shown that, with some materials, this effect becomes more important as the particle size is reduced.

The relation of grain size and tablet strength is given in Fig. 3. For hexamine, the slope is $-0{\cdot}47$. This is in very close agreement with the equation (1) where $S_0$ is measured by the crushing force, $F_c$. For sodium chloride, the slope is $-0{\cdot}2$. However the data for both sodium chloride and hexamine satisfies the equation (2).

The difference in the behaviour of sodium chloride and hexamine on the one hand and aspirin on the other derives from the strength of the interparticulate bond and its effect on failure. Thus, grain size effects should entirely disappear if the interparticulate bond is sufficiently weakened. This weakening was produced in sodium chloride and hexamine systems using a thin coat of stearic acid, and Fig. 4 shows that, with a coated batch of hexamine, the strength of the tablet is independent of the grain size. The argument is further supported by an examination of the fractures which showed that failure occurred around rather than across the grain.

In contrast to coated hexamine, sodium chloride coated with stearic acid produced the strongest tablets from the coarsest fraction; a virtual inversion of the effect described above. However, the sodium chloride crystal is stronger than that of hexamine and is cubic, thus presenting relatively sharp edges. In compression, the load per particle/particle contact will increase with increase in particle size. Thus, in a coarser fraction there will be a greater tendency to penetrate the interparticulate film of stearic acid to form strong welds and so a stronger tablet.

*The Capping of Hexamine*

At a pressure of about 1100 kg. cm.$^{-2}$ hexamine tablets cap and laminate, an effect which then increases progressively with increase in pressure. Although the strength test on a laminated compact must be regarded with caution, the results in Fig. 2 indicate that the disruptive processes are more marked in the case of finer powders despite their higher intrinsic particulate strength. A practical outcome of the results is that, in the production of a hexamine tablet of acceptable strength, capping can be reduced and the effective strength maintained by reducing both the particle size and the applied pressure.

The results given in Fig. 6 for the compression of hexamine under normal and evacuated conditions show identical strength and capping characteristics. We can assume, therefore, that entrapped air is not responsible for capping in this instance. This assumption is further justified by the elimination of capping in the hexamine coated with stearic acid. The geometry of the deforming system will be roughly the same and, if entrapped air was the causative agent, capping should still occur. Although the theories of capping proposed by Train (1956) and Long (1960) differ in fundamental respects, both agree that capping occurs through stresses produced by compression. Train considered that capping occurred on ejection because part of the tablet underwent radial recovery while the remainder was still confined by the die. Long considered that the stress pattern produced by radial forces after the withdrawal of the punches was the mechanism involved.

The variations in the behaviour of the materials studied may be explained by the following mechanisms. Sodium chloride, which produced strongly bonded compacts without capping, can elastically or plastically accommodate the strains produced by the recovery following compression and ejection. The minimum porosity attained in the series was 7 per cent and it is possible that these strains were relatively small. Hexamine, which also produced a strongly bonded compact, but with marked lamination, may be presumed to fail under recovery stresses and, because of the strength of the interparticulate bond, failure propagates across grains to produce extensive failure planes and laminae. The pressure required to consolidate the material completely was certainly exceeded and the relaxation was therefore large. Aspirin compressed to an equal degree, showed no tendency to cap. Because of the mechanical weakness of the interparticulate bond, the failure of an individual particle is not transmitted to neighbouring particles. Stresses are dissipated by a partial separation of the particles and extensive zones of failure are not produced. Thus, a weakening of the interparticulate bond, demonstrated by a round-grain fracture should reduce capping tendencies. This is clearly shown in Fig. 4 where capping in hexamine is eliminated by a thin coat of stearic acid. The introduction of stearic acid weakens the interparticulate bond to give a compacting system, similar to that of aspirin, in which failure occurs around the constituent particles; grain size effects and capping are absent and the resultant tablet is mechanically weak. Finally it is of interest to note that in a preliminary experiment, similar effects were found with a 30–40 mesh hexamine coated with 0·2 per cent hard paraffin.

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The paper was presented by MR. GANDERTON. The following points were made in the discussion.

When lubrication with stearic acid was confined to the die wall the strength of the tablets obtained was the same as when no stearic acid was present. Tablets were tested within minutes of compression, and the interval was kept constant. There was slight attrition of the corners of the crystals, particularly cubic ones. Grain size was identified with particle size. Porosity of the tablets had been inferred from the density of the material, and the volume occupied by the tablets after compression