

Fundamental studies of granule consolidation Part 2: Quantifying the effects of particle and binder properties

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Abstract

Glass ballotini of surface mean size 10, 19 and 37 μm were granulated in a 30 cm diameter tumbling drum. Water, glycerol and surfactant solutions used to vary the viscosity and surface tension of the binder liquid. The change of porosity with time was found to be well described by a simple, empirical, exponential decay equation:

$$\frac{\varepsilon - \varepsilon_{\min}}{\varepsilon_0 - \varepsilon_{\min}} = \exp(-kN)$$

where ε is the average granule porosity after N drum revolutions, ε_0 is the initial average porosity of the feed, $\varepsilon_{\min}$ is the minimum porosity reached by the tumbling granules and k is the consolidation rate constant. $\varepsilon_{\min}$ and k were complex functions of binder and solid properties—decreasing particle size and increasing liquid viscosity both reduced k ; decreasing surface tension increased $\varepsilon_{\min}$; and the effect of binder content on $\varepsilon_{\min}$ varied with binder viscosity. Where possible, these relationships were expressed quantitatively and compared with consolidation models in the literature. k was also related to the previously measured dynamic yield stress of the granules. The results show that capillary, viscous and friction forces all play a role in granule consolidation. © 1998 Elsevier Science S.A. All rights reserved.

Keywords: Granulation; Agglomeration; Consolidation; Inter-particle friction; Capillary forces; Viscous dissipation; Impact deformation; Dynamic yield stress

1. Introduction

Granulation is the process of agglomerating particles together into larger, semi-permanent aggregates by spraying a liquid binder onto the particles as they are agitated in a tumbling drum, fluidised bed, high shear mixer or similar device. The liquid binds the particles together by a combination of capillary pressure, surface tension and viscous forces until more permanent bonds are formed by subsequent drying or sintering. Some advantages of agglomerated materials include improved flow properties, reduced dustiness, increased bulk density and the co-mixing of particles which would otherwise segregate. Improper granulation causes problems in down stream processes such as caking, segregation and poor tableting performance. However, in spite of its importance and over 40 years of research, it is still impos-

sible to quantitatively predict the growth behaviour of a new formulation from only a knowledge of its fundamental properties.

As granules are agitated in a granulator they experience many collisions with other granules and with walls of the equipment. These collisions cause *consolidation* of the granules (also called densification or compaction) which reduces their porosity and size, squeezes out entrapped air and possibly even squeezes liquid binder to the granule surface.

Granule porosity controls granule strength, an important product property. Porous granules are weak, friable and disperse quickly in solution, whereas low porosity granules are strong and slow to disperse [1,2]. Granule porosity also controls granule deformability and liquid saturation which have a strong influence on granule growth mechanisms [3]. If consolidation causes liquid to be squeezed to granule surfaces, this may trigger rapid granule growth after a long period of little or no growth [4]. However, consolidation also reduces granule deformability, reducing the area of contact formed between colliding granules, and hence may decrease the likelihood of successful coalescence [3].

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Hence, consolidation affects both the granule product properties and the growth mechanisms that form them. However, until recently, work in the literature studying granule consolidation was limited. Several workers have measured granule porosity changes during batch granulation and noted that porosity initially decreases rapidly and then levels off to a stable value [5,6]. Others have noted that increasing moisture content increases the initial rate of granule consolidation [7,8]. This was thought to be because additional moisture increases particle mobility by acting as a lubricant at points of inter-particle contact [1,3]. However, none of these workers studied the effects of binder viscosity or surface tension, and seldom were systems allowed to reach their minimum porosity.

There are only two theoretical treatments of granule consolidation in the literature. Ouchiya and Tanaka [8] assumed granules were held together by the capillary pressure of the binder. This pressure generates a normal force causing friction at inter-particle contacts. They considered how the coordination number of particles in the granular assembly increased when forces were applied. They ignored any particle detachment that might occur due to dilation of the assembly.

They found that the rate of granule consolidation was:

$$\frac{d\varepsilon}{d\tau} \cong - \left\{ 1 - \frac{(1-\varepsilon)^3}{\varepsilon \cdot K_c} \right\}^n \quad (1)$$

where

$$\tau = (13/256) \cdot q \cdot t \quad (2)$$

$$K_c = \frac{16}{3} \cdot \frac{\rho_s g H_{\max}}{f(\mu)} \cdot \frac{d}{\phi(s) \cdot \gamma \cos(\theta)} \quad (3)$$

ε = granule porosity at time t , K_c = dimensionless granule compaction rate, n = parameter describing distribution of granule impact energies, τ = dimensionless compaction time, q = frequency of granule impacts, ρ_s = density of constituent particles in the granule, g = gravitational acceleration, $H_{\max}$ = maximum height of granule fall in tumbling granulator, $f(\mu)$ = function of coefficient of sliding friction at inter-particle contacts, $\phi(s)$ = function of granule saturation and particle assembly ($\cong 6$ for saturated matrix of monosized spheres), γ = liquid surface tension and θ = liquid–solid contact angle.

Hence, the dimensionless compaction rate, K_c , is proportional to particle size and inversely proportional to liquid surface tension. Compaction rate is also proportional to $\rho_s g H_{\max}$, which is a measure of the energy of granule collisions. Setting $d\varepsilon/d\tau = 0$, the minimum porosity the system reaches, $\varepsilon_{\min}$, after an infinite time is:

$$\frac{\varepsilon_{\min}}{(1-\varepsilon_{\min})^3} = \frac{1}{K_c} \quad (4)$$

Hence, this model predicts that the dimensionless compaction rate and minimum porosity are linked. Increasing the

compaction rate will decrease the minimum porosity (i.e., increase the amount of consolidation).

The second theoretical treatment of granule consolidation was by Ennis et al. [9] who considered only the effect of binder viscosity. They found that the amount of consolidation per collision would increase with increasing viscous Stokes number, St_v :

$$\frac{\Delta x}{h} = 1 - \exp(-St_v) \quad (5)$$

where Δx = reduction in inter-particle gap distance h , $St_v = 8\rho u_0 a / 9\mu$, ρ = particle density, u_0 = collision velocity, a = particle radius and μ = binder viscosity. Hence, increasing binder viscosity and decreasing particle size should both decrease the amount of consolidation and increasing the impact energy should increase the amount of consolidation.

Recently, Iveson et al. [10] studied the batch consolidation of glass ballotini using water and glycerol binders. Their results fitted a simple empirical exponential decay model:

$$\frac{\varepsilon - \varepsilon_{\min}}{\varepsilon_0 - \varepsilon_{\min}} = \exp(-kN) \quad (6)$$

where ε is the average granule porosity after N drum revolutions, ε_0 is the initial porosity of the feed, $\varepsilon_{\min}$ is the minimum granule porosity reached by the system and k is the consolidation rate constant. Increasing binder viscosity and decreasing binder content both decreased k . The effect of binder content on $\varepsilon_{\min}$ varied—at low binder viscosities, increasing binder content decreased $\varepsilon_{\min}$, whereas at high viscosities, increasing binder content increased $\varepsilon_{\min}$. This variable effect was attributed to the opposite effects of binder content on friction and viscous forces. Increasing binder content increases viscous forces but decreases inter-particle friction forces due to lubrication of inter-particle contacts.

This paper extends the earlier work of Iveson et al. [10] by quantifying the effects of particle and binder properties on the rate of granule consolidation, and also includes new experimental measurements of surface tension effects. These results are compared to the above two theoretical models and a mechanistic framework proposed to explain the variations. In addition, the consolidation rate constant is compared with the measured dynamic yield stress of the granules and the implications of this relationship discussed. From these studies of granule consolidation, it is hoped to be able to improve our understanding of granule growth behaviour.

2. Experimental

2.1. Materials

Glass ballotini were chosen as a model material since they are uniform, non-soluble particles. Three different grades were used with surface mean sizes of 10, 19 and 37 μm . The 10 μm ballotini was purchased from Cataphote (PO Box

Table 1
Properties of the glass ballotini

Surface-mean size ^a $x_{3,2}$ (μm)	Mass-mean size ^a $x_{4,3}$ (μm)	Coefficient of variation ^a $\sigma_{3,3}/x_{4,3}$ (–)	Dry-tapped bulk porosity ^b (–)	Wet-tapped bulk porosity ^b (–)
37	71	0.31	0.388	0.372
19	37	0.49	0.51	0.382
10	29	0.38	0.44	0.385

^aMean sizes and standard deviation ($\sigma_{3,3}$) measured by Malvern Mastersizer/E.

^bBulk porosities measured by tapping sample in 30 mm diameter volumetric cylinder.

2369, Jackson, MS, 39225-2369, USA) and had a skeletal density of 2.37 ± 0.01 g/ml (measured by both water-displacement and Helium pycnometry). The 19 and 37 μm ballotini were purchased from Potters Industries (Lot 4 Boundary Rd., Laverton, Victoria, 3028, Australia) and had a density of 2.45 ± 0.01 g/ml. Complete details of the ballotini are given in Table 1.

Solutions of glycerol were used to vary binder viscosity between 0.001 and 1 Pa s. Solutions of the surfactant sodium dodecylbenzene sulphonate (NDBS) were used to vary binder surface tension between 31 and 72 mN/m. The complete details of these solutions are given in Table 2.

2.2. Technique

Approximately 1 kg of glass ballotini were hand mixed with a known amount of binder in a plastic bag. This feed was then pushed through a 2 mm sieve into a 30 cm diameter, 20 cm long stainless-steel granulation drum. The drum had six 5 mm high triangular lifter bars spaced evenly around the interior to aid in tumbling the mass and a flexible scraper bar to prevent excessive build up of material on the drum wall. The drum was tumbled for up to 2 h at 30 rpm. During this time 50 g samples were periodically taken from the tumbling mass.

In this paper, porosity is defined as the volume fraction of the granule occupied by pores. The pores may be occupied by both liquid and air phases. Granule porosity was calculated indirectly from the granule apparent density. This was measured by the volumetric displacement of kerosene in a 50 ml volumetric flask. The resulting porosity versus time data was then fitted to the three-parameter exponential decay model of Iveson et al. [10] (Eq. (6)). The uncertainty of fitting the consolidation rate constant, k , to the data is high, typically of the order to 25 to 50%, whereas ε_{min} was much more precise with typical uncertainty less than 5%. A complete description of the experimental technique and data analysis is found elsewhere [10,12].

The present technique is restricted to systems where granule growth was slow or zero. All granules sampled were between 2 and 5 mm in size—any larger and they would not fit down the neck of the 50 ml volumetric flask. Using a larger flask introduced too large an error in measuring the kerosene volume to be able to accurately determine the porosity.

Table 2
Properties of the liquid binders

Liquid	Density ^a (g/ml)	Viscosity ^b (Pa s)	Surface tension (mN/m)
Water	0.997	0.0011	72 ^c
50 wt.% glycerol	1.123	0.0054	70 ^d
85 wt.% glycerol	1.216	0.070	66 ^d
Glycerol	1.256	1.1	63 ^d
NBDS solutions	–	–	–
0.00094 M			31 ^c
0.00053 M			48 ^c
0.00044 M			51 ^c
0.00032 M			52 ^c
0.00019 M			60 ^c

^aMeasured in 25 ml specific gravity bottle.

^bMeasured in Contraves Rheomat 115 rheometer. Shear rate was varied between 20 and 150 s^{-1} for glycerol and between 300 and 3650 s^{-1} for water.

^cMeasured by the Wilhemy Plate method using 2 cm square pieces of filter paper.

^dFrom Ref. [11].

3. Experimental results

3.1. Effects of particle size distribution

Fig. 1 shows a typical plot of porosity vs. time for the three different sized ballotini granulated with glycerol. Fig. 2

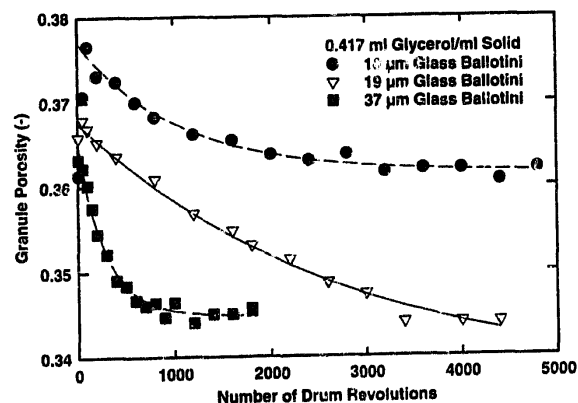


Fig. 1. Granule porosity vs. number of drum revolutions for the three grades of glass ballotini granulated with 0.417 ml/ml of glycerol. Lines show model fit to the data (from Ref. [10]).

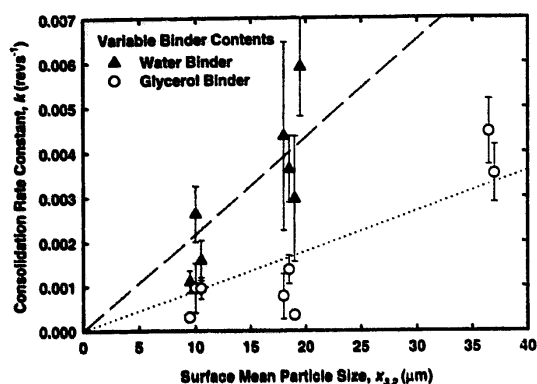


Fig. 2. Consolidation rate constant, k , vs. surface mean particle size for glass ballotini granules bound with water or glycerol binders across a range of binder contents (0.42 to 0.49 ml binder/ml solid).

shows k versus the specific surface mean particle size for both water and glycerol binders. Error bars show the 95% confidence intervals on the parameters determined by fitting Eq. (6) to the experimental data.

In general, the consolidation rate constant was proportional to specific surface mean particle size (Fig. 2). This result is consistent with the prediction of Kapur [4] and the models of Ouchiya and Tanaka [8] and Ennis et al. [9]. The trend is very clear, except when comparing 10 and 19 μm ballotini granulated with glycerol. The consolidation rate constant was very low when glycerol binder was used, making it difficult to distinguish the effect of particle size in this case.

The 10 μm ballotini did not consolidate as far as the 19 and 37 μm ballotini (Fig. 1; similar trends were also seen at other liquid contents [12]). This is consistent with the work of Capes [13] who found that granules made from fine particles ($d_p < 30$ μm) needed a greater volumetric amount of binder to reach saturation (i.e., had a higher porosity) than granules made from coarse particles. There was no significant difference between the extent of consolidation of the 19 and 37 μm ballotini (Fig. 1), but this was probably due to the wider size distribution of the 19 μm ballotini (see Table 1) which allowed small particles to fill interstitial spaces and lower the porosity.

3.2. Effects of binder content and binder viscosity

Fig. 3 is a typical plot of the effect of varying water content on granule consolidation behaviour. Figs. 4 and 5 show the effects of varying binder content on ϵ_{min} and k , respectively, for the granulation of 19 μm ballotini with a range of glycerol/water solutions. Similar trends were also observed with the 10 and 37 μm ballotini [12]. Fig. 6 plots k versus binder viscosity for the 19 μm ballotini. The range of binder contents that could be studied was limited to between 0.417 and 0.490 ml binder per ml solid. At lower binder contents, the granules were too weak and tended to fall apart or suffer pore penetration when immersed in kerosene. At higher binder contents

the granules quickly grew larger than 5 mm and could not fit through the neck of the 50 ml volumetric flask.

In agreement with the model of Ennis et al. [9], increasing binder viscosity always decreased k (Figs. 2, 5 and 6). In fact, k was inversely proportional to binder viscosity to the power 0.26 ± 0.13 for the 19 μm ballotini (Fig. 6). The combined effects of surface-mean particle size, d_p , (Fig. 2)

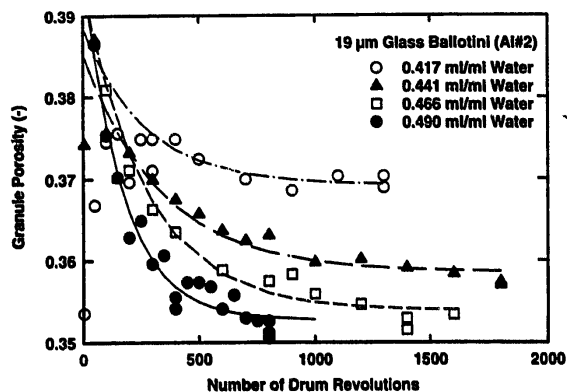


Fig. 3. Granule porosity vs. number of drum revolutions for 19 μm ballotini granulated with varying amounts of water ([10]).

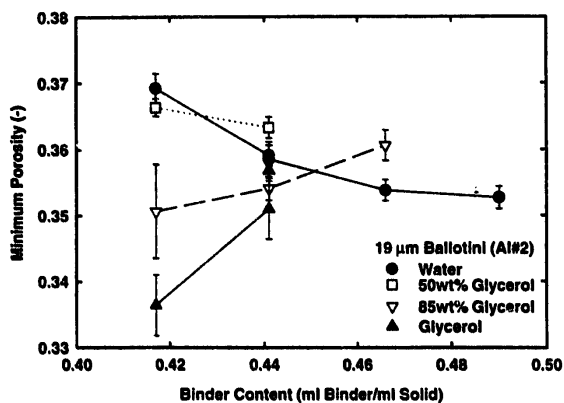


Fig. 4. Granule minimum porosity vs. binder content for 19 μm ballotini granulated with water, 50 wt.%, 85 wt.% and 100 wt.% glycerol ([10]).

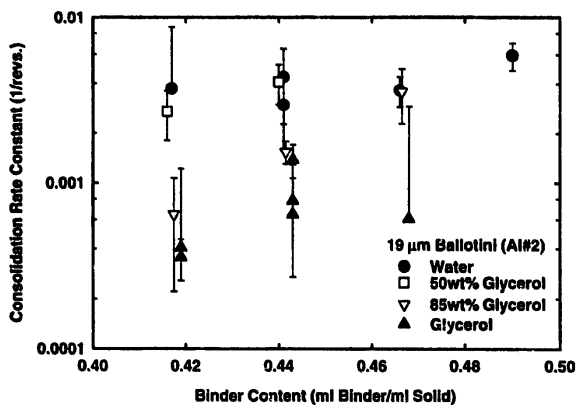


Fig. 5. Consolidation rate constant vs. binder content for 19 μm ballotini granulated with water, 50 wt.%, 85 wt.% and 100 wt.% glycerol.

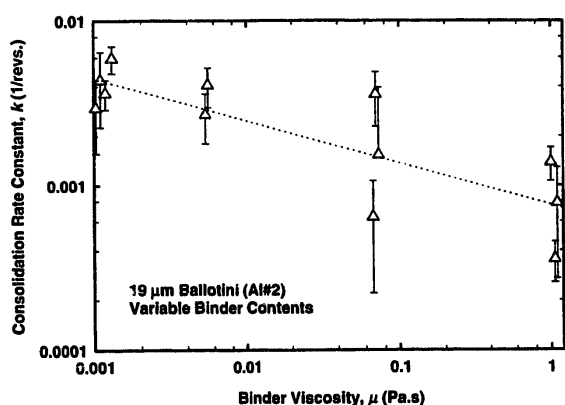


Fig. 6. Effect of binder viscosity on the consolidation rate constant of 19 μm ballotini for a range of binder contents. Dotted line shows Eq. (2).

and binder viscosity, μ (Fig. 6), can be expressed by the equation:

$$k \propto \frac{d_p}{\mu^a} \quad (7)$$

where $a = 0.26 \pm 0.13$ for 19 μm ballotini granulated in a 30 cm drum at 30 rpm.

Increasing the amount of water binder always increased the extent of consolidation (lowered $\varepsilon_{\min}$) (Figs. 3 and 4). In contrast, increasing the amount of glycerol binder decreased the extent of consolidation (Fig. 4). There appears to be an intermediate solution viscosity (between 50 and 85 wt.% glycerol in the case of 19 μm ballotini) at which $\varepsilon_{\min}$ is insensitive to the binder content (Fig. 4). Note also that the effect of increasing binder viscosity depends on the binder content. At low binder contents, viscous binders consolidated more than non-viscous ones. However, at high binder contents, viscous binders caused less consolidation than non-viscous ones.

For the water binder, the *initial* rate of consolidation was greater at higher binder contents (Fig. 3). However, this was only because the granules had further to consolidate, i.e., the 'driving force' for consolidation ($\varepsilon_0 - \varepsilon_{\min}$) was greater. The actual *consolidation rate constant* (k) was not significantly effected by binder content (Fig. 5) (this conclusion differs from that of Iveson et al. [10], who, because they did not calculate the uncertainty in k , concluded that k increased with increasing binder content).

Kapur and Fuerstenau [7] also found that increasing the amount of water binder increased the *initial* rate of granule consolidation. However, they did not reach a stable minimum porosity, so no conclusions about the effects of water content on $\varepsilon_{\min}$ or k can be made from their data [12]. Ritala et al. [14] measured granule consolidation in a high-shear mixer with different viscosity binders. However, they added binder continuously and also did not reach a stable final porosity. Hence, there is no published data against which the present results can be directly compared.

3.3. Effects of binder surface tension

Fig. 7 shows typical results of the effect of binder surface tension on granule porosity versus number of drum revolutions. Decreasing binder surface tension (increasing surfactant concentration) decreased the extent of consolidation (increased $\varepsilon_{\min}$). A phenomena that has not been observed previously was the sudden increase in granule porosity that occurred towards the end of some experiments. This was observed to occur at the onset of rapid granule growth, and thus may be an artefact of the granule size starting to increase. Data points that occurred after the start of this rise in porosity were not used when fitting results to Eq. (6).

Figs. 8 and 9 show plots of $\varepsilon_{\min}$ and k versus binder surface tension for glass ballotini of two sizes over a range of binder contents. Decreasing binder surface tension always increased $\varepsilon_{\min}$ (decreased the extent of consolidation). Decreasing binder surface tension generally increased k although there was a minimum or leveling off in k at surface tensions near 60 mN/m. The model of Ouchiya and Tanaka [8] agrees with the result that decreasing binder surface tension increases the rate of consolidation due to the reduced frictional forces at inter-particle contacts. However, that model also predicts that lowering surface tension should decrease

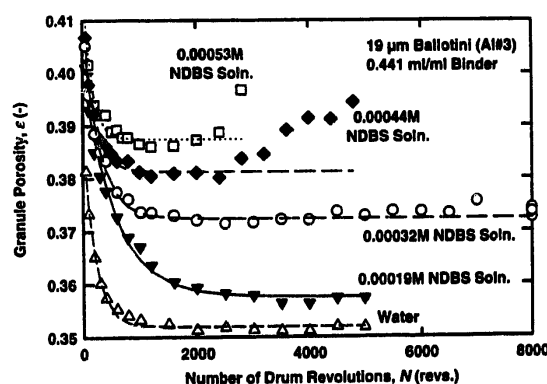


Fig. 7. Granule porosity versus number of drum revolutions for 19 μm ballotini granulated with varying concentration NDBS solutions.

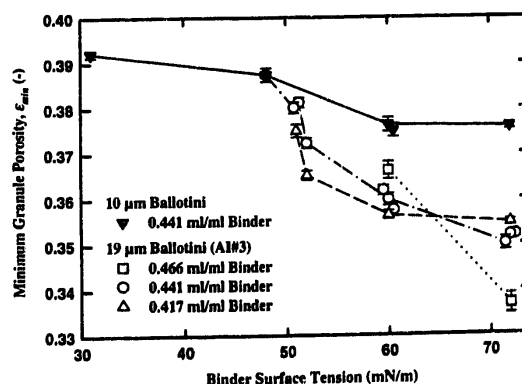


Fig. 8. Minimum granule porosity versus binder surface tension for two sizes of ballotini over a range of binder contents.

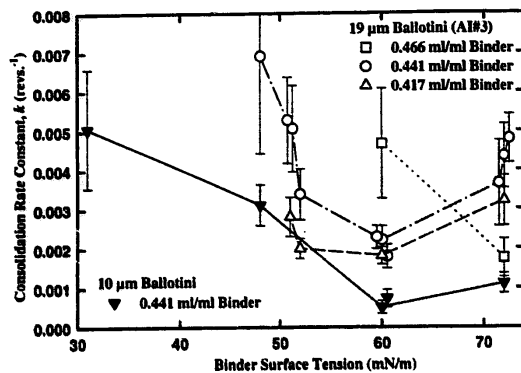


Fig. 9. Consolidation rate constant versus binder surface tension for two sizes of ballotini over a range of binder contents.

the minimum porosity (increase the amount of consolidation) whereas these experiments found the opposite effect.

4. A mechanistic explanation of the consolidation results

Granules may both dilate and compact during impacts [4]. Three forces control this behaviour: inter-particle friction, capillary and viscous. Inter-particle friction and viscous forces are dissipative, i.e., they resist both consolidation and dilation of the particle matrix. In contrast, capillary forces are conservative, i.e., they always act to pull particles together and thus aid consolidation and resist dilation.

As granules consolidate, the inter-particle friction and viscous forces increase (due to a greater number of inter-particle contacts and smaller gap distances). Capillary forces also increase until the granule becomes saturated [1,5]. Hence, granules consolidate until they become strong enough to resist any further consolidation. At this minimum porosity, there is a dynamic equilibrium between their tendency to dilate and consolidate during impact.

As average particle size (d_p) decreases, capillary, viscous and friction forces all increase (on a per unit area basis). Inter-particle friction increases because the number of inter-particle contacts increases [4]. Capillary forces increase because the liquid bridge curvature between particles increases as the pore size decreases (which increases the capillary suction pressure) [1]. Viscous forces also increase as the average gap-distance between particles is decreased [9]. This is why the rate of consolidation is proportional to particle size (Fig. 2 and Eq. (7)).

The effect of binder content is complex because it has opposite effects on inter-particle friction and viscous forces. Increasing binder content reduces inter-particle friction due to lubrication [2,4] but increases viscous forces since there is more binder to be squeezed between particles as they rearrange. When inter-particle friction dominates (e.g., low viscosity binders like water) increasing binder content increases the extent of consolidation (Fig. 3). However, when viscous forces dominate (e.g., high-viscosity binders) increasing the

amount of binder decreases the extent of consolidation (Fig. 4). There is an intermediate binder viscosity at which the decrease in frictional resistance is balanced by the increase in viscous resistance. At this stage, binder content does not affect the extent of consolidation (somewhere between the 50 and 85 wt.% glycerol curves in Fig. 4). This is a finding of major importance. *Unless the relative magnitudes of the viscous and frictional forces are known, it is impossible to predict beforehand the effect of changing binder content, even qualitatively!*

Increasing binder viscosity always decreases the rate of consolidation due to the increase in viscous forces (Figs. 5 and 6). However, at low binder contents, the glycerol-bound granules consolidated further than the water-bound granules (Fig. 4). This may also be due to the equilibrium between dilation and consolidation. The glycerol-bound granules are more able to resist dilation. Hence, although they consolidate more slowly, they can consolidate to a greater extent. Ritala et al. [14] also found in high-speed mixers that at some impeller speeds, viscous bound granules consolidated further than non-viscous bound granules. They speculated that this was because viscous binders are better lubricants and hence promote consolidation in systems where inter-particle friction forces dominate.

Inter-particle friction and surface tension forces are coupled. Reducing liquid surface tension reduces the capillary suction pressure. This in turn lowers the normal forces acting at points of inter-particle contact and hence reduces the frictional resistance to particle rearrangement. Hence, lowering binder surface tension reduces both the frictional resistance to consolidation and also reduces the capillary forces resisting particle dilation and rearrangement. This explains why lowering surface tension increases the consolidation rate. The reason for the apparent minimum in k near 60 mN/m is uncertain (Fig. 9). Lowering γ also lowers a granule's resistance to dilation which shifts the dynamic equilibrium between consolidation and dilation towards a more porous state. Hence, decreasing surface tension increases $\varepsilon_{\min}$ (Fig. 8). Thus, granules with low surface tension binders consolidate more quickly, but do not consolidate as far.

This mechanistic description successfully explains the experimental results and is supported by the limited amount of other data in the literature. It highlights a flaw in the consolidation model of Ouchiyaama and Tanaka [8] which considers the force required to further consolidate a granule but totally neglects granule dilation. Hence they incorrectly predict that the minimum granule porosity will decrease as binder surface tension decreases, whereas in reality the decrease in binder surface tension means granules more easily dilate and thus do not consolidate as far. Also, their model predicts that the minimum porosity always decreases as the rate of consolidation increases, whereas these experimental results show that the two are independent.

Clearly further work is required to understand the phenomena of granule dilation during impact. Although referred to in the early literature [4], the authors are not aware of any

detailed study of this phenomena. Without understanding what controls granule dilation, it will be impossible to predict the maximum extent of consolidation that a formulation will undergo.

5. Comparison with single-impact granule deformation

The complex interaction of capillary, viscous and inter-particle friction forces makes a priori prediction of consolidation behaviour difficult from first principles. However, since granule consolidation results from a large number of individual impacts, it should be related to granule impact deformation. To test this hypothesis, the consolidation rate constant (k) was plotted against the dynamic yield stress (Y) measured during granule impacts [15] for all cases where identical particles and binders were used (Fig. 10). The consolidation rate decreases with increasing yield stress according to the equation:

$$k = A \cdot \exp(-B \cdot Y) \quad (8)$$

where $A = 0.0032 (\pm 0.002) \text{ rev}^{-1}$ and $B = 0.0036 (\pm 0.0015) \text{ kPa}^{-1}$ for 30 cm drums operated at 30 rpm compared to pellets of 41% porosity. This relationship covers two different particle sizes, 10 and 19 μm , a range of binders (glycerol, water and 0.00019 M NDBS soln.) and a range of binder contents (0.417 to 0.490 ml/ml).

This decrease in consolidation rate as dynamic yield stress increases makes intuitive sense — granules with a low yield stress are ones in which the forces holding particles together are weak. Hence the particles are easily rearranged into a lower-porosity packing. The effects of particle size and binder viscosity are the most significant. Increasing binder viscosity and lowering particle size both increase granule yield stress and decrease the rate of consolidation. Increasing binder surface tension also increases granule yield stress and decreases the consolidation rate constant. The effects of binder content were of only secondary order in the range of conditions studied.

This relationship between consolidation rate and dynamic yield stress is a discovery of great importance. Even though a priori prediction of consolidation behaviour from fundamental properties of the powder–binder mixture is difficult, it may be possible to predict the consolidation behaviour of materials directly from their behaviour in simple impact experiments. This also avoids the need to perform lengthy and costly granulation experiments. The drum consolidation experiments described in this paper are tedious and require a skilled operator, whereas the single-impact deformation experiments of Iveson and Litster [15] are relatively inexpensive and quick to perform. In other work, the present authors have proposed a granule growth regime map based on the dynamic yield stress and consolidation behaviour of granules [16]. This could greatly decrease the time and expense of testing new formulations.

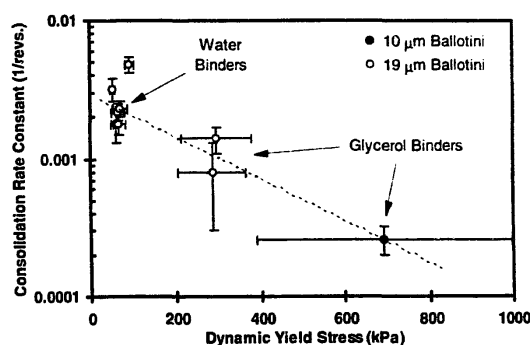


Fig. 10. Consolidation rate constant versus dynamic yield stress measured by Iveson and Litster [15] for two ballotini sizes and a range of binders and binder contents.

However, further work is required to verify and extend this discovery. The effects of equipment size and operating conditions have not been measured. Both theoretical models of granule consolidation [8,9] and some limited experimental data [8,14] suggest that the rate of consolidation increases in more energetic systems (e.g., larger drums, high-shear mixers). While the yield stress measurements are equipment independent, the consolidation measurements apply to only one piece of equipment at one set of operating conditions. Therefore the relationship between dynamic yield stress and consolidation rate will depend on the equipment type, size and operating conditions. This relationship needs to be investigated.

6. Conclusions and implications

This work is the most comprehensive experimental study to date of batch granule consolidation. Granule consolidation behaviour can be characterised by the consolidation rate constant, k , and the final equilibrium porosity, $\epsilon_{\min}$, determined by fitting experimental results to an empirical exponential decay equation.

Decreasing particle size (d_p) and increasing binder viscosity (μ) decrease the consolidation rate constant (k) according to the equation:

$$k \propto \frac{d_p}{\mu^a}$$

where $a = 0.26 (\pm 0.13)$ for 19 μm ballotini granulated in a 30 cm drum at 30 rpm. The consolidation rate constant also generally decreases as binder surface tension is increased, but there was no measurable effect of binder content on k in the range of conditions studied.

Decreasing d_p tends to increase the minimum porosity, although $\epsilon_{\min}$ also depends on the shape of the particle size distribution. The effect of binder content and binder viscosity on the minimum porosity are inter-related. Increasing the amount of a low-viscosity binder decreases $\epsilon_{\min}$ (increases the extent of consolidation) whereas increasing the amount

of a high-viscosity binder increases $\epsilon_{\min}$ (decreases the extent of consolidation). There is a binder viscosity at which the final granule porosity is insensitive to binder content. Decreasing binder surface tension always increased $\epsilon_{\min}$.

Granule consolidation is a balance between the tendency for granules to both dilate and compact during impacts. There are (at least) three forces that affect granule consolidation behaviour: inter-particle friction, capillary and viscous forces. Increasing binder content decreases inter-particle frictional forces due to its lubricating effect, but increases viscous forces due to the larger amount of binder. Unless the relative magnitude of these two forces is known, it is impossible to predict beforehand the effect of binder content on granule consolidation, even qualitatively!

However, it is possible to predict consolidation behaviour from the impact behaviour of the powder-binder mixture. The granule consolidation rate constant (k) in a tumbling drum decreases with increasing dynamic yield stress (Y) according to the equation:

$$k = A \cdot \exp(-B \cdot Y)$$

where $A = 0.0032 (\pm 0.002) \text{ rev}^{-1}$ and $B = 0.0036 (\pm 0.0015) \text{ kPa}^{-1}$ for 30 cm drums operated at 30 rpm compared to pellets of 41% porosity. This relationship holds true across a range of particle sizes, binder contents, binder viscosities and binder surface tensions. Thus, it may be possible to predict the consolidation behaviour of powder-binder mixture without requiring granulation experiments. This may lead to methods for better predicting the consolidation and growth behaviour of new formulations.

Both existing consolidation models [8,9] successfully predict that decreasing particle size and increasing surface tension will decrease granule consolidation rate due to the increase in capillary, viscous and frictional forces. The model of Ennis et al. [9] also successfully predicts that increasing binder viscosity decreases granule consolidation rate due to increased viscous forces. However, the model of Ouchiyama and Tanaka [8] incorrectly predicted that decreasing binder surface tension would increase the amount of consolidation. This is because they failed to take into account the occurrence of dilation during granule collisions. Any general model of granule consolidation must consider capillary, viscous and inter-particle friction forces, and must also include the pos-

sibility of both consolidation and dilation during granule impacts.

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