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Rapid Communication

The normalization of the solid fraction of particulate compacts

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Summary

Meaningful comparison of the compressibility and compactibility of different powder materials cannot be made using the solid fraction as a yardstick for comparison, due to the different ranges of solid fraction over which different materials consolidate. Consequently, the solid fraction has been normalized as follows: $P = (\Theta_{rc} - \Theta_{rf})/(1 - \Theta_{rf})$, where Θ_{rc} is the solid fraction of the compact at a given consolidation state and Θ_{rf} is a reference solid fraction marking the begin of interparticle bond formation. This work evaluates comparatively the appropriateness of the relative tap density, θ_{rT}, (which is the tap density expressed as a ratio of the true density) and the solid fraction, Θ_{ri}, at the point where the punch forces begin to increase and subsequently remain above the noise level of the load transducers, as reference densities. It was deduced that, Θ_{ri} is more appropriate than the tap density as the reference density. In the event where Θ_{ri} cannot be determined, the relative tap density may be used for noncohesive materials; it cannot however be used for cohesive materials. It was also shown that Θ_{ri} is independent of compression speed.

The mechanical properties of a compact are mainly dependent on the degree of interparticle bonds. Consequently, in the evaluation of the compactibility of different powder formulations, it is of utmost importance that the formulations are compared at the same degree of interparticle bonding. It is therefore desirable to have an estimate of the particle-particle area of bonding.

Owing to the difficulty in directly determining the true areas of interparticle contact in a compact, the solid fraction, Θ_r, is commonly used as an indirect measure of the degree of interparticle three-dimensional bonding. However, the solid fraction does not allow meaningful comparison to be made between different powder substances (Holman and Leuenberger, 1991). This is attributable to the fact that the range of solid fraction over which particulate systems consolidate differ for different substances as a result of the different powder bulk densities. Consequently, it is difficult to interpret the mechanical significance of the slopes of the relationship between some mechanical properties (e.g., indentation hardness, tensile strength) and the solid fraction of different powders. Infact it has been shown that the tap density and the slope of the relationship between the indentation hardness and the solid fraction are correlated (Holman and Leuenberger, 1991).

To alleviate this problem, the solid fraction has to be normalized. To this end, it was reasoned that a parameter which expresses the de-

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gree of interparticle bonding as a fraction of the maximum number of bonds which can potentially be formed will prove to be standardized. The maximum number of bonds, $B_{\max}$ (100% particle-particle bonds) that can be formed corresponds to the solid fraction range equal to the difference between unity, i.e., when the compact is fully dense, and a reference solid fraction, Θ_{rf}, at which point bonds begin to form. That is:

$$B_{\max} = 1 - \Theta_{\mathrm{rf}} \tag{1}$$

Likewise, the number of bonds formed at a given consolidation state of a compact, B_{c}, would correspond to the difference between the solid fraction of the compact at this state, Θ_{rc}, and the reference solid fraction. That is:

$$B_{\mathrm{c}} = \Theta_{\mathrm{rc}} - \Theta_{\mathrm{rf}} \tag{2}$$

The fraction of bonds, P, formed at any given solid fraction can thus be expressed as a quotient of B_{c} and $B_{\max}$. That is:

$$P = \frac{\Theta_{\mathrm{rc}} - \Theta_{\mathrm{rf}}}{1 - \Theta_{\mathrm{rf}}} \tag{3}$$

For all substances therefore, $P = 0$ at the reference solid fraction, Θ_{rf}, i.e., $\Theta_{\mathrm{rc}} = \Theta_{\mathrm{rf}}$, and $P = 1$ when the compacts are fully dense, i.e., $\Theta_{\mathrm{rc}} = 1$. The normalized solid fraction, P, has been shown to be a standardized parameter describing three-dimensional interparticle bonding (Holman and Leuenberger, 1991).

Based on the theory that bonds begin to form appreciably after the particles assume a state of closest packing, Holman and Leuenberger (1988, 1989, 1990, 1991) used the relative tap density which is the tap density expressed relative to the true density, as the reference density. Recently, the solid fraction, Θ_{ri}, at which point the upper punch force begins to increase and thereafter remain above the noise level of the load transducer during single-ended compression was taken as the reference point (Holman, 1991a,b). This solid fraction value marks the end of the extensive rearrangement stage and the begin of interparticulate bond formation. The object of this paper is to comparatively evaluate these two reference points, the relative tap density and Θ_{ri}.

The rationale behind the use of the relative tap density, Θ_{rT}, as the reference point has been treated in detail by Holman and Leuenberger (1991). The use of Θ_{ri} as the reference density is based on the following observations. At the start of compression, the punches confine the powder to reduced volumes with negligible force. This stage is followed by a stage where the punch forces repeatedly increase above and decrease to the noise level of the transducers. This is indicative of on-going particle rearrangement whereby columns of particles which are formed in the direction of force application are broken up and then reconstituted (Endersby, 1940). The columns are broken up because some interparticle bonds are disrupted when the direction of force at a contact point deviates from the direction normal to the area of contact to a greater degree than friction allows (Drescher and De Josselin de Jong, 1972). The disruption of even one contact point creates a movement of a great number of particles resulting in the disruption of particle-particle bonds and a reorientation of relative positions of particles (Drescher and De Josselin de Jong, 1972). At some point, the punch forces do not decline to zero but to a measurable force above the noise level of the transducers.

The decline of the upper punch force to zero suggests that most of the bonds previously formed are broken by the reorientation of the particles and an insignificant number of interparticle bonds survive the rearrangement. On the other hand, a decline of the punch force to a measurable value indicates that an appreciable number of bonds survive the rearrangement. Thus, the begin of constructive bond formation (formation of bonds which are not subsequently totally disrupted) is indicated when the upper punch force begins to increase and thereafter remains above the noise level of the load transducers. It should be noted that the solid fraction, Θ_{ri}, corresponding to this point in the compression is higher than the solid fraction at which point a measurable force is first exerted on the punches.

To compare the two reference points, the relative tap density, Θ_{rT}, and Θ_{ri} were determined for the following materials; microcrystalline cellulose (Avicel® PH102) (manufacturer: FMC, Philadelphia, PA, U.S.A.); β-anhydrous lactose (manufacturer: Sheffield, Norwich, NY, U.S.A.); dicalcium phosphate dihydrate (Emcompress®) (manufacturer: Edward Mendell Co., Carmel, NY, U.S.A.); acetaminophen, USP Fine Powder (80% of the particles are smaller than 38 μm), (manufacturer: Penco of Lyndhurst Inc., Lyndhurst, NJ, U.S.A.); starch 1500 (manufacturer: Colorcon, Inc., West Point, PA, U.S.A.).

The powders were conditioned at 40 ± 10% RH for a minimum of 1 week and were passed through a 0.5 mm sieve to break down any agglomerates before being used for the determinations. The tap densities were determined with a tapping machine (Vanderkamp Tap Density Tester, Vankel Industries, Inc., Edison, NJ, U.S.A.). Depending on the bulk volume of the powder, 30–70 g of the powder was weighed into a 100 ml glass cylinder and subjected to a series of cycles of taps, with each cycle of taps comprising 1250 taps, until the change in volume between two consecutive cycles of taps was less than 1 ml. The volume of powder was then read off as the tap volume and used to calculate the tap density.

The solid fraction marking the end of the extensive rearrangement stage, Θ_{ri}, was determined during compression of the powders. The powders were compressed at speeds of 1.5, 15, 30 and 60 rpm on a compaction simulator (Mand Testing Machines Ltd. Stourbridge, West Midlands, U.K.) programmed to mimic a 16 station rotary press as described by Holman (1991a). The solid fraction at which point the upper punch force began to increase and subsequently remained above the noise level of the load transducers was determined from a printout of the punch forces and the corresponding solid fractions of the powders recorded during compression.

The relative tap densities and Θ_{ri} of the powders are listed in Table 1. It is obvious from Table 1 that the solid fraction marking the end of

TABLE 1

Densities of the powder materials used in this study

Substances	Relative densities [a] $\pm$SD (n [b] = 3)		True density [c] (g cm^{-3})	Solid fraction of powder at the end of the extensive rearrangement, Θ_{ri}, during compression at different compression speeds; $\pm$SD (n = 3)			
	Poured	Tap		1.5 rpm	15 rpm	30 rpm	60 rpm
Microcrystalline Cellulose (Avicel® PH102)	0.197 ±0.003	0.286 ±0.004	1.59	0.299 ±0.004	0.298 ±0.006	0.294 ±0.001	0.314 ±0.009
β-Anhydrous lactose	0.356 ±0.000	0.548 ±0.009	1.56	0.530 ±0.009	0.534 ±0.004	0.516 ±0.001	0.536 ±0.008
Dicalcium phosphate dihydrate (Emcompress®)	0.371 ±0.008	0.466 ±0.004	2.36	0.448 ±0.004	0.454 ±0.000	0.440 ±0.007	0.440 ±0.006
Acetaminophen USP Fine	0.265 ±0.018	0.490 ±0.005	1.29	0.666 ±0.011	– [d]	0.638 ±0.010	0.642 ±0.008
Starch 1500	0.381 ±0.004	0.501 ±0.004	1.56	– [d]	– [d]	0.474 ±0.004	– [d]

[a] Poured and tap densities are expressed relative to the true densities.

[b] Number of determinations.

[c] Determined with a helium gas pycnometer (Autopycnometer 1320, Micromeritics, Norcross, GA, U.S.A.).

[d] Value not determined.

the extensive rearrangement, Θ_{ri}, is essentially the same at all speeds of compression. The dependence of the compression behaviour of powders on the compression speed is mainly due to the time-dependent deformation characteristics of the powder particles. Since the particles undergo no deformation or at most undergo time independent elastic deformation at the low pressures (approx. 0.25 MPa) operative at this stage, it is not surprising that Θ_{ri} exhibits an independency of the compression speed.

The relative tap densities of the substances, with the exception of the cohesive acetaminophen, differ from Θ_{ri} in the range of 0.00 to ±0.03. The relative tap density can therefore be considered as being practically the same as Θ_{ri} for all the noncohesive, free flowing materials. The relative tap density of the cohesive acetaminophen, however, is very much lower than Θ_{ri}. The behaviour of the fine (micronized) acetaminophen particles can be explained by the fact that they exist as agglomerates in spite of their being sieved prior to the determinations (the particles immediately agglomerated after being sieved). The particles forming the agglomerates are held together by relatively weak attractive forces. Nevertheless, the forces which are normally operative during tapping were apparently not strong enough to overcome these attractive forces. The basis for rearrangement during tapping is as follows: During tapping the powder particles are forced to jump and lose contact with each other for a moment during which period there is no friction between the particles. This frictionless moment allows the particles to rearrange themselves. Due to the inability of the particles within the agglomerate to experience this frictionless moment during tapping, the rearrangement of the powder only proceeded to an intermediary point and not to the point representing the closest possible packing of the particles. Thus, for powders which spontaneously agglomerate, the tap density must not be used as a measure for the begin of extensive interparticle bond formation, since it does not reflect the closest packing state.

In contrast to the relative tap density, Θ_{ri} of the acetaminophen powder represents the closest possible packing of the particles, since the force applied during compression can overcome the forces holding the particles together in the agglomerates. Hence, whereas the tap density can only be used as a reference point marking the begin of interparticle bond formation for noncohesive, free-flowing materials, Θ_{ri} can be used as a reference point for all powder materials, cohesive as well as noncohesive. Further, the use of Θ_{ri} as opposed to using the relative tap density offers the advantage that the reference solid fraction is determined under the same conditions (die-wall lubrication, die diameter and manner of force application) under which the powders are being compacted. Moreover, the reference solid fraction determined during compression is specific for the singular compression event as well as for the particular powder mass unit being compacted in the die. In contrast, the tap density is determined under conditions different from those used for compaction. For example, the wall of the vessel used for the determination is not lubricated, the diameter of vessel may be different from the die diameter and the manner in which force is applied is different. Besides, not the same powder sample used for the determination of tap density is compressed. It is assumed that the samples used for compression and the determination of tap density are true representatives of the bulk powder and therefore identical in composition and physical characteristics. Such an assumption is not always valid.

It can therefore be concluded that the determination of the reference solid fraction during the compression event is more relevant to the problem and offers many advantages over the tap density. However, in cases where there is no possibility of determining Θ_{ri}, the tap density may be considered as an alternative for noncohesive and free-flowing materials.

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