Fundamental studies of granule consolidation
Part 1: Effects of binder content and binder viscosity

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Abstract

Granule consolidation was studied experimentally using a 0.3 m diameter laboratory granulation drum with fine glass ballotini as the model powder and glycerol–water mixtures as model liquid binders. Granule consolidation during tumbling was found to be a complex process controlled by the balance between the different mechanisms that resist granule deformation: interparticle friction and viscous dissipation. The rate of consolidation decreased with decreasing particle size. As liquid content increased, interparticle friction effects decreased but viscous losses became more significant. Thus, the effects of binder viscosity and liquid content were highly interactive. Unless the balance between the two mechanisms is accurately known for a given system, the effect of changes to binder parameters on granulation behaviour cannot be predicted, even qualitatively. To overcome these difficulties a new methodology for relating formulation properties to granulation behaviour is suggested based on bulk powder properties measured by triaxial consolidation tests and the development of a new granulation criterion for deformable granules. A procedure for testing critically the proposed methodology is presented.

Keywords: Granule behaviour; Interparticle friction; Viscous dissipation; Binder parameters; Granule deformation

1. Introduction

Granulation (also called pelletization, balling or agglomeration) is a size enlargement process that produces granules with controlled properties from liquid or particulate feeds. It is a key process in many industries including mineral processing, fertilisers, agrochemicals and pharmaceuticals. Key granule properties important for product quality are granule size distribution and porosity (which controls granule attrition and dispersibility). These properties are in turn fixed by the rate and extent of various macroscopic growth mechanisms in the granulation process. These mechanisms (nucleation, consolidation, coalescence, layering, etc.) have been clearly identified and classified [1–3]. What is not clearly understood is the quantitative relationship between feed properties, operating conditions and these mechanisms. For example, it is still not possible to predict the granulation behaviour of a formulation from the fundamental particle and binder properties except for a few special cases.

This is a particular problem in industries where there are many different feed formulations with widely varying properties, such as pharmaceuticals and agricultural chemicals. Extensive, and expensive, pilot plant testing is required for each newly developed formulation. Even so, failure to meet specification on scale-up from pilot plant to full scale are very common. In these industries, regulations often require the process to be registered even before sufficient feed powder is available for laboratory and pilot scale granulation tests.

This paper reports experimental results for the consolidation of granules during tumbling as a function of granule and binder properties. The mechanisms which resist granule deformation are discussed. A new approach to relating formulation properties to granulation behaviour is presented.

2. Current knowledge of granule deformation, consolidation and coalescence

The traditional approach to granule strength is based on the classic work of Rumpf [4] which predicts that the static tensile strength of a single granule depends on binder surface tension, particle size and granule porosity. To understand granule growth mechanisms, however, a much more detailed understanding of deformation behaviour is needed. Schubert et al. [5] and Kristensen et al. [6] have measured the stress–strain behaviour of granules and powder compacts under uniaxial tension and compression respectively. For coarse powders, granule tensile strength and plasticity increase as
liquid content increases towards saturation. However, Kristensens found that for fine powders, granule strength could decrease with increasing liquid content. This was postulated to be because interparticle friction was a significant contributor to granule strength and that the liquid decreased interparticle friction by acting as a lubricant at particle contacts. However, the range of experimental parameters covered in these studies was quite limited, for example, binder surface tension and viscosity were not varied. Also, strain rates were low and invariant, in contrast to impacts colliding with granules during processing. Our studies of single granule deformation on impact [7] show very plastic behaviour. Binder viscosity was found to have a strong influence on granule deformation. Adams et al. [8] claimed viscous and friction effects, not bridge rupture (surface tension effects), account for most of the energy dissipation on impact, based on computer simulation studies. In contrast Simons et al. [9] claim bridge rupture to be a key mechanism in determining granule growth.

Several workers have shown granules consolidate considerably during tumbling [10,11] or mixing [12]. Granules made from coarse particles compact more quickly than fine particle aggregates. Again, these studies covered only a very limited range of binder and powder properties. It is accepted that granule strength and deformability strongly influence coalescence in tumbling and mixer granulators [2,6,13]. Weak granules that deform easily grow quickly (sometimes referred to as crushing and layering). Stronger granules grow slowly by coalescence and can reach a maximum stable granule size. Capes and Dankwerts [10] proposed an empirical relation for granulation regimes based on binder surface tension and particle size. More recently, Ennis et al. [14] considered the dynamic strength of the mobile liquid bridge between colliding particles and developed a criterion for successful coalescence based on the balance between the kinetic energy of collision and viscous dissipation in the liquid bridge as given by a viscous Stokes number. This approach gave for the first time a quantitative approach to predicting granulation regimes (layering or coalescence) and we have used these results to explain the behaviour of several different granulation systems [15-17]. However, the original analysis considers collisions between effectively elastic granules and ignores significant granule deformation. Further work [18] accounts for deformation but requires empirical parameters which are not known a priori and must be determined from granulation tests.

In summary, granule growth and compaction in tumbling and mixer granulators is strongly affected by granule deformation behaviour which is still not well understood. Frictional, viscous, elastic and capillary effects may all be important in determining granule deformation.

3. Experimental

The experiments measured the change in granule porosity with time while tumbling in a laboratory granulation drum.

<table>
<thead>
<tr>
<th>Binder</th>
<th>Density (kg m⁻³)</th>
<th>Viscosity (Pa s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water</td>
<td>997</td>
<td>0.0011</td>
</tr>
<tr>
<td>50 wt.% glycerol</td>
<td>1123</td>
<td>0.0054</td>
</tr>
<tr>
<td>85 wt.% glycerol</td>
<td>1215</td>
<td>0.070</td>
</tr>
<tr>
<td>100 wt.% glycerol</td>
<td>1256</td>
<td>0.65</td>
</tr>
</tbody>
</table>

The effects of both powder and binder properties were investigated. Three size fractions of glass beads were used as the model powders. The powders had specific surface mean sizes ($\bar{X}_{50}$) of 8, 15 and 39 μm respectively (or mass mean sizes of 27, 40 and 70 μm) as measured by a Malvern laser sizer. Glycerol–water mixtures were used as binders so that the binder viscosity (measured by a concentric cylinder viscometer) could be varied over a wide range. Measured binder properties are given in Table 1.

The stainless steel granulation drum was 0.3 m in diameter and 0.2 m long. The drum had six small wedge shaped lifters, each 5 mm higher, to aid in tumbling the mass. The drum speed was kept constant at 30 rpm. A flexible scraper bar was used to prevent powder build up on the drum surface. This drum design is similar to that reported by Sastry and Fuerstenau [1].

To begin an experiment, approximately 1 kg of powder was mixed with a known amount of binder in a plastic bag. The mixture was hand kneaded to distribute the binder evenly. The wet mass was then pushed through a 2 mm sieve into the drum. This procedure gave a consistent feed material for the start of each experiment. The drum was then tumbled for up to 5000 revolutions. 20 to 40 g samples were taken from the tumbling mass at regular interval to measure granule apparent density and liquid content. Up to 50% of the original charge was removed as samples during an experiment. Experiments were finished when either the granule porosity had reached a stable value or when the granules had grown larger than 10 mm (where the porosity could no longer: be measured accurately).

Granule porosity was calculated from the binder content measured, the apparent granule density and the density of the binder and particles. Binder content for water-bound granules was measured by mass difference before and after drying. Glycerol-bound granules were weighed, then carefully washed with water before drying and final weighing. Previous workers measured granule apparent density by benzene displacement [10] or mercury porosimetry [19]. We chose a technique of kerosene displacement [20]. Approximately 30 g of granules were weighed and immersed in kerosene in a 50 ml volumetric flask. The volume of kerosene displaced was calculated knowing the mass of the full flask and the density of the kerosene. The technique was precise to ± 1%. This level of precision is important, since the increase in granule apparent density during an experiment was usually less than 10%. Under conditions where granules grew...
quickly, a larger volumetric flask (100 ml) was used to handle granules larger than 5 mm. In these experiments precision was reduced to ± 3% (hence the larger scatter in results for 0.239 g/g glycerol in Fig. 2(b)). Above 10 mm size, granule density could no longer be measured precisely enough to show consistent behaviour.

This technique assumes that the non-wetting kerosene does not displace binder or air in the intragranule pores. Granules maintained their shape and strength even after long periods immersed in kerosene which indicates that the binder was not displaced. For loose porous granules there was in fact some displacement of air from the pores. This led to spurious high density measurements (low porosity) early in some experiments (see, for instance, the early measurements for 0.17 g/g water in Fig. 2(a)). However, the granules eventually compacted sufficiently to resist the displacement of air by penetrating kerosene. This technique also showed good reproducibility (see duplicate experiments in Fig. 2(b)).

4. Results and discussion

Fig. 1 shows typical results for changes in granule porosity during tumbling. Granule porosity decreases with time before levelling off at a final minimum value. Similar trends were shown by other workers [10–12]. We found that powder and binder properties effected both the rate of consolidation and the final minimum porosity.

Fig. 1 also shows the effect of particle size on consolidation. The rate of consolidation decreased as particle size decreased. Similar trends were observed for all binder contents and viscosities. Decreasing the particle size increases the specific surface area of particles and increases the resistance to granule deformation by both interparticle friction and viscous dissipation. Therefore, the observed experimental trend is expected.

The results were fitted to an empirical model (the lines shown in Figs. 1 and 2). This model assumes that for a given system, the rate of granule consolidation at any time should be inversely proportional to the porosity at that time. The simple equation to describe this behaviour is an exponential decay:

\[
\varepsilon - \varepsilon_{\text{min}} = \exp(-kN)
\]

where \(\varepsilon\) is the granule porosity, \(\varepsilon_{\text{min}}\) is the minimum granule porosity, \(k\) is the rate constant (revs.\(^{-1}\)), and \(N\) is the number of drum revolutions (revs.). The rate constant, \(k\), and the minimum porosity, \(\varepsilon_{\text{min}}\), are expected to be complex functions of drum speed and geometry, particle properties (for example, size and shape), binder properties (viscosity, surface tension and contact angle) and binder content. Any initial spurious results due to kerosene pore penetration were neglected in the curve fit.

Changes to binder viscosity and liquid content yielded some interesting and surprising results (see Figs. 2–4). Fig. 2(a) shows the effect of increasing liquid content on granule porosity for a low viscosity binder (water). Increasing the liquid content increased the degree of consolidation. Kristensen et al. [12] found similar results for consolidation during high speed mixer granulation. Contrast these results with Fig. 2(b) which shows a similar set of data for the high viscosity glycerol binder. Increasing the liquid content
decreased the extent of consolidation. Thus, there is no simple trend of consolidation with liquid content. Fig. 3 summarizes the results of many experiments and shows this more clearly. As binder viscosity increases, the effect on minimum porosity, $e_{\text{min}}$, reverses. Note that at intermediate viscosities consolidation is actually insensitive to liquid content. Fig. 3 also shows that the effect of viscosity on consolidation is not straightforward. Final consolidation decreases with viscosity at low liquid contents, but increases with viscosity at high liquid contents. Clearly, the effects of viscosity and liquid content are highly interactive.

We propose these unusual results because both interparticle friction and viscous dissipation are important mechanisms that resist granule deformation. To consolidate, granules must deform when they collide. Stronger granules, that is those with greater resistance to deformation, will deform less. To deform, particles within the granule must slide against each other. This motion is resisted by interparticle friction. Liquid in the granule pores lubricates the particle–particle contacts. Therefore, increasing the liquid content reduces the resistance to consolidation by interparticle friction. Granule deformation also requires liquid in the granule to squeeze through the intragranule capillaries. This liquid motion is resisted by viscous dissipation in the fluid. The contribution of viscous dissipation increases with increasing liquid content. Thus, increasing liquid content (or viscosity) shifts the controlling dissipation mechanism from interparticle friction towards viscous dissipation. Unless the balance between the two mechanisms is known accurately for a given system, the effect of changes to binder parameters on granulation behaviour cannot be predicted even qualitatively.

Fig. 4 shows the effect of binder content and viscosity on the empirically fitted rate constant, $k$. This shows that the rate of consolidation decreases as binder viscosity increases which is to be expected from the above discussion. However, Fig. 4 also shows that the rate of consolidation appears to increase with increasing binder content in all four cases (the plotted lines are a straight line fit to the data points) which is contrary to expectations from the above discussion. At this stage we have no explanation for this trend.

This discussion has ignored surface tension (capillary effects). Capillary forces are certainly important in determining the static strength of granules [4]. Their contribution to the dynamic processes of consolidation and coalescence is still under debate. Note that contribution of capillary forces to consolidation will be fundamentally different from liquid and particle frictional effects because the capillary forces are not dissipative. Capillary forces always act to pull particles together, whereas frictional and viscous forces resist particle movement in both directions as granules consolidate and dilate during collisions.

5. A new approach to predicting granulation behaviour for different formulations

These results and discussion emphasize the very complex nature of granule consolidation. Predicting coalescence behaviour is even more difficult. To overcome these difficulties, a new approach is proposed. Instead of trying to relate directly microscopic particle and binder properties to growth regimes directly, soil mechanics theory can be used to develop a constitutive model for the stress–strain relationship of the wet powder and this used as a basis for developing a new granulation criterion.

5.1. Constitutive model development

The failure strength of a dry powder is well described by the Mohr–Coulomb plasticity theory and easily characterized by direct shear measurement, for example, Jenike shear cell. In contrast, suspensions are characterized by a viscoelastic relationship between the applied stress and strain rate. In granulation, the wet powder or paste lies somewhere between these extremes. There is a strong analogy with the behaviour of partially and fully saturated soils. Fig. 5 shows a typical stress–strain relationship for such a system [21,22]. Upon
Fig. 5. Stress–strain behaviour of a wet bulk solid [21].

loading, the element immediately deforms (OA) and some of this deformation is recoverable, or elastic (AB). With time, under load, the sample yields and continues to deform (AC). On removal of the stress, the sample recovers elastically (CD) but due to permeability effects, it takes time to dissipate the fluid pressure (DE). At point E, we observe the full plastic deformation of the element.

Fig. 6 shows conceptually a possible lumped parameter model to describe the combined plastic and viscoelastic effects involved in such behaviour. The differential elastic effects can be written as a linear elastic model in terms of bulk and shear moduli. The plastic model essentially gives the shape of the yield surface and could be described by a Mohr–Coulomb model. Viscous effects are modelled by relating the applied stresses in excess of the yield stress to strain rates. Finally, pore pressure effects can be represented by Darcy's Law to a first approximation.

A full constitutive model considers the combined behaviour due to these separate effects. Standard triaxial consolidation tests as used in soil mechanics can also be used to validate the constitutive model and extract the associated constants. Thus the deformation behaviour of wet compacts (granules) can be characterized for a wide range of powder and binder properties and using different stress paths, degrees of saturation and impact conditions.

5.2. A new criterion for granule consolidation and coalescence

When two granules collide, they will deform until the initial kinetic energy is dissipated through viscous and frictional interactions or stored elastically. If sufficient energy is stored, the granules can rebound which may account for collisions between well-compacted (strain hardened) granules or individual particles. This is described by the existing theory of Ennis et al. [14].

However, for more deformable granules, most of the energy is dissipated and rebound does not occur. A dumbbell intermediate results which will eventually form a single, roughly spherical granule if it survives shear and tensile stresses from subsequent collisions. This will depend to a large degree on the contact area developed during the collision. In an extreme case of collision between two very deformable granules, the smaller granule may be squashed flat and the probability of successful coalescence will be high. As tumbling or mixing time increases, compaction and strain hardening reduce the plastic deformation on impact and increase the elastic component of the collision, making coalescence less likely.

If the impact velocity is known, the constitutive relationship for the granule can be used to derive the degree of deformation as the energy dissipation is simply the integral of stress with respect to incremental strain. For given granule constitutive properties, the required impact velocity for rebound can be calculated. The end result will be a new granulation criterion which will relate collision velocity, which is controlled by operating variables such as mixer impeller speed or rotating drum speed, and the bulk material parameters of the wet particulate system to growth mechanisms and growth rate. A similar approach can be used for predicting the rate of granule compaction with time as the result of many collisions.

Fig. 7 shows a block diagram of the proposed methodology along with experimental studies designed to validate the approach at different levels. Should it be successfully validated, the key advantage of this approach is the ability to predict granulation behaviour from measurable bulk solids properties, therefore limiting significantly the amount of lab-
oratory and pilot scale granulation tests needed in the design and scale up of granulation processes.

6. Conclusions

Granule consolidation during tumbling is a complex process controlled by the balance between the mechanisms which resist granule deformation: interparticle friction and viscous dissipation. The rate of consolidation decreases with decreasing particle size as the influence of both dissipative mechanisms is increased. As liquid content increases, interparticle friction effects are decreased but viscous losses become more significant. Thus, the effects of binder viscosity and liquid content are highly interactive. Unless the balance between the two mechanisms is accurately known for a given system, the effect of changes to binder parameters on granulation behaviour cannot be predicted even qualitatively.

To overcome these difficulties a new methodology for relating formulation properties to granulation behaviour is suggested based on bulk powder properties measured by drained and undrained triaxial consolidation tests and the development of a new granulation criterion for deformable granules. The methodology remains to be tested at a range of levels from individual granule deformation experiments to pilot plant trials.

References