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A Study of the Compaction Mechanism of Granulated Materials

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Abstract

Using model granules produced from pharmaceutical powders, a mechanism for their compaction was established which was then applied to ceramic materials. Different granulation techniques, for example, spray drying, slugging and agitating granulation, were used to produce granules with different properties. Basic characteristics such as granule size and size distribution, granule shape, specific surface area and apparent specific volume were measured. It was found that, in general, the strength of the compacts formed from the model granules was higher for the granules with higher apparent specific volume. Compacts with higher strength were formed from granules with low specific surface area and large pores while granules with high specific surface area and small pores formed low strength compacts.

In this paper, a mechanism for compaction is established and the compaction behaviour of ceramic materials is considered. © 1996 Elsevier Science Limited.

Introduction

The compaction of ceramic powders and granules has not received the same degree of attention as the more novel and sophisticated methods for producing high performance ceramics for advanced applications. Green compact studies generally emphasise the role of binders and plasticiser on compact strength rather than relating the characteristics of the powder particles or granules themselves.[1,2] Given the emphasis on pressureless sintering as one of the few economically viable methods of mass-producing ceramic components, the refinement of processing conditions such as compact formation has not received due attention.

On the other hand, compaction behaviour is pivotal to tablet production in the pharmaceutical industry and the shaping and forming of compacts has dominated the research carried out in this area for the last ten to fifteen years. It is therefore beneficial to look to pharmaceutical materials to establish a mechanism for compaction, to then compare the compaction behaviour of ceramic materials and to apply the established mechanism.

To date, no one mechanism for compaction has been universally agreed. Likewise, no single granule or powder property such as size or shape has been identified as directly affecting the compaction of all materials. Instead, studies of individual materials have led to conflicting theories. Evidently, factors which influence the compaction of one particular material do not necessarily apply to all. For example, McKenna and McCafferty[3] found that particle size variation had a marked effect on the tensile strength of spray-dried granules. Hüttenrauch[4] found from various experiments that smaller particles gave stronger tablets, as a general rule. He put forward the activation theory that small particles usually lead to large friction during compression causing greater activation of particles. This activation theory has not found general use. It applies to some materials but not to others. Wallace *et al.*,[5] despite a clear difference in particle size, did not observe a significant difference in tablet strength. Also the results of Pesonen and Paronen[6,7] disagreed with the concept that smaller particles produce stronger tablets. Similar contradictions occur throughout the work carried out to date and are in no way aided by the vagueness associated with the terms compression and compaction. These terms are used interchangeably. Cooper and Eaton[8] referred to 'compaction mechanisms' for ceramic powders and derived a mathematical relationship generally known as the Cooper–Eaton equation which referred to the volume reduction occurring at a given pressure. However, Heckel[9] used the term compaction to describe compact strengthening rather than volume

reduction. For the purpose of clarity, compaction and compression as used in this paper are defined as follows:

Compression is 'the reduction in volume of a powder bed under a specified pressure'.[10] It was proposed[11] that the term 'compressibility' should be used to indicate the extent to which the density of a powder is increased by a given pressure. Thus the compression ratio is generally defined as a ratio of the compact density obtained at a given pressure to the apparent density of the loose powder.[9]

The term compaction is used to describe the process of pressing a powder in a die to produce a compact.[12] More specifically, compaction is defined as 'the increase in mechanical strength of a compact under a specified pressure.' [10] It is a technique of size enlargement of small particles by compressing them into a coherent mass.[13] Compaction involves consolidation, size reduction, plastic deformation and fusion occurring in series or simultaneously, depending on the pressures used for compaction and on the material being compacted. Therefore, compactibility is a measure of the minimum pressure needed to produce a given green strength.[11]

The overall purpose of this study was to establish a method of predicting the compaction of any granulate by the measurement of a granulate property (or properties).

Experimental Procedures

Pharmaceutical granules

Model granules were produced by agitating granulation using mixtures of two well-known pharmaceutical excipients with very diverse properties. The predominant compaction process for microcrystalline cellulose is plastic flow and it has a high moisture-absorbing capacity.[14] Lactose has been found to consolidate by brittle fracture[3,15,16] and its compactibility has been found to remain unaffected by moisture uptake.[17] Granules of mixtures of microcrystalline cellulose (mcc):lactose were formed in ratios of 100:0, 80:20, 60:40, 40:60, 20:80 and 0:100. The moisture content, average particle size, apparent specific volume and fragmentation of the granules were measured.

The percentage moisture content of the powders was measured using an infrared detector, Kett FD-220. This instrument calculates moisture content by measuring the amount of water vapour present in the atmosphere above 2–3 g of the sample while it is being heated.

Particle size distribution was estimated from the weight fractions of granules on a series of sieves. The average particle size and deviations were calculated from normal distribution paper (Cumulative Weight Percent versus Log Particle Size).

Apparent specific volumes were calculated from measurements of the unpacked volume of 25 g of each granulate. The granulate was poured into a glass tube which was standing, self-supported, in a graduated cylinder of greater diameter. To ensure that the entire 25 g was transferred to the glass tube, a rubber funnel was attached to the opening. The tube/funnel was then pulled out slowly from the graduated cylinder, taking care not to touch one with the other. The surface of the powder in the graduated cylinder was lightly swept level with a small brush and a volume reading taken. It was essential that no packing occurred during this procedure which was carried out on a shock-absorbing surface.

The −1410+700 μm fraction of each granulate was measured for fragmentation using a Fudo Industry Co. Rheometer NMR2025. Individual granules were placed on a level platform and force was applied at a rate of 20 mm/min. On deformation or fracture of a granule, a change in the slope in the plot of force versus time occurred. The force corresponding to this change in slope is the granule fragmentation.

Tablets were uniaxially pressed from each granule composition using 0·5 g powder charge. Tablets of each sample were pressed at 5, 15 and 25 kPa. Maximum pressure was maintained for 10 s and five tablets were pressed from each sample.

The 'breaking force' of tablets was measured using a Schleuniger-2E tester in which the tablet was crushed in the axial direction by a moving platen. This instrument measures the exact point at which the tablet fractures and is used extensively in the pharmaceutical industry (to measure the 'hardness' of tablets). As the acceleration and the area of the tablet were constant, tablet breaking force results are quoted in kilograms force [kgf].

The data obtained from the characterisation of the pharmaceutical granules were analysed using multiple regression analysis by eqn (1), to identify the influence of these granule characteristics on tablet breaking force, i.e. the tablet breaking force is the 'response' variable and the granule characteristics are the 'predictor' variables.

$$R = K_1 \cdot f_1(x_1) + K_2 \cdot f_2(x_2) + \ldots\ldots + K_p \cdot f_p(x_p) \quad (1)$$

where R is the correlation coefficient for tablet breaking force, K_1, $K_2 \ldots K_p$ are unknown parameters and $f_p(x_p)$ is the linear function of p predictors (average particle size, average granule fragmentation, apparent specific volume, microcrystalline cellulose content and the amount of binder present). JUSE QCAS/MA1 Software, (Japan) was

employed to calculate R, presented as the correlation coefficient from the predictor variables using eqn (1).

The apparent specific volume (ASV), of pharmaceutical granules was examined in detail. A ternary composition combining microcrystalline cellulose (mcc), lactose and cornstarch was used. This granulate, devised by the Japanese Society of Standard Recipes[18] is used as an excipient for drugs in the pharmaceutical industry. To vary the ASV, different binder types, granulating times and granulating techniques were used. Six granule samples which provided a wide range of ASV were selected for analysis and their characteristics (particle size, granule fragmentation, moisture content, specific surface area and granule density) were measured. Tablets were formed and their breaking force recorded using the procedure outlined above.

Ceramic granules

Talc and pyrophyllite were prepared by spray drying at different temperatures. The characteristics of these granules, particle size and distribution and apparent specific volume were measured as for the model pharmaceutical granules. The specific surface area was measured using a Quantasorb Sorption Analyser (Quantachrome GmbH) which utilises the BET method (adsorbate gas, N_2; carrier gas, He).

Tablets of each ceramic powder were uniaxially pressed using 1·0 g powder charge, at pressures of 20, 35 and 50 kPa. Five tablets were pressed at each pressure. The tablet breaking force was obtained using an Instron tensile tester. As with the Schleuniger-2E tester used for the pharmaceutical materials, the acceleration and tablet size were constant and so the units were again kilograms force [kgf]. This method, the diametral compression test (or Brazilian test),[19] has been recommended for testing ceramic compacts in place of the normal three or four point bend tests which report modulus of rupture. This type of test overcomes the inherent difficulty of tensile testing in the form of stresses from gripping devices and tests the bulk material rather than just the surface.[1]

Results and Discussion

Assessment of pharmaceutical model granules

The moisture content, average particle size, apparent specific volume and fragmentation of the granules is shown in Table 1. There are no obvious trends in granule property changes as the amount of mcc is increased. The forces required to break tablets formed from these granules are shown in Fig. 1. The tablet breaking force increased with increasing pressure. However, because of the number of parameters involved and the volume of data obtained for each granulate, a multiple regression analysis technique was introduced to assess the influence of each measured property of the model granules on their compaction behaviour at different pressing pressures. This software, JUSEQCAS/MA1, was used to calculate correlation coefficient values, R, for each granule property (input). The highest value possible is $R = 1$. Parameters with high R values, i.e. R close to 1, influence the output (tablet breaking force) more than parameters with lower R values. In addition, cumulative R values for all input should be ≈ 1. Low cumulative R values suggest that some input is missing and that some influential parameter has been excluded in the granule evaluation. In the case of the model pharmaceutical granules, cumulative R values close to 0·9 and greater were obtained at each pressing pressure indicating that the most influential parameters had been measured and the granule evaluation carried out was complete.

Correlation coefficient values were high for system-related parameters such as the amount of

Table 1. Model granule evaluation

#	*Sample PH101:200M+binder*	*Moisture content (%)*	*Apparent specific volume (cm^3/g)*	*Granule fragmentation (kgf)*		*Particle size distribution*	
				Mean (kgf)	*Standard deviation*	*Mean (μm)*	*Standard deviation*
1	100:0 + 800 g 3% HPC-L	3·6	1·36	1·38	0·61	638·3	0·16
2	100:0 + 700 g 3% HPC-L	3·6	1·50	1·00	0·64	524·8	0·18
3	80:20 + 650 g 3% HPC-L	4·8	1·30	1·74	0·33	631·0	0·17
4	80:20 + 600 g 3% HPC-L	3·6	1·40	1·60	0·13	582·1	0·17
5	60:40 + 600 g 3% HPC-L	2·2	1·50	1·99	0·35	707·9	0·20
6	60:40 + 550 g 3% HPC-L	2·1	1·40	1·50	0·32	656·1	0·27
7	40:60 + 500 g 3% HPC-L	3·1	1·60	0·97	0·80	595·7	0·21
8	40:60 + 450 g 3% HPC-L	2·6	1·56	1·68	0·35	608·1	0·19
9	20:80 + 350 g 3% HPC-L	3·4	1·84	0·96	0·52	676·1	0·21
10	20:80 + 300 g 3% HPC-L	3·4	1·76	1·11	0·31	758·6	0·29

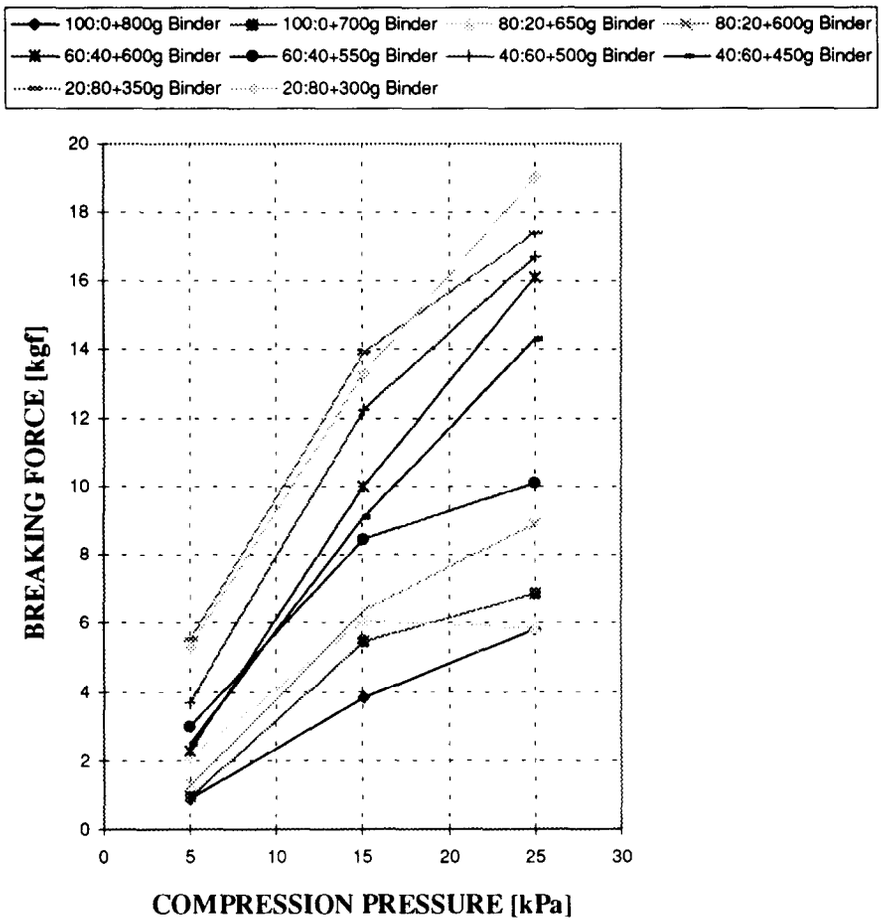

Fig. 1. Compaction of model granules.

mcc:lactose. With increasing amounts of mcc, the tablet breaking force of the compacts decreased. However, of the granule-related parameters such as granule size or apparent specific volume, it was found that the apparent specific volume (ASV) exerted most influence on tablet compaction. Figures 2, 3 and 4 show tablet breaking force plotted against average particle size, granule fragmentation and apparent specific volume, respectively, for tablets which were pressed at 15 kPa pressure. The correlation coefficient obtained for the apparent specific volume, R = 0·85, was much higher than that obtained for either granule fragmentation or particle size (R = 0·241 and R = 0·593, respectively).

Similar results were obtained at 5 and 25 kPa pressing pressures. However, although model granules with a higher ASV formed stronger compacts which required a higher applied force before breakage occurred than those with lower ASV, no direct relationship between the tablet breaking force and the ASV was identified. As this result was for one model system, further investigation and verification was required. Therefore, the ASV and factors affecting ASV (specific surface area, pore volume) were examined in more detail for a standard composition of microcrystalline cellulose, lactose and cornstarch.

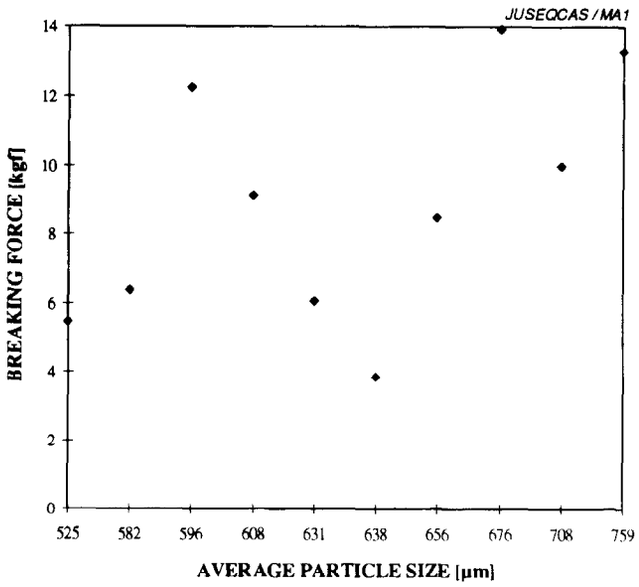

Fig. 2. Average particle size versus tablet breaking force for model pharmaceutical granules, R = 0·593.

Mechanism of compaction

Instead of using model granules of different composition which inevitably influences their compaction behaviour, a series of granulates of the same composition but with different ASV were produced. Figure 5 shows the compaction behaviour of these

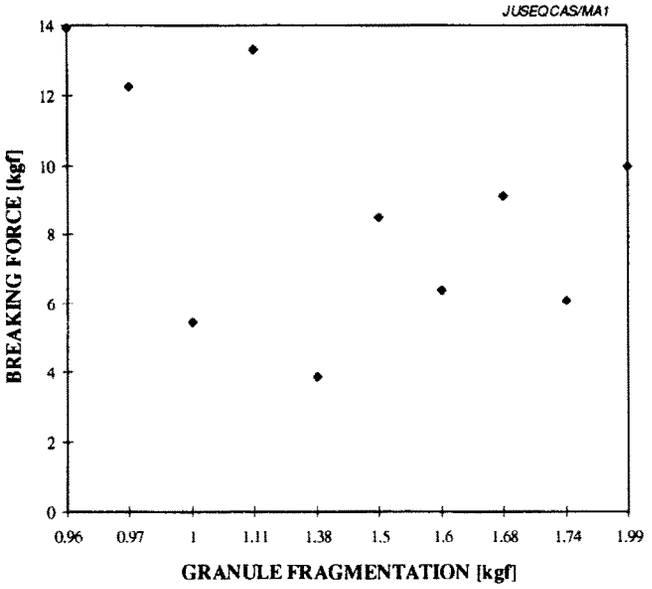

Fig. 3. Granule fragmentation versus tablet breaking force for model pharmaceutical granules, $R = 0{\cdot}241$.

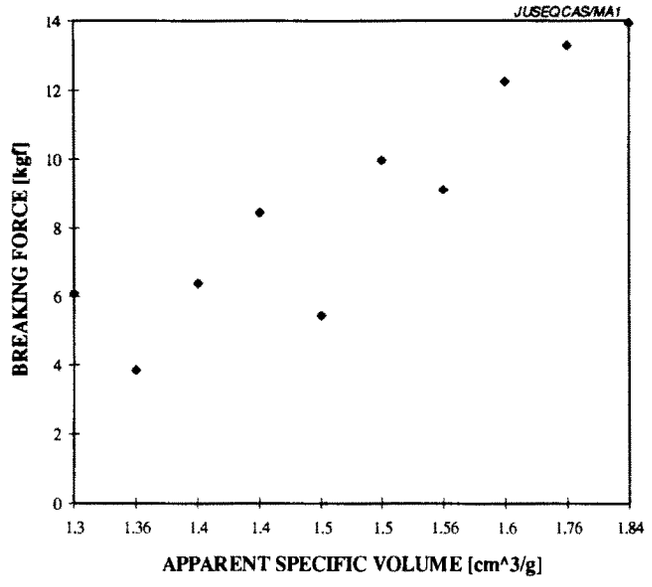

Fig. 4. ASV versus tablet breaking force for model pharmaceutical granules, $R = 0{\cdot}85$.

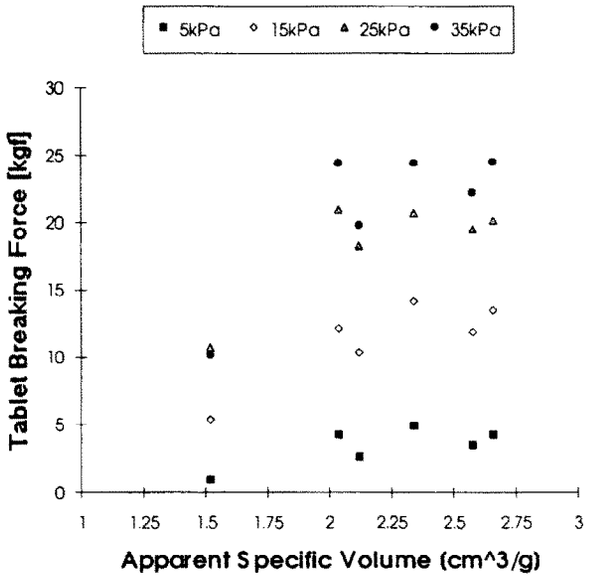

Fig. 5. Compaction behaviour of granules formed from a standard composition with varying ASV.

granules at a range of pressing pressures. Although in general, as with the model granules, the tablet breaking force increased with increasing ASV, the breaking force of some samples decreased. The factors which contribute to the ASV of each granulate, the specific surface area and the pore volume, were therefore measured and from these measurements, the size and number of pores was estimated.[17] It was estimated that granulates with lower tablet breaking force (in Fig. 5) had smaller pores present. The granules of these granulates are made up of particles which are relatively tightly packed together. For samples with higher tablet breaking force, larger pores were present. When pressure is applied resulting in the collapse and break-up of the granules and particles, this looser packing allows more interaction and interlocking between particles compared to the relative inactivity of the tightly packed granules. During compaction, the number of faces/planes available, i.e. the area of true contact,[3,4] which can form the bonds that increase compaction, is much greater when large pores are present than if small pores are present. When large pores collapse and fragment under applied pressure, the area of surface available on which bonding can occur is greater than for the small pores. Regardless of the forces involved for a material (interfacial forces such as liquid bridges, capillary forces, adhesive and cohesive forces or attractive forces[11]) the larger pores lead to more surface interaction and a greater possibility of strengthening of the compact formed.

Ceramic granules

As stated previously, the overall purpose of this study was to establish a method of predicting the compaction of any granulate by the measurement of a granulate property (or properties). For pharmaceutical granules, the ASV can be measured and the degree of compaction can be predicted with a reasonable amount of confidence. For a more accurate understanding, the specific surface area and pore volume are also required. The next stage of this study was to apply the findings of the pharmaceutical work to ceramic granules. To this end, a random selection of granulated ceramic materials was characterised, compacted and their tablet breaking forces were measured.

Two pyrophyllite granulates with largely different properties were compared. Pyrophyllite 1 was produced by sinter-crushing. It is made up of

Table 2. ASV and breaking force of ceramic granules and compacts

Material	*ASV (cm^3/g)*	*Breaking force @ 35 kPa pressing pressure (kgf)*
Pyrophyllite 1	0·84	5·30
Pyrophyllite 2	1·34	24·17
Talc 1	1·72	19·79
Talc 2	1·76	21·25
Talc 3	1·81	26·08
Talc 4	1·83	28·66

individual irregularly shaped particles and from observations using scanning electron microscopy, there were no pores evident. Pyrophyllite 2 on the other hand, was prepared by spray drying, which gave the characteristic spherically shaped granules. Compared with the surface of the sinter-crushed pyrophyllite, the spray-dried pyrophyllite granules were relatively porous. In addition, these spray-dried pyrophyllite granules were found to be hollow. The ASV and tablet breaking forces of these materials at 35 kPa pressing pressure are given in Table 2. The higher the ASV, the greater the degree of compaction obtained at a given pressure. Tablets produced from pyrophyllite 1 required a force of only 5·3 kgf before breakage occurred compared with a force of 24·2 kgf for the spray-dried pyrophyllite. The ASV of pyrophyllite 1 was much lower than that of pyrophyllite 2. As with the pharmaceutical materials, larger pores and hollow-centred granules collapsed during pressing and resulted in the formation of stronger compacts.

Four talc samples, granulated by spray drying at different temperatures, were also compared. Compared to the spray-dried pyrophyllite granulate, these granules were not hollow but composed of layers of flaky particles, similar to a snowball. ASV and tablet breaking forces for these materials are shown in Table 2. Their ASV ranged from 1·72 to 1·83 cm^3/g and it was found that these granulates followed the same trend — for a granulate with a higher ASV, a higher force was required to break tablets produced at a given pressure. The tablet breaking force ranged from 19·79 kgf for compacts produced from spray-dried talc with an ASV of 1·72 cm^3/g to 28·66 kgf for spray-dried talc with an ASV of 1·83 cm^3/g.

For this random selection of ceramic materials the trend established using pharmaceutical materials which links compaction to the ASV can be applied. It is proposed that for a given material/composition, a granulate with a higher ASV should compact to a higher degree than a granulate with lower ASV.

Conclusions

The level of compaction increased with increasing apparent specific volume for granules from a standard pharmaceutical composition. However, the mechanism of compaction was also influenced by the number and size of pores present in the granules. Larger pores led to a greater degree of compaction.

It was found that the compaction behaviour of selected ceramic granules was similar to that of the pharmaceutical materials studied. In general, compaction increased with increasing apparent specific volume but the pore size was also influential in the determination of the extent of compaction. As was found for the pharmaceutical materials, larger pores led to a greater level of compaction.

The apparent specific volume (ASV) can be measured and used to predict the level of compaction of a granulate.

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