Growth Regime Map for Liquid-Bound Granules

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A regime map of granule growth behavior is proposed based on granule deformation during collision and the granule liquid content measured as the maximum pore saturation. The granule deformability on collision is represented by a deformation number, which is a ratio of granule impact energy to the plastic energy absorbed per unit strain. Granule growth regimes such as steady growth, induction, nucleation, crumb, and slurry are defined. This regime map qualitatively explains the variations in granulation behavior. Laboratory drum granulation experiments were used to test the regime map. Experiments were performed in a 0.3-m-dia. drum using three sizes of glass ballotini (19, 31, and 60 μm) with water and glycerol as liquid binders. Increasing granule yield stress by decreasing particle size and increasing binder viscosity caused the system to move from steady growth to induction behavior as predicted by the regime map. Preliminary validation with literature data was also encouraging. More work, however, is required to better quantify the boundaries between different growth regimes and to investigate the effect of process agitation intensity. This regime map has great potential to help design and control granulation systems, because it is based on properties of the powder/binder system that can be measured or estimated without performing any granulation tests.

Introduction

In the chemical industry it has been estimated that 60% of products are manufactured as particulates and a further 20% use powders as ingredients. The annual value of these particulates is estimated at $1 trillion in the U.S. alone (Ennis, 1997). Granulation is a key step in many of these industries including mineral processing, agricultural chemicals, pharmaceuticals, and detergents.

Granulation is the process of agglomerating particles together into larger, semipermanent aggregates by spraying a liquid binder onto the particles as they are agitated in a tumbling drum, fluidized bed, high shear mixer, or similar device (Ennis and Litster, 1997). 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 downstream processes such as caking, segregation, and poor tableting performance.

In spite of its economic importance and over 40 years of research, granulation is still more of an art than a science. We have a qualitative understanding of the mechanisms of granule growth and the effects of different variables on granule growth behavior. *However, it is still impossible to predict the granulation behavior of a new formulation from only its fundamental properties.* Neither is it known how to vary a formulation in order to obtain a desired change in product properties. This necessitates expensive and extensive laboratory and pilot-scale testing of all new materials (Pietsch, 1991). This is a particular problem in industries where there are many and frequently changing formulations with widely varying properties (such as food, pharmaceuticals, and agricultural chemicals). Regulations often require these new formulations to be registered before there is even sufficient material available for laboratory and pilot-scale granulation tests. Even when pilot-scale testing does occur, there is still a significant failure rate during scale-up to industrial production.

Granule growth mechanisms have traditionally been divided into categories such as nucleation, layering, coalescence, abrasion transfer, and crushing and layering (Sastry, 1975). However, these categories are somewhat arbitrary depending on the definition of granular and nongranular material. Nu-

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cleation can be considered as the simultaneous coalescence of many small granules. Layering is the coalescence of many small granules with one very large one. Abrasion transfer and crushing and layering can be considered as two-stage processes, first granules break or attrite, and then the fragments coalesce with other granules.

Hence, granulation behavior has more recently been divided into three broad classes of phenomenon (Ennis and Litster, 1997): (1) wetting and nucleation; (2) consolidation and growth; and (3) breakage and attrition.

This article is concerned with granule consolidation and growth in systems where no simultaneous drying takes place. We first survey and discuss the different models and classes of growth observed in the literature. Then, we propose a regime map for predicting granule growth behavior. Preliminary verification of this regime map is attempted using literature data and our own experimental results. The implications of this regime map are then discussed.

Granule Growth Behavior

Capes and Danckwerts (1965) studied the granulation of coarse silica sands with water-ethanol solutions. They found there was a critical ratio of binder surface tension to particle size of 0.46 mN/m/μm, below which the material would not granulate satisfactorily, but only formed a weak "crumb."This ratio of surface tension to particle size is proportional to granule tensile strength as predicted by the theory of Rumpf (1962). *This suggests that particle-binder mixtures must have a critical tensile strength before they will granulate satisfactorily.*

Ouchiyama and Tanaka (1975) developed a model to predict the probability of permanent granule coalescence during collision. They modeled granules as plastic or elastic, surface-dry spheres. Successful coalescence occurred if the bond formed between colliding granules was strong enough to resist being broken by forces within the granulator. This bond strength was assumed to be proportional to the area of contact formed during collision. They found that there was a critical average granule size, above which coalescence would not occur due to the large torque imposed on the dumbbell by subsequent collisions.

Kristensen et al. (1985) extended the work of Ouchiyama and Tanaka by assuming that granule deformation for a given force was proportional to the ratio of critical strain (l_{cr}) to stress (σ_{cr}) at failure measured during compression of bulk samples and also that the bond strength formed between granules was the same as the bulk tensile strength of the granule. They found that the limiting size for coalescence, δ (the size of granules above which no collisions result in permanent coalescence), was given by:

$$\delta^{2/a} = A \cdot \frac{l_{cr}^3}{\sigma_{cr}} \tag{1}$$

where A and a are constants ($a = 1/4$ for perfectly plastic systems, and $a = 1/3$ for elastic impacts). This predicts that granule coalescence is a strong function of granule deformability (l_{cr}). Experimentally it has been found that l_{cr} always increases with increasing liquid saturation (Schubert et al., 1975; Holm et al., 1985). *This suggests that liquid saturation plays a major role in controlling granule growth.* Experimental evidence supports this prediction. Ritala et al. (1988) found that the mean granule sizes of a wide range of materials granulated in a high-shear mixer all collapsed onto one curve when plotted against liquid saturation.

Ennis et al. (1991) took a different approach to modeling coalescence by assuming that granules were elastic spheres with viscous surface liquid layers. They assumed that permanent coalescence took place if the collision kinetic energy was entirely absorbed by viscous dissipation in the binder and elastic losses in the solid. No account was taken of granule plastic deformation or the formation of bonds between the two granule surfaces. From this model, they derived a viscous Stokes' number (St_{vis}) and critical viscous Stokes' number (St^*_{vis})

$$St_{\text{vis}} = \frac{8\rho r u}{9\mu} \tag{2}$$

$$St^*_{\text{vis}} = \left(1 + \frac{1}{e}\right) \ln\left(\frac{h}{h_a}\right) \tag{3}$$

where ρ is the granule density, r is the harmonic mean granule radius, e is the coefficient of restitution, h_a is the typical height of surface asperities, h is the thickness of the surrounding liquid layer, μ is the viscosity of the liquid layer, and u is the half-collision velocity. If $St_{\text{vis}} > St^*_{\text{vis}}$, then inertial effects dominate and all collisions are unsuccessful (coating regime). If $St_{\text{vis}} < St^*_{\text{vis}}$, then viscous effects dominate and all collisions are successful (noninertial regime). This model is significant because it is the first to account for viscous dissipation, which has experimentally been found to be significant by several workers (Ennis et al., 1990; Iveson et al., 1996; Iveson and Litster, 1998a,b; Adams et al., 1994). However, granule impacts can be almost entirely plastic (Iveson and Litster, 1998a), which limits the general applicability of the elastic assumption used in this model.

These two different models of granule coalescence by Ouchiyama and Tanaka (1975) and Ennis et al. (1991) highlight the fact that different forms of granules can exist: granules can be elastic or plastic with either wet or dry surfaces. In fact, a system may change between these states during the course of granulation (see Figure 1). Initially, the granules may be porous, weak and deformable with no surface binder

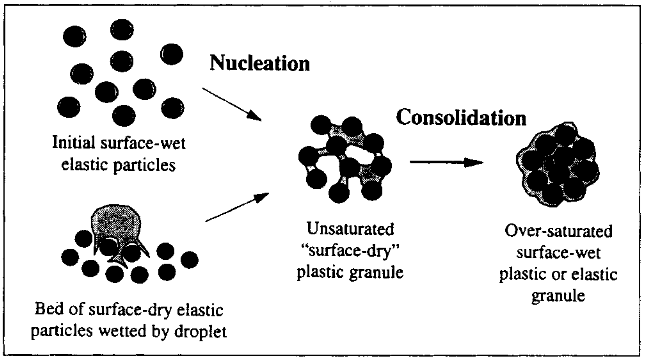

Figure 1. Transitions between plastic and elastic, and surface-dry and surface-wet conditions that can occur during granulation.

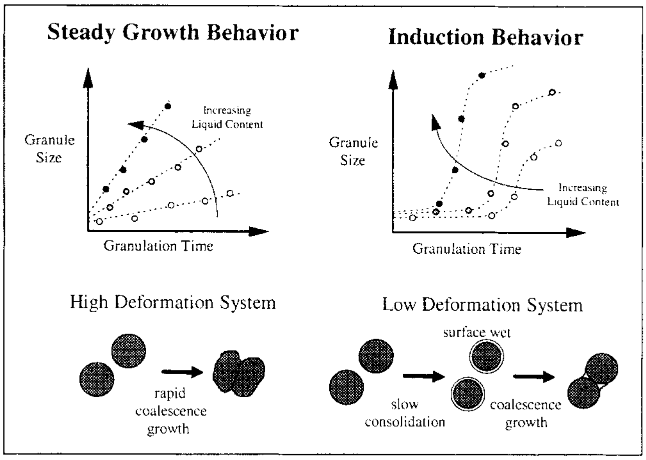

Figure 2. Steady and induction growth behavior and the controlling effect of granule deformability.

(apart from a thin adsorbed surface layer). As they consolidate, they become stronger, less-deformable, and even elastic in nature. If sufficient consolidation occurs, liquid binder may eventually be squeezed to their surface. These variations help explain the various growth behaviors observed in different granulation systems.

There are two broad classes of growth behavior which can occur—either steady growth or induction time behavior (Figure 2). Which type of growth occurs depends on the deformability and rate of consolidation of the granules. Weak, deformable granules form a large area of contact during collision and liquid binder may be squeezed into this contact zone. If this bond is strong enough to resist the separating forces within the granulator, then the pair of granules will be rounded into a new larger granule. This behavior leads to a steady increase in granule size (steady growth) and is common in systems with coarse, narrowly-sized particles and low surface tension and/or low viscosity binder liquids (Newitt and Conway-Jones, 1958; Capes and Danckwerts, 1965; Linkson et al., 1973).

Strong, nondeformable, slowly-consolidating granules do not deform sufficiently during impact to form a strong contact bond. Pairs of collided granules quickly break apart, and there is a period of little or no granule growth—the "induction" period (also called the "nuclei" region (Kapur, 1978) or "consolidation" period (Hoornaert et al., 1994)). The length of this induction period decreases with increasing liquid content. If granules consolidate sufficiently, then liquid binder may eventually be squeezed to their surface. This surface liquid enables strong bonds to form between granules without the need for large amounts of deformation. This triggers rapid granule growth until the granules become so large that the torque exerted on granule dumbbells prevents further coalescence. This class of behavior is frequent in systems with fine particles and/or viscous binders (Linkson et al., 1973; Sastry et al., 1977; Kapur, 1978; Hoornaert et al., 1994).

Proposed Granule Growth Regime Map

From the preceding discussion, any general model of granule growth behavior must consider the following factors:

- Particle-binder mixtures below a minimum strength do not granulate and only form a weak crumb material.
- Coalescence growth is promoted by increased liquid content.
- Granule properties vary with time. Consolidation can cause a system to change from deformable and surface-dry, to elastic and surface-wet.
- There are two broad classes of granule growth behavior, steady growth and induction behavior, depending on the deformability and consolidation rate of the particle-binder matrix.

We propose that granule growth behavior is a function of the system's liquid content and impact deformation during granulation. Granule saturation will vary during batch granulation as the granules consolidate. Therefore, we use the maximum granule pore saturation ($s_{\max}$) as the measure of liquid content

$$s_{\max} = \frac{w\rho_s(1-\epsilon_{\min})}{\rho_l\epsilon_{\min}} \tag{4}$$

where w is the mass ratio of liquid to solid, ρ_s is the density of the solid particles, ρ_l is the liquid density, and $\epsilon_{\min}$ is the minimum porosity the formulation reaches for that particular set of operating conditions. $\epsilon_{\min}$ is a complex function of formulation properties and operating conditions (Iveson et al., 1996; Iveson and Litster, 1998b). For low intensity drum granulation, it can be estimated by measuring the wet-tapped porosity of the formulation.

The granule impact deformation is a function of both the granule rheology and the process agitation intensity. We consider the granule as a rigid-plastic material with strain rate independent dynamic yield stress. For a cylinder of such a material impacting on a flat surface at velocity U, the increase in contact area of the cylinder due to plastic deformation is given by (Hawkyard, 1969)

$$2\left[\frac{A_0}{A} - 1 + \ln\left(\frac{A}{A_0}\right)\right] = \frac{\rho U^2}{Y} \tag{5}$$

where A_0 and A are the area of the impact face before and after collision, and ρ and Y are the density and dynamic yield stress of the material. Equation 5 enables the dynamic yield stress of a material to be determined from simple impact deformation measurements (Iveson and Litster, 1998a).

By analogy, we postulate the extent of deformation of a granule during an impact in a granulator will be a function of the deformation number (De)

$$De = \frac{\rho_g U_c^2}{Y_g} \tag{6}$$

where U_c is a representative collision velocity for the granulator and represents the process intensity. Ennis et al. (1991) discuss suitable choices for the collision velocity in different types of granulation equipment. For drum granulation, the maximum collision velocity is of order ωD where ω is the drum rotational speed and D is the drum diameter. Y_g is the granule dynamic yield stress. It will vary with formulation properties and granule porosity. Y_g and ρ_g should be measured at the porosity reached by the granules in the granulator ($\epsilon_{\min}$).

De is a measure of the ratio of impact kinetic energy to plastic energy absorbed per unit strain. It takes into account both the process agitation intensity and the granule rheology and indicates the amount of deformation during a typical impact.

In summary, the position of a system on a plot of *De* vs. $s_{\max}$ will characterize the type of growth behavior observed:

- At very low liquid contents (low $s_{\max}$), particles will either remain as a dry, free-flowing powder, or else will form nuclei due to van de Waals interactions, but not grow any further.
- At slightly higher liquid contents, either nuclei will form which will not grow or, if the system is extremely weak (high *De*), it will form a nongranular "crumb" material (Capes and Danckwerts, 1965).
- At medium levels of liquid content, if the granules are deformable and consolidate quickly (medium *De*), then they will grow steadily. If they are low-deformation, slowly-consolidating systems (low *De*), then they will exhibit induction-time behavior.
- At high liquid contents ($s_{\max} \rightarrow 1$), both fast and slow consolidating systems will grow rapidly.
- At very high liquid contents ($s_{\max} > 1$), a slurry or over-wetted mass will be formed.

A qualitative map of these growth regimes is given in Figure 3. Figure 4 illustrates the expected effects of different variables on the position of a formulation on the growth regime map. These effects are now discussed.

Effect of process agitation intensity

Increasing process agitation intensity will increase the typical impact velocity and, hence, increase the deformation number. The consolidation models of Ouchiyama and Tanaka (1980) and Ennis et al. (1991) both predict that granule consolidation will increase with increasing frequency and energy of impacts. This has been confirmed experimentally in tumbling pans, high-shear mixers, and agitated fluidized beds where it was found that increasing pan or impeller speed increased the initial rate of consolidation (Ouchiyama and Tanaka, 1980; Ritala et al., 1986; Watano et al., 1995). Likewise, the porosities of granules made in high-shear mixers are generally much lower than those made in tumbling drums and pans (such as 20 to 40% measured by Kristensen et al. (1985) compared with 40 to 50% measured by Kapur and Fuerstenau (1969)). Hence, increasing process agitation intensity will increase the extent of granule consolidation and, hence, increase the maximum pore saturation.

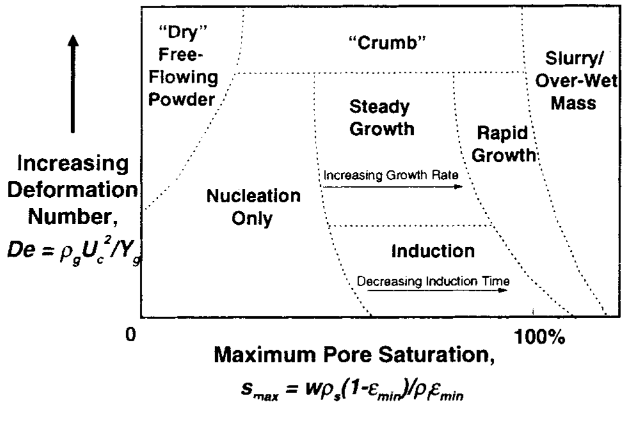

Figure 3. Proposed growth regime map showing the effect of granule consolidation rate and liquid content on granulation behavior.

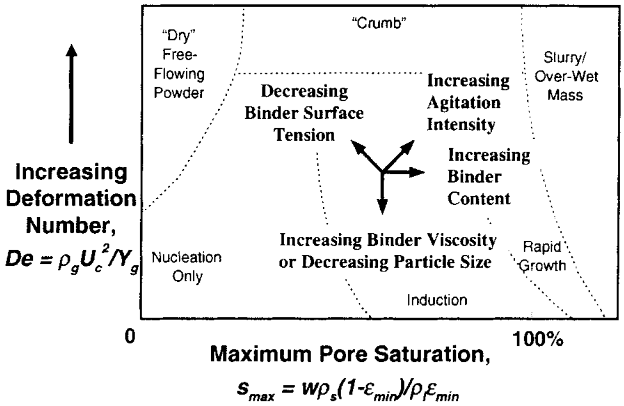

Figure 4. Effect of different variables on a formulation's position on the granulation regime map.

Therefore, increasing agitation intensity should shift a system's behavior towards the top-right of the proposed growth regime map, that is, from nucleation to steady or induction growth; from induction to steady or rapid growth; and from steady growth to crumb, rapid, or over-wetted growth (see Figure 4). There is some evidence for this in the literature. Poskart (1988) used a tumbling drum mounted on a spinning arm and found that increasing the centrifugal force from 1 to 10–50 G significantly increased the product density. At 50 G, destructive forces dominated and very few pellets were formed ("crumb" region). The process also became very sensitive to binder content and could easily "sludge"(that is, form an over-wetted mass or slurry). Similarly, Keningley et al. (1997) found that increasing impeller speed shifted a material's behavior from the steady growth to the crumb region (their so-called "paste" region). However, no work is currently available to confirm whether increasing agitation intensity will also shift a material's behavior from induction to steady growth as predicted by this regime map.

Effect of liquid viscosity and particle size

Increasing liquid viscosity or decreasing particle size both increase granule dynamic yield stress, but have variable effects on minimum porosity (Iveson et al., 1996; Iveson and Litster, 1998a,b). Hence, increasing binder viscosity and decreasing particle size will decrease *De* and should shift a formulation's behavior towards the bottom of the regime map, that is, from crumb to steady growth, and from steady growth to induction time behavior (Figure 4).

Results in the literature support these predictions. Capes and Danckwerts (1965) found that decreasing particle size shifted behavior from the crumb into the steady growth region. The increasing frequency of induction time behavior with decreasing particle size can be seen by surveying the existing literature. Workers who studied coarse, narrowly-sized silica sands did not observe induction behavior (Capes and Danckwerts, 1965; Newitt and Conway-Jones, 1958). Linkson et al. (1973) saw induction times of up to 200 revolutions with widely-sized, 43 μm-silica sands. Induction times of over 1,000 revolutions have been frequently observed with finely ground limestone powders (0.71 m^2/g) and hematite ores (0.295 m^2/g) (Kapur, 1978; Sastry et al., 1977).

Keningley et al. (1997) found that increasing liquid viscosity shifted a material's behavior from the crumb region to the steady growth region. However, there is no work available to confirm whether increasing binder viscosity can also shift a material's behavior from steady to induction growth. Hence, as part of this work, drum granulation experiments were undertaken to test whether increasing binder viscosity does increase the frequency of induction time behavior (see the subsection on granule growth experiments).

Effect of liquid surface tension

Decreasing binder surface tension has been found to decrease both dynamic yield stress and the extent of granule consolidation (Iveson and Litster, 1998a,b). Hence, lowering binder surface tension will increase De and decrease $s_{\max}$. This will shift a formulation's behavior towards the top-left of the regime map, that is, from induction to steady growth, and from steady growth to crumb or nucleation behavior. Capes and Danckwerts (1965) found that lowering binder surface tension did shift a material into the crumb region. However, no studies have been done to test whether lowering binder surface tension can shift a material's behavior from induction to steady growth.

Other variables

A range of other possibly significant variables, such as particle shape and roughness, have not been discussed. However, a feature of this regime map is that the effects of these variables should be implicitly accounted for by their effect on the granule deformation number and maximum pore saturation. However, as it is extremely difficult to isolate the effect of these variables on granulation behavior, it is currently impossible to verify this assertion.

Experimental Methods and Results

Two sets of experiments were performed to verify the proposed growth regime map: measurements of granule deformation during single impacts and measurements of granule growth behavior in a tumbling drum. The procedures and results of these experiments are outlined below. For a fuller discussion, see the work of Iveson (1997).

Materials

Glass ballotini were chosen as the model material since this is a well defined system that avoids complications due to solids dissolution. Three different grades of glass ballotini were used with surface mean sizes of 19, 31 and 60 μm purchased from Potters Industries (Lot 4 Boundary Rd., Laverton, Victoria, 3028, Australia) and had a density of 2.45 ±0.01 g/mL (measured by both water-displacement and helium pycnometry). Full details of these ballotini are given in Table 1.

Table 1. Properties of Glass Ballotini

Nominal Size	$x_{3,2}$* (μm)	$x_{4,3}$* (μm)	$\sigma_{4,3}$* (μm)	$\sigma_{4,3}/x_{4,3}$*	Dry-Tapped Porosity**	Wet-Tapped Porosity**
90–150 μm	60	122	48	0.39	0.385 ±0.004	0.373 ±0.005
44–90 μm	31	68	20	0.29	0.380 ±0.004	0.367 ±0.004
< 53 μm	19	36	15	0.42	0.51 ±0.01	0.382 ±0.005

*Surface mean size ($x_{3,2}$), mass mean size ($x_{4,3}$), and mass-mean standard deviation ($\sigma_{4,3}$) measured by Malvern Laser Mastersizer/E.

**Bulk porosities measured by tapping sample in a 30-mm-dia. volumetric cylinder.

It was desirable to cover a wide a range of binder surface tension and viscosity. Binder surface tension was varied between 31 and 72 mN/m by using solutions of the surfactant sodium dodecylbenzene sulphonate (NDBS). Binder viscosity was varied between 0.001 and 1 Pa·s by using solutions of glycerol. The full details of these solutions are given in Table 2.

Impact experiments

Cylindrical liquid-bound pellets of 41% porosity were made in a 20 mm diameter press. These pellets were dropped from various heights onto a stainless steel plate. The increase in impact area was measured and used to calculate the dynamic yield stress of the granules (Eq. 5). This was used to characterize the effect of varying particle and binder properties (Iveson and Litster, 1998a).

Granule growth experiments

Three different grades of glass ballotini (surface mean sizes 19, 31, and 60 μm) were granulated with varying amounts of water or glycerol (0.42 to 0.54 mL liquid per mL solid). Approximately 1 kg of ballotini was hand mixed with a known amount of liquid in a plastic bag. This feed was then pushed through a 2-mm sieve into a 30-cm in 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

Table 2. Properties of Liquid Binders

Liquid	Density* (g/mL)	Viscosity** (Pa·s)	Surface Tension (mN·m)
Water	0.997	0.0011	72†
Glycerol	1.256	1.1	63‡
0.00094 M NDBS Solution	0.997	0.0011	37†

*Measured in 25 mL specific gravity bottle.

**Measured in Contraves Rheomat 115 rheometer. Shear rate was varied between 20 and 150 s^{-1} for glycerol and between 300 and 3,650 s^{-1} for water.

†Measured by the Wilhemy Plate method using 4-cm^2 squares of filter paper.

‡From Weast (1981–1982).

in tumbling the mass, and a flexible scraper bar to prevent excessive material buildup on the drum walls.

The drum was rotated at 30 rpm for up to 4,000 revolutions (over 2 h). Samples of 50 to 100 g were taken periodically from the top of the tumbling mass. While these samples were not representative (in fact they were biased towards the smaller granules), they were sufficiently accurate to detect the gross differences between steady growth and induction time behavior. The samples were frozen in liquid nitrogen to increase their strength, and then sieved to measure the mass-median size of the sample. In some experiments, the entire contents of the drum were frozen and sieved and the experiment started again from scratch.

Results

The results of some of the impact deformation experiments are shown in Figure 5. Increasing binder viscosity and binder surface tension always increased the dynamic yield stress (Y). Decreasing particle size increased Y when water was used, but had no significant effect when glycerol was used. The water-based binders all displayed a maximum in Y with increasing binder content, whereas when glycerol was used, Y steadily increased in the range of conditions covered (Iveson and Litster, 1998a).

The results of the drum granulation experiments are shown in Figures 6a–6f. The water-bound systems (Figures 6a, 6c and 6e) all exhibited steady-growth behavior. Granule size increased steadily with time and increasing binder content increased the rate of granule growth. At low binder contents, very little growth took place (cf. Figure 2). The 60 μm glass ballotini also exhibited steady-growth behavior when granulated with glycerol (Figure 6b).

However, when granulated with glycerol, the 31-μm ballotini (Figure 6d) had one example of induction-time behavior and the 19-μm ballotini (Figure 6f) showed several examples of induction-time behavior (cf. Figure 2). At low binder contents, granules remained as nuclei for the length of the experiment, and at high binder contents, granules grew rapidly from the start (zero induction time). Increasing binder content generally decreased the induction time, although there is significant variation in the induction time of duplicate experiments (probably due to variations in the initial consolidation of the hand-mixed feed).

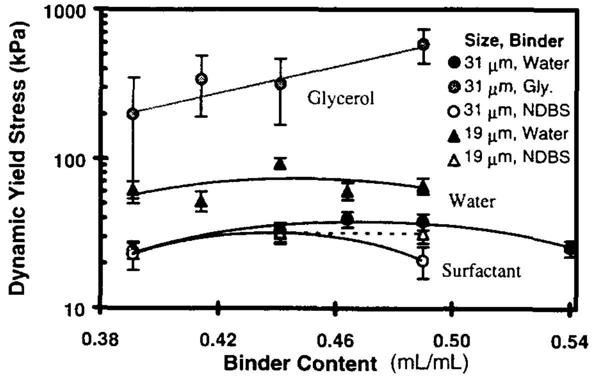

Figure 5. Dynamic yield stress vs. binder content for pellets made from a range of particle sizes using water, glycerol, and surfactant (NDBS) solutions.

Courtesy of Iveson and Litster (1998a).

These results qualitatively support the proposed growth regime map which predicts that the frequency of induction time behavior should increase as the granule yield stress increases (that is, as particle size is decreased and binder viscosity increased). This is the first time that this effect of binder viscosity has been reported in the literature.

Preliminary Verification of Proposed Regime Map

The drum granulation results in Figure 6 were used to test the regime map. The growth rates demarcating the different granulation regimes were arbitrarily defined according to Table 3. The dynamic yield stress of the granules was taken to be the value measured by Iveson and Litster (1998a) for pellets of 41% porosity. The characteristic impact velocity was taken to be ωD. The minimum granule porosity reached at each set of conditions was not measured. Instead, it was assumed that the glass ballotini consolidated to its minimum wet-tapped porosity. This assumption ignores the influence of binder viscosity and binder content on minimum granule porosity (cf. Iveson et al., 1996). The granule density was calculated at this wet-tapped porosity.

The results plotted in Figure 7 show good agreement with the proposed regime map (Figure 3). All cases of rapid growth occur on the righthand side of the plot ($s > 0.78$). Steady growth occurs only in the upper middle of the plot ($De > 0.02$) and induction behavior occurs in the lower middle of the plot. Several cases of nucleation occur in the induction region, but this is not surprising since induction behavior will always appear to be nucleation if the experiment is stopped before the induction time is reached.

It is difficult to use data in the literature to directly test this regime map, because few workers report the yield stress of their formulations or the minimum porosity reached. However, we can make use of some data with some simplifying assumptions. We assume that the dynamic yield stress is proportional to granule tensile strength (σ_t), as predicted by the theory of Rumpf (1962). This assumption can only be made with any degree of accuracy in systems where frictional and dynamic effects are small (since these are ignored in Rumpf's model). Hence, only data for coarse particles and low viscosity binders can be used.

The drum granulation data of Newitt and Conway-Jones (1958), Capes and Danckwerts (1965), and Linkson et al. (1973) was tested. They all granulated relatively coarse silica sands with water and ethanol binders in drums of similar size and speed. Again, the minimum wet-tapped porosity was used to calculate the maximum saturation and was also used in calculating the theoretical tensile strength. Newitt and Conway-Jones were the only ones to report wet-tapped porosity, so their data was used to estimate the wet-tapped porosities of the sands used by Capes and Danckwerts and Linkson et al.. Newitt and Conway-Jones were also the only ones to report the surface mean sizes of their sands. In the other two cases arithmetic mean sieve size was used in place of the surface-mean size in Rumpf's theory. For widely-sized sands, this would lead to an underprediction of granule strength, so only data for narrowly-sized sands were used. A full description of

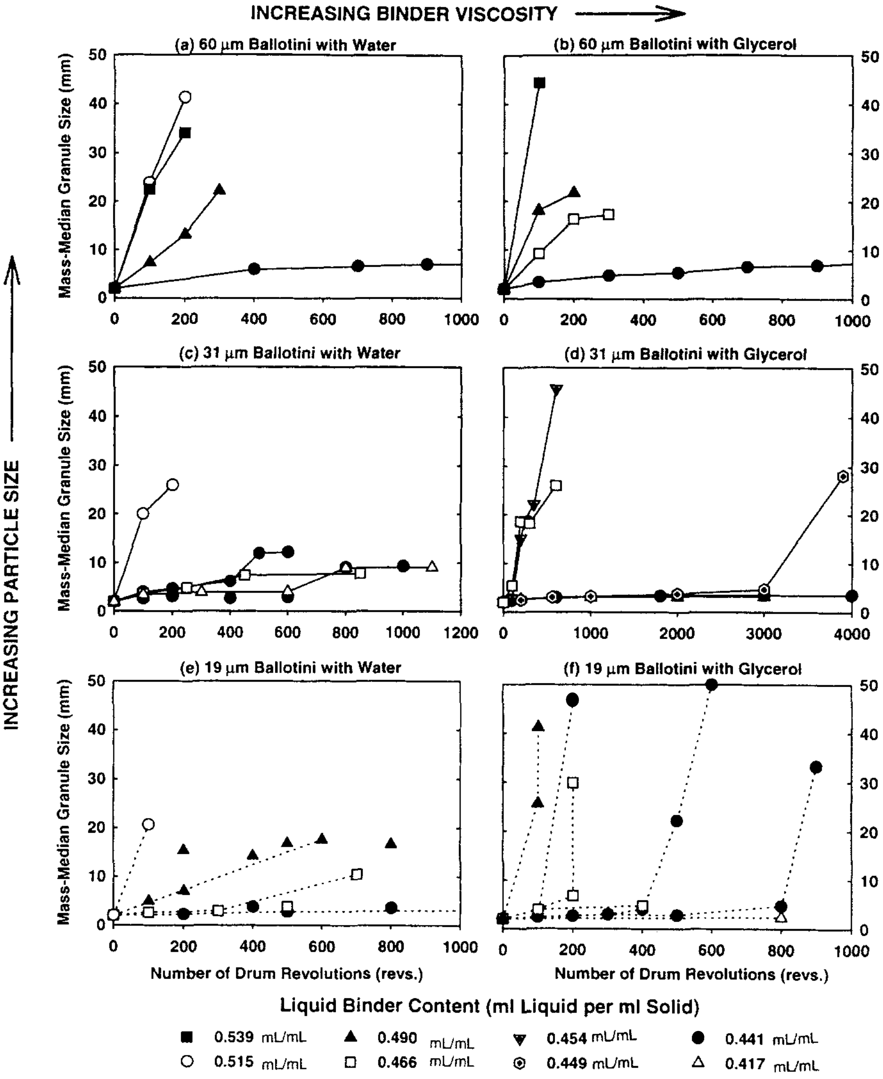

Figure 6. Mass-median size vs. granulation time for 19-, 31- and 60-μm ballotini granulated with water and glycerol binders across a range of liquid contents.

Points connected by solid lines represent multiple samples taken from the same experiment. Points connected by dotted lines represent experiments where the entire contents of the drum were sampled and the experiment restarted from scratch (see subsection on granule growth experiments).

the estimates and assumptions used in analyzing the data from these three articles is given in Iveson (1997).

Figure 8 plots the results of this analysis. The group $\rho_g U^2/\sigma_t$ is used instead of the deformation number. Given the many assumptions and approximations used, the results are encouraging. The crumb results all lie at the top of the plot ($\rho U^2/\sigma_t > 0.2$), most of the steady-growth results lie in the middle region, most of the rapid growth results lie on the righthand side, and the majority of the nucleation results are on the lefthand side ($s < 0.92$).

Table 3. Definition of Granule Growth Regimes

Growth Regime	Growth Rate (mm/1,000 Revs)
Nucleation	< 1
Slow growth	1 to 5
Steady growth	5 to 25
Rapid growth	> 25
Induction	< 5 followed by rapid growth

The ordinates of Figure 7 and Figure 8 merge together well. The crumb region occurs when $De > 0.2$, steady growth occurs for $0.2 > De > 0.02$, and induction growth occurs for $De < 0.02$. However, the abscissas do not align together. Figure 7 indicates that steady growth occurs in the region of $0.7 < s_{\max} < 0.8$, whereas Figure 8 shows steady growth as occurring for $0.9 < s_{\max} < 1.1$. This difference reflects the difficulty in accurately estimating a formulation's maximum pore saturation from the limited data available in the references analyzed.

These preliminary verifications of the proposed regime map are very encouraging. However, further work is required to extend this validation and to further quantify the boundaries between the different growth regimes. In particular, work is required testing a given material across a range of agitation

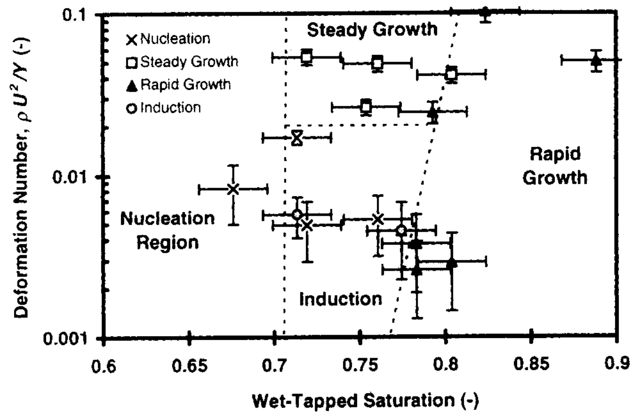

Figure 7. Drum granulation results of Figure 6 plotted on regime map using wet-tapped porosity to calculate liquid saturation.

intensities in order to measure the effects of scale-up and process agitation intensity.

Implications of Regime Map

If correct, this regime map has the potential to significantly improve our ability to design, scale up, and operate granulation devices. The control strategies for different formulations will depend on which area of the regime map they lie in.

If a powder-binder mixture lies in the induction growth region (strong, nondeformable granules), then, provided its induction time is not exceeded, it will never grow rapidly. The granule size distribution will be controlled by the nucleation conditions. Layering growth could be promoted by spraying binder onto the surface of pre-existing granules, whereas smaller granules could be formed by spraying the binder onto the fresh feed to promote nucleation. Therefore, the product size distribution will be strongly influenced by the positioning of the liquid nozzle(s) and powder feed, liquid droplet-size distribution, powder wetting characteristics, and the flow and mixing of material in the granulator.

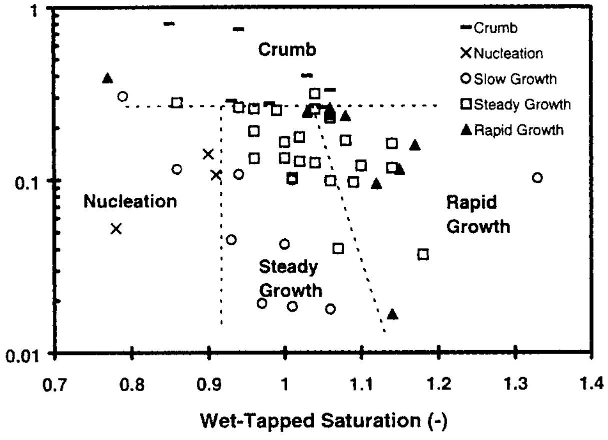

Figure 8. Drum granulation data of Newitt and Conway-Jones (1958), Capes and Danckwerts (1965), and Linkson et al. (1973).

Plotted on regime map using theoretical tensile strength according to Rumpf (1962) to calculate the deformation number and wet-tapped porosity to calculate liquid saturation.

This kind of formulation will be relatively easy to scale with respect to granule size distribution since there is a long period of little growth. However, care must be taken that the process agitation intensity does not increase significantly during scale-up (such as from a small to large pan). Such an increase may shift the formulation's behavior from the induction region to the steady growth region, in which case scale-up will be much more difficult.

The growth behavior of powder-binder mixtures in the steady growth region (weak, easily deformable granules) is dominated by coalescence, not layering or nucleation. The rate of coalescence is very sensitive to binder content and scale of operation, because these variables both affect the granule deformability. This type of formulation is difficult to scale up with respect to granule size distribution. Granule size is controlled by the rate of liquid addition and is also a strong function of the residence time. On the other hand, porosity will be insensitive to residence time since granules will quickly reach their minimum porosity.

This regime map is the first of its kind. It implicitly or explicitly includes all of the parameters of interest. It has the potential to help us customize formulations to give different granulation behavior and product characteristics *without the need to perform any granulation experiments*. For instance, as demonstrated above, an increase in binder viscosity can be used to shift a formulation from the steady growth to induction regime. This makes control of granule growth much easier and also slows down the rate of consolidation to give a less-porous product.

However, this regime map does have some limitations. First, it does not consider wetting and nucleation effects. This is a function of droplet size, spray position, and powder wetting characteristics. These variables will control the size distribution and porosity of the initial nuclei.

Secondly, this regime map does not predict the rate or extent of granule growth. These will be a function of the granule strength, process intensity, and process residence time. This more detailed prediction will require the development of a coalescence model, which accounts for granule consolidation, deformation and the effects of agitation intensity.

Conclusions and Recommendations

It has been shown that granule consolidation and deformation behavior can be used to predict granule growth behavior. Highly deformable, quickly consolidating systems grow steadily with time. Low deformation, slow consolidating systems tend to exhibit induction behavior. For the first time, it has been shown that increasing binder viscosity can shift a material's behavior from steady growth to induction time behavior.

A regime map of granulation behavior in terms of the granule deformation number and maximum pore saturation has been proposed. These two variables are bulk parameters of the powder-binder mixture and also functions of the process agitation intensity. Hence, they implicitly take into account all the variables affecting granule growth. This regime map qualitatively explains the variations in granulation behavior seen in the literature. Preliminary validation of this regime map with present results and data in the literature is encouraging. However, more work is required to confirm its validity, quantify the boundaries between different growth regimes, and investigate the effect of process agitation intensity.

This regime map has the potential to greatly aid in designing and controlling granulation systems, because it predicts the granule growth behavior of a formulation without the need for extensive laboratory and pilot-scale granulation experiments.

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Since the submission of the original manuscript of this article, Tardos et al. (1997) defined a Stokes deformation number (St_{def}) which is very similar to our deformation number (De). They showed that this number determined the deformation and breakup of wet granules in a uniform shear field of fluidized powder and used this to predict that the maximum size granules would reach in a granulator.

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